

Deformation and Fracture of Soft Polymers: Experiments, Modeling and Computations

By

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INTRODUCTION

Background

Soft polymers are a broad class of polymers with chain networks formed by permanent or dynamic chemical or mechanical crosslinks, leading to elastic moduli in the range of a few kilopascals to tens of megapascals. This category encompasses elastomers, viscoelastic and elastic-viscoplastic polymers, polymer gels, and biological tissues. Their physical forms can vary from very viscous, slowly flowing soft solids (Konale et al., 2023) to elastomeric rubber-like materials (Steck et al., 2019).

These materials are essential in engineering due to a wide array of applications, including targeted drug delivery (Huebsch et al., 2014; Rodell et al., 2016; Balakrishnan et al., 2019; Xin, 2022), biological and structural adhesives (Yuk et al., 2016; Hong et al., 2018; Yuk et al., 2019; Liu et al., 2020a; Deng et al., 2021), mechanical sensing and actuation (Yuk et al., 2017; Liang et al., 2020; Liu et al., 2021; Zhang et al., 2021; Li et al., 2021) and flexible impact mitigation (Wang et al., 2016; Liu et al., 2020b; Sang et al., 2021; Zong et al., 2021; Tang et al., 2022). Significant variations in their mechanical properties are driven by their underlying crosslinking mechanisms. Permanent crosslinks yield rate-independent behavior with high elasticity, as in elastomers (Nunes, 2011). On the other hand, dynamic reversible crosslinks lead to significantly rate-dependent mechanical behavior (Kurkin et al., 2021). In the case of dynamically crosslinked polymers, the material response is dictated by the loading timescale: longer timescales lead to highly viscous, fluid-like

behavior, while shorter timescales result in elastomer-like behavior. Additionally, these dynamic crosslinks can facilitate the autonomous self-healing of micro-fractures (Lai et al., 2019). Hence, depending on the nature of crosslinks in a soft polymer, its behavior can vary between the extremes of purely elastic and highly viscous response.

Polyborosiloxane (PBS), a key rate-stiffening soft polymer, has dynamic boron (B)-oxygen (O) coordinate-bond crosslinks and exhibits significantly different mechanical behavior at different timescales (Li et al., 2014). PBS has been used to develop self-healing sensing and actuation interfaces (Narumi et al., 2019) and has been shown to mitigate impact loads (Zhang et al., 2020). Despite PBS’s broad applications, its large strain material response characterization and predictive model development are lacking.

Although the mechanics community has made significant strides in the experimentation and modeling of elastomers, rate-dependent soft polymers continue to present substantial challenges due to their complex material responses. Currently, there is a lack of deformation, damage, and fracture models capable of predicting the strongly rate-dependent, non-linear behavior of a broad class of soft polymers. As soft polymers become increasingly integral to both daily and industrial-scale applications, comprehensive, microstructure physics-motivated models and simulation capabilities for end-to-end design alongside extensive experimental data are necessary from both scientific and practical perspectives.

To address these critical gaps, the objective of this dissertation is threefold: (i) to experimentally characterize the deformation and fracture response of PBS, a representative and important rate-stiffening soft polymer; (ii) to develop microstructural physics-motivated deformation and damage constitutive models for PBS that are broadly applicable to other soft polymers; and (iii) to implement these models numerically using the traditional finite element method (FEM) and emerging machine learning (ML) based approaches for validation and predictive simulations.

Publications related to this dissertation

The following is a list of publications in peer-reviewed journals with content related to this dissertation.

Konale, A., and Srivastava, V., A physics-informed neural network for modeling fracture without gradient-damage: Formulation, application, and assessment, *Journal of the Mechanics and Physics of Solids*, 106395, 2026.

Konale, A., and Srivastava, V., On modeling fracture of soft polymers, *Mechanics of Materials*, 105346, 2025.

Konale, A., Ahmed, Z., Wanchoo, P., and Srivastava, V., A large deformation model for quasi-static to high strain rate response of a rate-stiffening soft polymer, *International Journal of Plasticity*, 168, 103701, 2023.

Manuscripts in preparation with content related to this dissertation are also listed below.

Konale, A., and Srivastava, V., A physics-informed neural network for modeling large deformation contact between elastomers and rigid bodies (*in preparation*).

Konale, A., and Srivastava, V., Double network composites using a rate-stiffening soft polymer: experimental response and simplified model-based insights (*in preparation*).

Konale, A., Gupta, A., Ma, K., Bazilevs, Y., Guduru, P., and Srivastava, V., Modeling damage and fracture in polymeric architected materials and structures (*in preparation*).

In fact, much of the text that comprises this dissertation is drawn from the above publications and manuscripts in preparation.

Dissertation outline

This dissertation compiles findings and discussions from several distinct projects, presenting experimental data alongside modeling and simulation techniques for a range of pressing problems relevant to the main topic of soft polymer mechanics. The work is organized into seven chapters, structured as follows:

In *Chapter 1*, we present a microstructural physics-motivated constitutive model for PBS and similar soft polymers and polymer gels with dynamic crosslinks to predict their large strain, non-linear loading-unloading, and significantly rate-dependent responses. Large strain (up to true strains of $\sim 125\%$) compression stress-strain data of PBS over a wide strain rate range of 10^{-3} s^{-1} to 10^4 s^{-1} is also reported. The semiphysical model reasonably accurately captures the response of PBS over six decades of strain rates. In *Chapter 2*, we propose and demonstrate that a critical value of stress work can serve as a measure of fracture resistance for a certain class of soft polymers. We also present a damage model to predict fracture in soft polymers. In the model, the energetic component of the critical stress work is proposed as a damage initiation criterion capable of capturing failure surfaces. Damage growth is modeled through a generalized gradient-damage framework. The fracture model is validated for several distinct soft polymers and can predict key physical phenomena. In *Chapter 3*, we present an alternative numerical method using a physics-informed neural network (PINN) to model fracture in elastomers undergoing large deformation, without the complex gradient-damage formulation. The PINN implementation here requires no training data and utilizes the collocation method to formulate physics-informed loss functions. The performance of the PINN has been evaluated by systematically varying key neural network parameters, providing an assessment and guidance for future applications.

In *Chapter 4*, we continue the theme of utilizing PINNs as an alternative numerical method for soft polymer mechanics. Specifically, we develop a PINN for modeling large

deformation frictionless contact between elastomers and rigid bodies. Various automatic differentiation (AD)-based approaches are presented and assessed for evaluating the tangential vector on the deformed body's surface necessary to enforce the frictionless conditions. The PINN's predictive capabilities have been validated for various contact surface configurations using baseline solutions from FEM. A preliminary extension of the formulation to elastomeric deformable-deformable body frictionless contact is also discussed. In *Chapter 5*, with the initial aim of addressing PBS's lack of shape stability for practical applications, we present the large strain material response data for PBS-polydimethylsiloxane (PDMS) interpenetrating double network (IDN) polymer composites. A basic model is shown to reasonably capture the IDN response and can explain the desirable, peculiar properties of a specific IDN composition. Results from the exploration of IDNs filled with conductive particles for flexible strain and strain rate sensing are also presented. In *Chapter 6*, we map the damage and fracture modeling approach for soft polymers from *Chapter 2* for polymeric architected materials. A damage initiation criterion is proposed for amorphous polymers. Experimental validation of the developed damage constitutive model and numerical simulation capability is also presented. Lastly, *Chapter 7* presents the closing remarks and highlights the future perspectives. In *Appendix A.1*, pseudo codes for the fracture model (in *Chapters 2* and *6*) implementation in the commercial FE software ABAQUS are presented.

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CHAPTER 1

A large deformation model for quasi-static to high strain rate response of a rate-stiffening soft polymer

1.1 Introduction

Soft polymers and polymer gels with dynamic, reversible crosslinks that exhibit rate-dependent mechanical behavior are an important class of materials with a wide range of applications. In this chapter, we only consider and refer to polymer gels [solvent-swelled polymer networks (Chester and Anand, 2010; Shukla et al., 2020; Tepole and Kuhl, 2016; Bosnjak et al., 2020)] that have negligible solvent diffusion in the mechanical loading timescales of interest, i.e., the concentration of the solvents is approximately constant both spatially and temporally. As an example, the diffusion of solvent in a polymer gel can be neglected to analyze its mechanical response during a high rate loading event.

Polyborosiloxane (PBS), a key rate-stiffening soft polymer has dynamic boron (B)-

oxygen (O) coordinate-bond crosslinks and exhibits significantly different mechanical behavior at different timescales (Li et al., 2014; Wang et al., 2014, 2016b; Tang et al., 2016; Wang et al., 2018; Zhao et al., 2021; Liu et al., 2019a; Zhao et al., 2020; Kurkin et al., 2021; Liu et al., 2022; Kim et al., 2022). PBS has been used to develop self-healing sensing and actuation interfaces (Narumi et al., 2019). It has been shown to mitigate impact loads (Zhang et al., 2020; Zhao et al., 2020; Kurkin et al., 2021; Kim et al., 2022). Lightweight and flexible impact-mitigating composite structures where PBS is integrated into polymer foams (Tu et al., 2023) or sandwich structures (Myronidis et al., 2022) have been shown to protect against impact. Material response characterization studies on PBS have been limited to small strain loadings, primarily oscillatory shear using a rheometer to quantify the effects of loading rates (Tang et al., 2016; Wang et al., 2016a; Kurkin et al., 2021; Zong et al., 2021; Myronidis et al., 2022; Zhao et al., 2021; Zhang et al., 2020; Wang et al., 2014, 2018). Large strain material response characterization studies have been conducted only on PBS with fillers to enhance its shape stability (Jiang et al., 2014; Zhao et al., 2021) or to provide sensitivity to magnetic fields (Wang et al., 2014, 2016b, 2018; Liu et al., 2019a). It is important to study and model the response of pure PBS without any fillers to understand its physical material behavior.

Despite the broad applications of PBS, its large strain material response has not been characterized over a wide range of strain rates. There is no large deformation constitutive model which can predict the response of PBS spanning the quasi-static (10^{-3} s^{-1}) to high (10^3 s^{-1}) strain rate range. The time dependent behavior of pure PBS is shown through our experimental observations considering two distinct timescales: (i) PBS flows under its weight over hours as shown in Figure 1.1(A) where a PBS sphere flattens in an hour and has negligible shape recovery, and (ii) when subjected to a high loading rate, PBS exhibits notable shape recovery after undergoing significant deformation as seen in Figure 1.1(B) where PBS is subjected to a strain rate of approximately 5000 s^{-1} using a Split Hopkinson Pressure Bar (SHPB). Figure 1.1(B) shows the PBS deformation over milliseconds.¹

¹PBS lacks shape stability as shown in Figure 1.1(A) for direct practical applications. Section 1.2.5

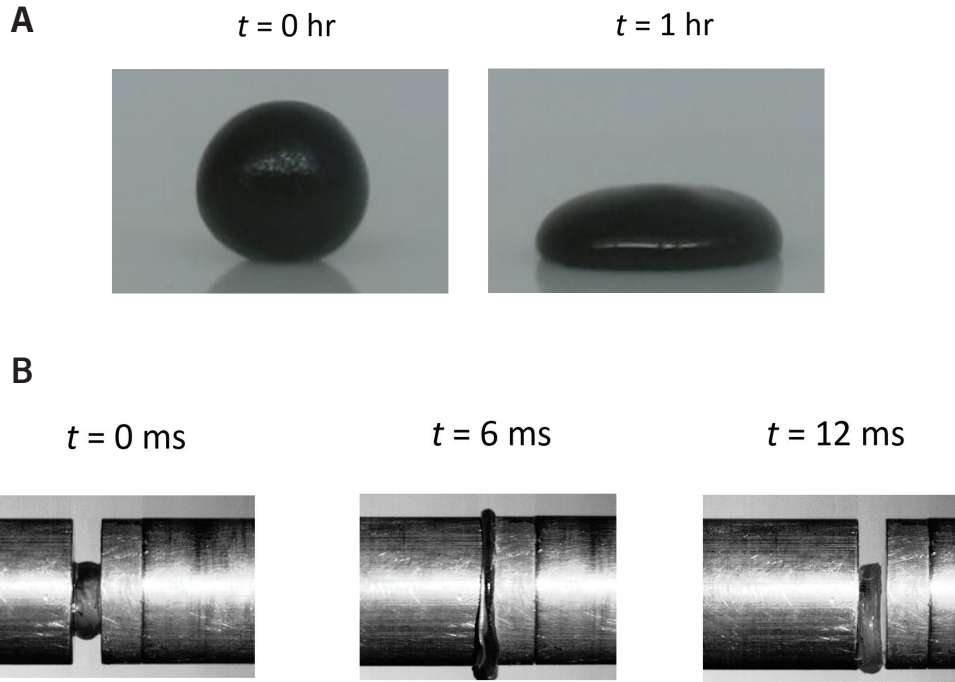


Figure 1.1: **Time dependent behavior of PBS: two distinct timescales.** (A) Flow of a PBS sphere under its weight in 1 hour. The images are shown at $t = 0$ and 1 hour. The PBS sphere was colored for easy visualization. (B) Response of PBS subjected to a strain rate loading of approximately 5000 s^{-1} using a Split Hopkinson Pressure Bar. The PBS deformation images are shown at $t = 0, 6$, and 12 milliseconds (ms). PBS experiences large deformation followed by significant shape recovery at high rates.

Important contributions in three-dimensional, large deformation modeling of soft polymers and polymer gels with dynamic crosslinks (Polyvinyl alcohol, Polyampholyte, Ethyl acrylate copolymers, Poly(acrylamide-co-acrylic acid) hydrogel) with different microstructures and micromechanisms are limited to the quasi-static (10^{-3} s^{-1}) to slow (10^{-1} s^{-1}) strain rate range (Hui and Long, 2012; Long et al., 2014; Guo et al., 2016; Meng and Terentjev, 2016; Meng et al., 2016; Lin et al., 2020; Pamulaparathi Venkata et al., 2021; Wang et al., 2023b). The numerical implementations of these models have also been limited to *one-dimensional, homogeneous* deformation problems. Additional micromechanisms with very short relaxation timescales can contribute to the mechanical

presents preliminary experimental results from a shape stability imparting approach utilizing a metal foam scaffold. A flexible counterpart with a soft polymer network as a scaffold is discussed in Chapter 5. The synthesis procedure, experimental characterization results, and basic modeling are presented along with results from a proof-of-concept study exploring strain and strain rate sensing through the incorporation of conductive filler particles.

response at high strain rates. Hence, large-strain material response characterization at high strain rates and corresponding constitutive models are crucial. In addition, numerical procedures to evaluate the convolution-like time integrals due to dynamic crosslink kinetics for model implementation in finite element programs are necessary to simulate complex inhomogeneous loading problems. The important gaps to be filled are (a) large-strain material response characterization of PBS and (b) physics-based, three-dimensional large deformation constitutive models and their numerical implementation spanning a wide strain-rate range for PBS.

We have conducted the following work: (i) experimentally characterized large strain compression response of PBS up to a true strain of $\sim 125\%$ over a wide strain rate range of 10^{-3} s^{-1} to 10^3 s^{-1} , (ii) developed a microstructure physics-motivated three-dimensional, large-deformation, rate-dependent, elastic-viscoplastic constitutive model for PBS and similar soft polymers and polymer gels with dynamic crosslinks. The mathematically straightforward model accounts for the dynamic crosslink kinetics and the dominant mechanisms of deformation in PBS, allowing for a smooth transition from dissipative resistance due to chain sliding dominating the mechanical response at low strain rates to the primary contribution from the network stretching resistance at high strain rates. The model captures PBS's response over the strain rates of 10^{-3} s^{-1} to 10^3 s^{-1} . The model is validated through inhomogeneous deformation experiments, (iii) propose two types of crosslinks in PBS and identify their significantly different macroscopic relaxation timescales (τ) based on the macroscopic mechanical response, namely B:O coordinate-bond dynamic crosslinks ($\tau \approx 3 \text{ s}$), and temporary entanglement lockups at high strain rates acting as crosslinks ($\tau \approx 0.001 \text{ s}$), and (iv) outline a numerical procedure evaluating the convolution-like integrals due to crosslink kinetics for model implementation in finite element softwares.

1.2 Experiments

All experiments were conducted at 25°C. Three repetitions were performed for each experimental result reported in this chapter.

1.2.1 Synthesis

PBS was synthesized by mixing hydroxy-terminated Polydimethylsiloxane entangled melt (PDMS, Sigma Aldrich 481963, 750 cSt) and Boric Acid (BA, Sigma Aldrich B0394, ACS reagent, $\geq 99.5\%$) in the ratio 160:1 by weight, stirring the mixture and then heating it at 120°C for 72 hrs (Seetapan et al., 2013; Tang et al., 2016). Hydroxy-terminated PDMS entangled melt with a viscosity of 750 cSt has a number-average and weight-average molecular mass of 30 kg/mol and 49 kg/mol, respectively, which are greater than 12 kg/mol, the critical entanglement molecular weight of PDMS.

1.2.2 Rheological experiments

Oscillatory shear and shear stress relaxation experiments were performed on an ARES G2 Rheometer to 1% strain using a cone and plate geometry (40 mm diameter and 0.04 rad cone angle). Oscillatory shear experimental results for PBS are shown in Figure 1.2(A). A network with only ideal, permanent crosslinks would exhibit frequency-independent storage and loss moduli (Parada and Zhao, 2018). The highly frequency-dependent storage, loss moduli, and plateauing of the storage modulus at high frequencies in Figure 1.2(A) suggest the presence of dynamic crosslinks with relaxation timescales within those of the experiments. For comparison, oscillatory shear experiments were conducted on the hydroxy-terminated PDMS entangled melt precursor, the results of which are shown in Figure 1.2(B). The consistently higher loss modulus than the storage modulus for the PDMS entangled melt in the high frequency regime (10 - 100 rad/s) illustrates its viscous fluid-like behavior. Comparing this with the corresponding response of PBS in Figure

1.2(B) confirms the condensation of BA and PDMS hydroxy end groups during the heating process to connect PDMS chains along their ends through Silicon (Si)-O-B bonds (Tang et al., 2016). Figure 1.2(C) shows the shear stress relaxation response of PBS.

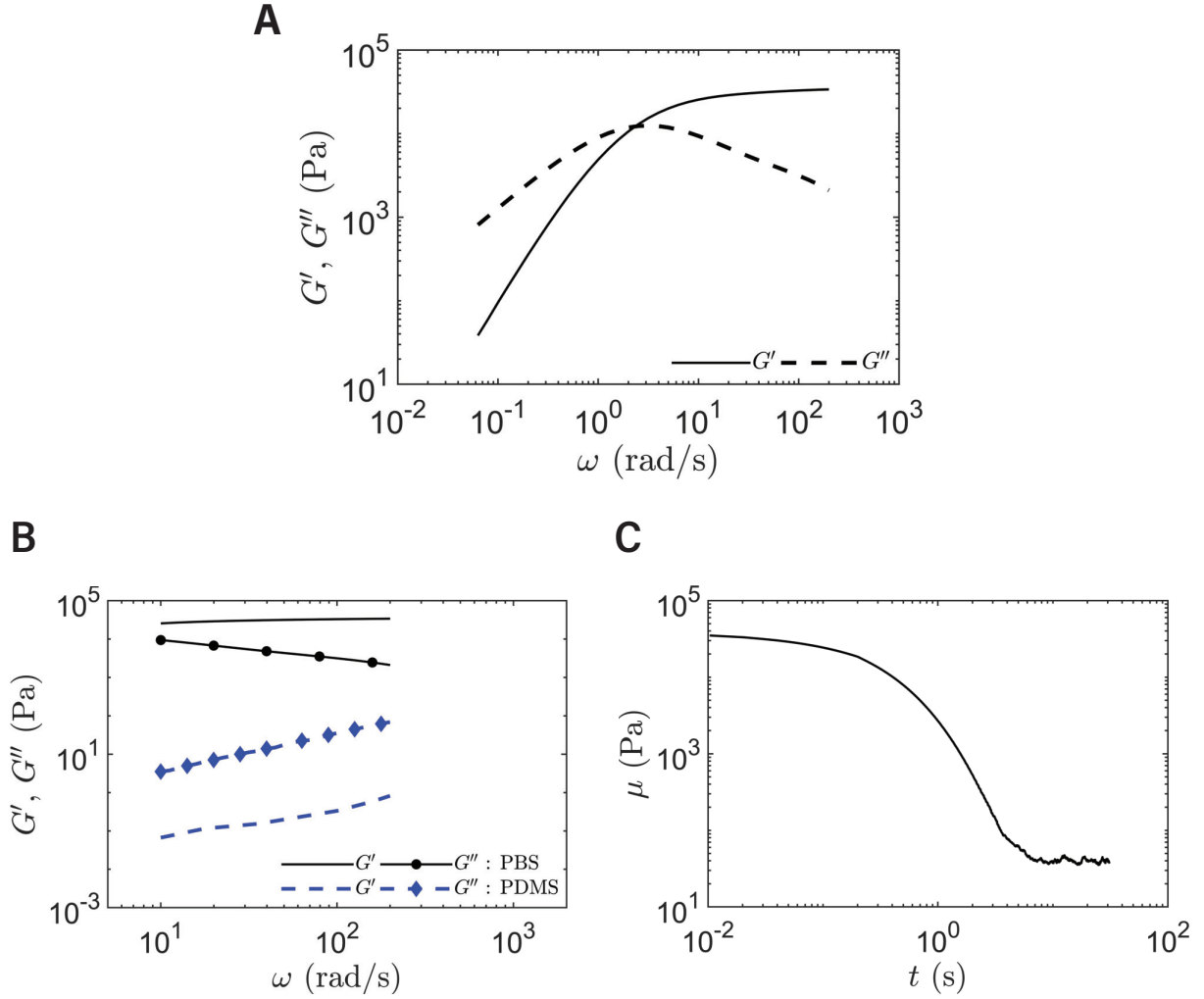


Figure 1.2: **Small strain rheological response of PBS: oscillatory shear and shear stress relaxation.** (A) Oscillatory shear response of PBS. (B) Comparison of PBS's oscillatory shear response along with its hydroxy-terminated PDMS precursor. (C) Shear stress relaxation response of PBS. The specimens were subjected to 1% strain in all rheological experiments.

1.2.3 Compression experiments

Quasi-static to moderate strain rates

Compression tests up to a true strain² of 125% were carried out using UniVert Mechanical Test System (CellScale) at true strain rates over three decades from 0.001 s^{-1} to 1 s^{-1} . The specimens were cylindrical with a diameter of $10 \pm 2 \text{ mm}$ and a height of $5 \pm 1 \text{ mm}$. The interfaces between the specimens and the platens were lubricated to prevent barreling. Figure 1.3(B) and (C) show the corresponding true stress-strain response and the substantial rate-dependent stiffening.³ The specimens showed significant permanent deformation after unloading ($\approx 100\%$ at 0.001 s^{-1} to $\approx 83\%$ at 1 s^{-1}) as shown by the unloading response curves.

High strain rate experiments with a SHPB setup

The SHPB setup (Kolsky, 1964) is used to characterize the dynamic, high strain rate response of materials and structures. The specimen is sandwiched between incident and transmission bars equipped with strain gauges that measure the respective strain histories (Srivastava et al., 2000, 2002; Khan and Lopez-Pamies, 2002; Khan and Farrokh, 2006; Kabirian et al., 2014; Ku et al., 2020). A stress pulse of finite length is generated in the incident bar by impacting it with a striker bar fired from a gas gun. Once the stress pulse arrives at the specimen, part of it gets transmitted through the specimen to the transmission bar, while the remaining pulse gets reflected into the incident bar due to impedance mismatch. Force history on both faces of the specimen and dynamic stress-strain curves can be obtained from the measured strain histories and standard analysis utilizing the theory of one-dimensional wave propagation (Chen and Song, 2010).

²True strains for the experiments in Section 1.2.3 were calculated using its standard definition and displacement data from the experiments.

³Section 1.2.4 presents a brief discussion on the effects of potential moisture exposure during practical applications on the mechanical response of PBS.

PBS was characterized in the high strain rate regime (10^3 s^{-1}) using a SHPB setup.

Due to the low stress wave impedance of PBS, a hollow aluminum transmission bar was used with inner and outer diameters of 16.51 mm and 19.05 mm, respectively. The hollow bar amplifies the transmitted stress signal compared to an equally long solid aluminum transmission bar with a diameter of 19.05 mm due to its reduced cross-sectional area. Each end of the transmission bar was plugged with aluminum end caps. Semiconductor strain gauges (Micron Instruments SS-060-033-1000PB-S1) were used on the hollow transmission bar. The solid incident bar was made of aluminum and had a diameter of 19.05 mm. Both incident and transmission bars were 1.83 m long. The specimen-incident bar and specimen-transmission bar interfaces were lubricated using a Molybdenum disulfide-based lubricant. To obtain a valid material response, it is essential to achieve dynamic force equilibrium throughout the PBS specimen. At strain rates in the order of 10^3 to 10^4 s^{-1} , the timescales of deformation are comparable to those of the stress wave propagation. The standard methods for evaluating material response in the form of stress-strain curves from these high strain rate experiments assume homogeneous conditions throughout the specimen thickness (Sharma et al., 2002; Weeks and Ravichandran, 2022; Prakash and Mehta, 2012), which are approximately obtained after a significant number of stress wave reverberations through the specimen thickness. Therefore, it is essential to ensure that the specimen experiences dynamic force equilibrium (reasonably equal forces on the front and back faces of the specimen from the incident and transmission bars, respectively) over a significant portion of the deformation period for the stress-strain curves to actually represent the material response. A careful consideration of dynamic force equilibrium in characterizing soft materials at high strain rates is warranted.

A significant number of stress wave reverberations through the specimen thickness are needed to achieve force equilibrium. Incident stress pulse duration generated using a SHPB is equal to the time taken for stress waves to make a round trip through the striker bar length (i.e., it is inversely proportional to the speed of stress waves in the striker bar)

after they originate at the striker bar-incident bar interface. In low impedance materials like PBS, the relatively lower speed of stress waves implies that attaining dynamic force equilibrium using conventional high impedance striker bars (short stress pulse durations) is very difficult (Sharma et al., 2002; Song and Chen, 2004). A low impedance striker bar made of HYDEX[®] (a type of nylon) with 0.3 m length and 12.7 mm diameter was used, which provided a sufficiently long incident stress pulse length allowing adequate time for stress to equilibrate across the specimen thickness. The cylindrical specimens had a diameter of 11.43 ± 1 mm and a thickness of 1.04 ± 0.1 mm. The corresponding true stress-strain response is shown in Figure 1.3(A), and the true strain rate reported is an arithmetic mean over the pulse length. The difference in stress magnitudes between the high strain-rate (4500 s^{-1} and 8500 s^{-1}) and moderate strain-rate (0.001 s^{-1} - 1 s^{-1}) response is seen in Figure 1.3(A). To get a quantitative sense, true stress-strain rate curves at different strain levels are shown in Figure 1.3(D). The stresses increase by slightly more than three orders of magnitude as we go from the strain rate of 0.001 s^{-1} to 8500 s^{-1} .

1.2.4 Effect of exposure to moisture in the air at 25° C on PBS

The presence of water dissociates the Si-O-B bonds in PBS, which are necessary for the dynamic crosslinking of the chains (Liu et al., 2023). The effect of moisture in the air at 25° C was studied. The ‘stretching using inserts’ setup discussed in the upcoming Chapter 2 (Section 2.8.4) was used. Figure 1.4 shows the extension force vs. displacement curves (20 mm/s insert separation speed) for 0 and 24 hours of exposure. The extension force decreased significantly upon exposure, which is consistent with the softening observed in (Liu et al., 2023). Therefore, strategies to prevent water contact with PBS are necessary for practical applications.

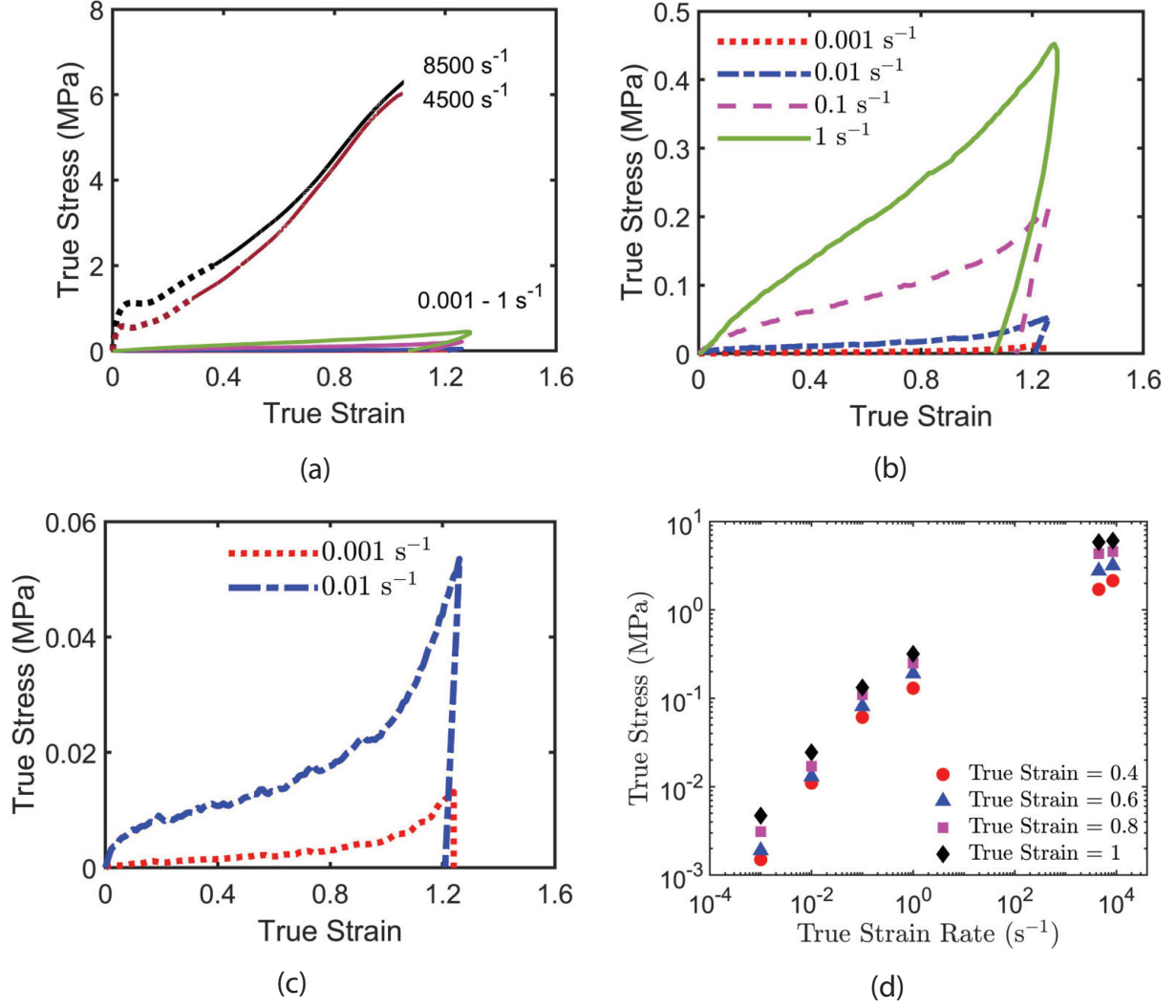


Figure 1.3: **The significantly rate-dependent, non-linear stress-strain response of PBS.** (A) Large strain compression response of PBS in the quasi-static to high (0.001 s^{-1} to 8500 s^{-1}) strain-rate range. The dashed line for 4500 s^{-1} and 8500 s^{-1} corresponds to the region where the ratio between the front and back face of the specimen was less than 0.7. (B) Zoomed-in view of the response spanning strain rates of 0.001 s^{-1} to 1 s^{-1} . (C) Zoomed-in view of the response at the strain rates of 0.001 s^{-1} and 0.01 s^{-1} . (D) True stress-strain rate curves at different true strain values showing a substantial rate-dependent stiffening of PBS. The stresses increase by more than three orders of magnitude as we go from the strain rate of 0.001 s^{-1} to 8500 s^{-1}

PBS: Effect of exposure to moisture in the air at 25° C

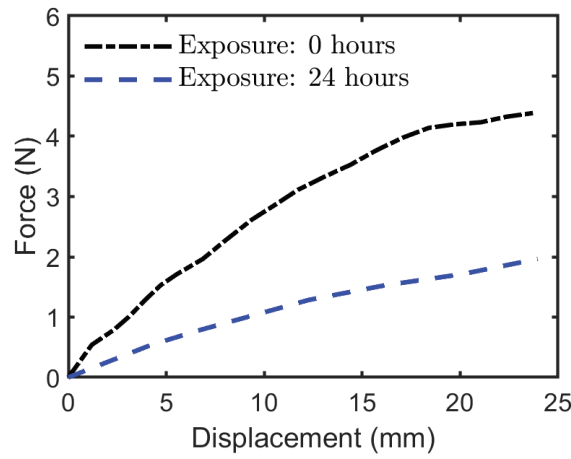


Figure 1.4: **PBS: Effect of exposure to moisture in the air at 25° C.** The ‘stretching using inserts’ setup discussed in the upcoming Chapter 2 (Section 2.8.4) was used to quantify the response of PBS exposed to moisture in the air at 25° C for 0 and 24 hours. The extension force (insert separation speed of 20 mm/s) was significantly reduced in the exposed specimens. Moisture in the air hydrolyzes the bonds necessary for dynamic crosslinking, resulting in softening of the response (Liu et al., 2023). Practical applications of PBS will require strategies to prevent water contact.

1.2.5 Imparting shape stability to PBS using open cell aluminum foam

As shown in Figure 1.1(A), PBS lacks shape stability and flows under its own weight. A strategy used in the literature to impart shape stability to PBS is to incorporate it into open-cell foams (Tu et al., 2023; Sang et al., 2021). Aluminum open-cell foam (Duocel Inc., Al 6101 T6, 1 in $\times$ 1 in $\times$ 0.5 in, approximately isotropic) was selected, which is a lightweight energy absorber. Filling it with PBS should enhance its impact protection capabilities. Metal foams are relatively insensitive to environmental factors compared with their polymer counterparts. However, a disadvantage is the lack of flexibility.

The two largest available pore sizes per inch (PPI), 20 and 40, were selected to facilitate efficient PBS incorporation. The PBS-filled foam composite was fabricated via a one-pot process in which the foam was placed in a container with PBS precursors and heated. The porosities of the 20 and 40 PPI unfilled foams were calculated using the constituent aluminum density (2.7 g/cc), foam dimensions, and foam weights (1.77 and 1.64 g), yielding values of 91.6% and 92.6%, respectively. The percentage of foams filled with PBS was calculated from the weights of the filled foams (7.37 and 7.04 g) and the density of PBS (1.1 g/cc), yielding 67.8% and 64.7% for the 20 and 40 PPI foams, respectively. The relatively low filling percentages can be attributed to the dissimilar surface properties of PBS and the foam. Figure 1.5(A) shows the top views of the unfilled, uncrushed; filled, uncrushed; unfilled, crushed (70% engineering strain); and filled, crushed 40 PPI foam specimens. PBS was satisfactorily constrained even in the crushed state by the foam. The compressive engineering stress-strain response of the 20 and 40 PPI filled and unfilled foams was characterized using a servo-hydraulic MTS machine. Figure 1.5(B) shows the approximately rate-independent response of the unfilled foam. The comparison of the unfilled, pure PBS, and filled foam responses (0.7 s^{-1}) for both PPIs is shown in Figure 1.5(C). The unfilled foam is significantly stiffer than PBS. The plateau stress increased

upon the incorporation of PBS; that is, more energy was absorbed during deformation at this strain rate.

Experimental characterization at higher strain rates is required to confirm the synergistic effect of PBS and foam for impact protection. A few studies have hypothesized the physical mechanisms underlying this synergy (Liu et al., 2019b). Further micro- and macroscale experimental studies are required to develop a comprehensive understanding. A different, flexibility-retaining approach for imparting shape stability to PBS, which uses a soft polymer network as a scaffold, is presented in Chapter 5.

1.3 Theory and constitutive model

The key features needed to be captured by a three-dimensional, large deformation constitutive model for PBS are: (i) non-linear large strain response, (ii) kinetics of the dynamic crosslinks involved, and (iii) significant rate-dependent stiffening of the material response.

We base our theory on the classical Kroner (Kröner, 1959)-Lee (Lee, 1969) multiplicative decomposition of the deformation gradient $\mathbf{F}$ into elastic and plastic parts $\mathbf{F}^e$ and $\mathbf{F}^p$, which is commonly used in elastic-viscoplastic and visco-elastic constitutive theories (Boyce et al., 1988; Vogel et al., 2014; de Rooij and Kuhl, 2016; Kothari et al., 2019; Shen et al., 2019; Saadedine et al., 2021; Niu et al., 2023; Lin et al., 2023; Wang et al., 2023a)

$$\mathbf{F} = \mathbf{F}^e \mathbf{F}^p \quad \text{with} \quad \det \mathbf{F}^e > 0 \quad \text{and} \quad \det \mathbf{F}^p > 0. \quad (1.1)$$

We utilize the parallel “multi-mechanism” generalization of the multiplicative decomposition in (1.1) that accounts for various rate-dependent microscopic relaxation mechanisms (Bergström and Boyce, 1998; Anand et al., 2009; Ames et al., 2009; Srivastava et al., 2010a,b; Li et al., 2017; Garcia-Gonzalez et al., 2017; Mahjoubi et al., 2019; Hao et al.,

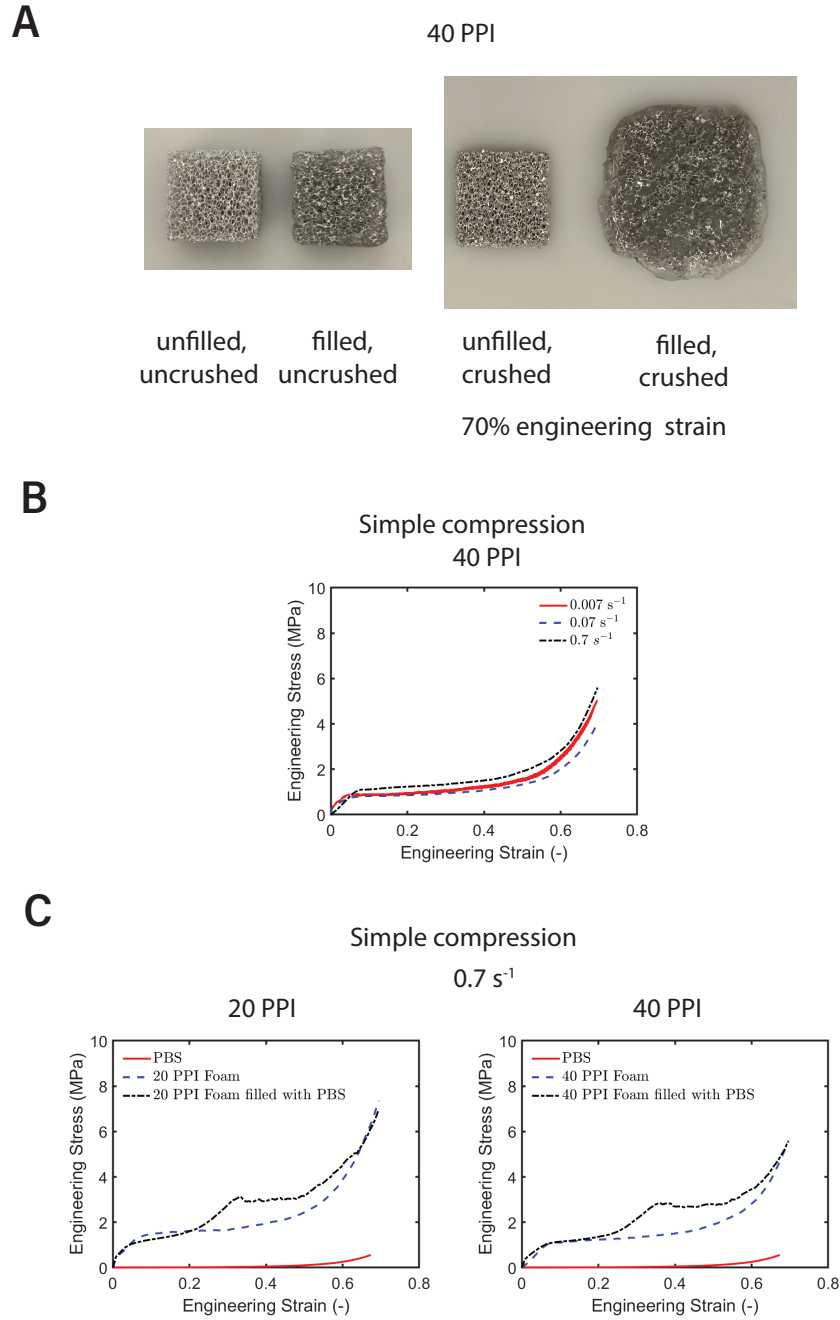


Figure 1.5: PBS: open cell aluminum foam for imparting shape stability. Aluminum open-cell foam (Duocel Inc., Al 6101 T6, 1 in x 1 in x 0.5 in, approximately isotropic) with the two highest available porosities (20 PPI - 91.6%, 40 PPI - 92.6%) was used. **(A)** Qualitative comparison of filled and unfilled 40 PPI foam before and after crushing (70% engineering strain). Satisfactory PBS containment was achieved even after crushing. **(B)** The approximately rate-independent response of the unfilled foam (40 PPI). **(C)** Comparison of compressive engineering stress-strain curves ($0.7 s^{-1}$) for pure PBS, unfilled, and filled foam specimens. The foam was significantly stiffer than PBS. The PBS-filled foam exhibited a higher plateau stress, indicating that it absorbed more energy during deformation. The presence of this phenomenon at higher strain rates will result in improved impact protection capabilities.

2022; Xiao et al., 2022), i.e.

$$\mathbf{F} = \mathbf{F}^{e(\alpha)} \mathbf{F}^{p(\alpha)} \quad \text{with} \quad \det \mathbf{F}^{e(\alpha)} > 0 \quad \text{and} \quad \det \mathbf{F}^{p(\alpha)} > 0, \quad \alpha = 1, \dots, M, \quad (1.2)$$

where each α represents one of the M number of mechanisms in a general case acting in parallel that captures a particular macroscopic aspect of the material response. For a given material point $\mathbf{X}$, with invertible $\mathbf{F}^{e(\alpha)}$ and $\mathbf{F}^{p(\alpha)}$,

$$\mathcal{M}_{\mathbf{X}}^{(\alpha)} \stackrel{\text{def}}{=} \text{range of } \mathbf{F}^{p(\alpha)}(\mathbf{X}) = \text{domain of } \mathbf{F}^{e(\alpha)}(\mathbf{X}), \quad (1.3)$$

is referred to as the intermediate structural space at a material point $\mathbf{X}$ for a given α , which is a purely mathematical construct. We assume PBS as an isotropic material. An isothermal constitutive model is developed for applications involving near room temperature conditions owing to a very small estimated temperature rise (only a few degrees Celsius) due to inelastic dissipation even for the strain rates of 4500 s^{-1} and 8500 s^{-1} .

1.3.1 Dynamic, reversible crosslinks

B:O coordinate-bond dynamic crosslinks, rheological test predictions

During the synthesis of PBS, hydroxy end groups on Boric Acid (BA) and hydroxy-terminated Polydimethylsiloxane (PDMS) chains in the entangled melt condense to form Silicon(Si)-O-B bonds connecting the ends of hydroxy-terminated PDMS chains through boron atoms and form longer chain molecules (Tang et al., 2016). The electron-deficient p orbitals of these connecting boron atoms can further receive a pair of electrons from oxygen atoms in the dimethylsiloxane backbone of neighboring end-connected PDMS

chains to form dynamic coordinate-bond B:O bonds that act as dynamic crosslinks with the ability to relax and reconnect (Li et al., 2014). We use “subchains” to denote parts of chain molecules between two elastically active crosslinks (connected to the elastically active network). We further use the superscript “ bd ” to denote quantities associated with subchains between B:O coordinate-bond dynamic crosslinks. We denote the referential number density of chain segments currently in their “subchain” state (S) corresponding to dynamic B:O coordinate-bond crosslinks as n_s^{bd} and that of chain segments which can be in the (S) state but currently are in the “not subchain” state (NS) as n_{ns}^{bd} . The underlying dynamic, reversible reactions can be represented as



where k_s^{bd} and k_{ns}^{bd} are the forward and backward reaction rate parameters. Assuming first-order kinetics to model the reactions, we have

$$\dot{n}_s^{bd} = k_s^{bd} n_{ns}^{bd} - k_{ns}^{bd} n_s^{bd}. \quad (1.5)$$

We now focus on the shear stress relaxation data in Section 1.2.2. We assume that (i) the new crosslinks and subchains are formed in their stress-free, relaxed configurations, (ii) B:O coordinate-bond crosslinks are the dominant type of dynamic crosslinks with relaxation timescales within those of the shear stress relaxation and oscillatory shear experiments, (iii) relaxation of B:O coordinate-bond dynamic crosslinks is the dominant relaxation mechanism in the timescales of the rheology experiments, (iv) the reaction rate parameters are approximately constant under the applied small strain loadings, and (v) we start from a spatially homogenous, stress-free, equilibrium state of the kinetics at $t=0$ i.e., $n_s^{bd}(0)$ and $n_{ns}^{bd}(0)$ related through $n_{ns}^{bd}(0) = [k_{ns}^{bd}/k_s^{bd}] n_s^{bd}(0)$.

For the case of stress relaxation, the subchains formed after the strain ramp do not

stretch and do not carry any load during the hold period. Further, following the theory of rubber elasticity (Treloar, 1943) and assumption (iii), the shear stress relaxation modulus $\mu(t)$ is given by

$$\mu(t) = n_{load}^{bd}(t)k_B\theta, \quad (1.6)$$

where k_B is the Boltzmann constant and n_{load}^{bd} denotes the referential number density of load bearing subchains during the hold period.⁴ Considering only the stress decay (relaxation) corresponding to the conversion of load-bearing subchains to the not-subchain state, the form of (1.5) for the evolution of n_{load}^{bd} during the hold period ($t \geq t_{ramp}$) reduces to

$$\dot{n}_{load}^{bd} = -k_{ns}^{bd}n_{load}^{bd}, \quad n_{load}^{bd}(t) = n_s^{bd}(0)e^{-k_{ns}^{bd}t} \quad (\text{for } t \geq t_{ramp}). \quad (1.7)$$

Substituting (1.7) in (1.6), we get

$$\mu(t) = \mu(0)e^{-k_{ns}^{bd}t}, \quad (1.8)$$

and

$$\mu(0) = n_s^{bd}(0)k_B\theta. \quad (1.9)$$

As seen in Figure 1.6(A), equations (1.8), (1.9) fit the shear stress relaxation experimental data well with $n_s^{bd}(0) = 6.4 \times 10^{24} \text{ m}^{-3}$ and $k_{ns}^{bd} = 0.35 \text{ s}^{-1}$. Further, (1.8) is identical to the expression for shear stress relaxation modulus of a Maxwell element,

⁴Since the strain ramp for the stress relaxation experiment is faster than k_{ns}^{bd} and k_s^{bd} , all $n_s^{bd}(0)$ subchains at $t=0$ will be stretched to 1% strain at the end of the strain ramp. Hence, $n_{load}^{bd}(t_{ramp}) = n_s^{bd}(0)$ and all $n_{load}^{bd}(t)$ load bearing subchains at any time instant $t \geq t_{ramp}$ are at 1% strain.

$$\mu(t) = \mu(0) e^{-\frac{t}{\tau}}, \quad (1.10)$$

and

$$\tau = \frac{1}{k_{ns}^{bd}}, \quad (1.11)$$

quantifying the B:O coordinate-bond dynamic crosslink *macroscopic* relaxation timescale.

This implies that PBS will behave like a Maxwell element in other cases of small-strain loadings as well. Considering the linear viscoelastic Maxwell element-like behavior, the storage and loss moduli $G'(\omega)$, $G''(\omega)$ for a sinusoidal loading corresponding to a frequency of ω for PBS are given as

$$G'(\omega) = \mu(0) \frac{\omega^2}{\omega^2 + (2\pi k_{ns}^{bd})^2}, \quad G''(\omega) = \mu(0) \frac{2\pi\omega k_{ns}^{bd}}{\omega^2 + (2\pi k_{ns}^{bd})^2}. \quad (1.12)$$

Response predictions for the oscillatory shear loading obtained using (1.12) and $\mu(0)$, k_{ns}^{br} from fit to the shear stress relaxation response are in good agreement with the experimental results as seen in Figure 1.6(B). The fit of the shear stress relaxation data and reasonably good prediction for the oscillatory shear loading support the assumption of B:O coordinate-bond dynamic crosslink relaxation being the dominant relaxation mechanism in the shear stress relaxation and oscillatory shear experimental timescales. This also supports the assumptions of new crosslinks and subchains being formed in their stress-free relaxed configurations and the reaction rate parameters being approximately constant under the applied small strain loading. When the loading timescales are such that $t_{\text{load}} \ll 1/k_{ns}^{bd}$, the dynamic B:O coordinate-bond crosslinks do not have sufficient time to dissociate. Consequently, PBS will resemble a network with “permanent” crosslinks. Therefore, PBS will behave like a Maxwell element up to frequencies near the crossover frequency $\omega_c =$

$2\pi k_{ns}^{bd}$ and will transition to “permanent” network-like behavior with B:O coordinate-bond crosslinks for frequencies beyond ω_c . The plateauing of G' after the crossover frequency, when a permanent-like network is approached, suggests approximately constant number density of load-bearing subchains, i.e., negligible formation of new subchains which are stretched in these relatively short timescales. This indicates that k_s^{bd} is comparable to or smaller than $k_{ns}^{bd} = 0.35 \text{ s}^{-1}$.

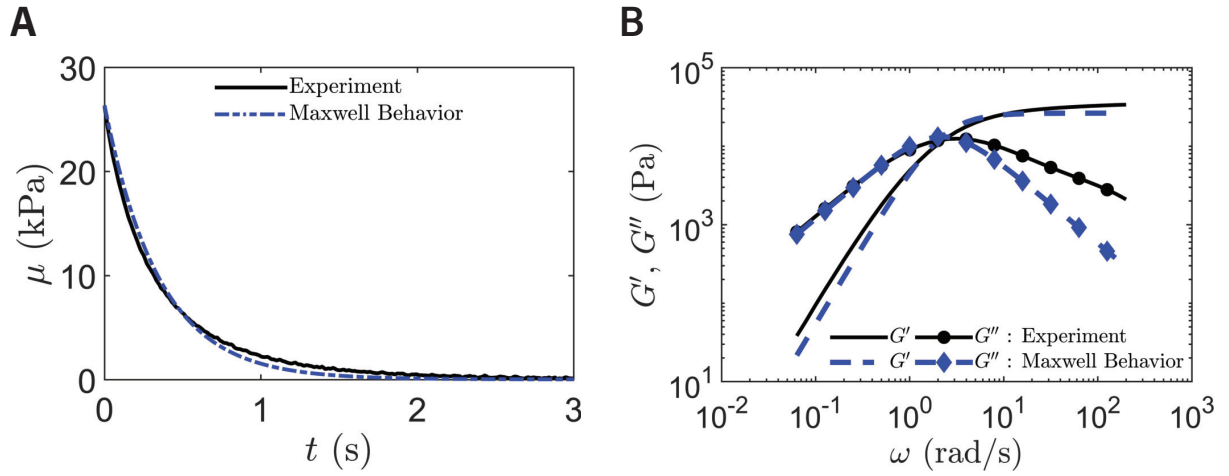


Figure 1.6: **The Maxwell-element like behavior of PBS under small strain deformation.** (A) Fit of the linear Maxwell-like response of PBS at small strains, assuming first-order kinetics for the B:O coordinate-bond dynamic crosslink relaxation mechanism. The new subchains are formed in their stress-free, relaxed configurations. (B) Oscillatory shear loading predictions of G' and G'' considering the Maxwell-like behavior with shear stress relaxation test fitted parameters $n_s^{bs}(0)$, k_{ns}^{bd} .

Temporary entanglement lockups at high strain rates

Using the fitted parameter $n_s^{bd}(0)$ and (1.9), we get a ground state glassy shear modulus $\mu(0)$ of approximately 26 kPa. For loading timescales $t_{load} \lll 1/k_{ns}^{bd}$, the subchain kinetics can be neglected, i.e., a “permanent” - like network is approached. B:O coordinate-bond dynamic crosslinks with $\mu(0) = 26 \text{ kPa}$ are not sufficient to explain the relatively substantially larger stress levels observed at the high strain rate of 4500 s^{-1} ($t_{load} = 1/(4500 \text{ s}^{-1}) \lll 1/k_{ns}^{bd}$) in Figure 1.3(A). This suggests a possibility of the presence of an additional type of crosslinked network with crosslink relaxation timescales much shorter

than those involved in the oscillatory shear experiments.

Previous works, including molecular dynamics simulations (Zheng et al., 2023) and macroscopic experiments (Malkin et al., 2012) have suggested that temporary entanglement lockups can occur in entangled polymer melts at high strain rates through the tightening of interchain twists and knots, which are randomly created and destroyed. At low strain rates, the twists and knots can disentangle, and chains can slip past each other and flow similar to the tube model predictions. The twists and knots tightening up at high strain rates enable the transition of entangled polymer melts from viscous flow to rubbery-like behavior at high strain rates. The entanglement lockups, which temporarily occur at high strain rates, can be thought of as reversible crosslinks with very short relaxation timescales. PBS shows behavior similar to entangled polymer melts at slow strain rates, we suggest an additional network stretching resistance coming from temporary entanglement lockups at high strain rates acting as crosslinks with very short relaxation timescales.⁵

We use the superscript “*el*” to denote quantities associated with subchains between entanglement lockups. Figure 1.7 shows a schematic representation of the B:O coordinate-bond dynamic crosslinks and entanglements.

Force dependence of reaction rate parameters

The reaction rate parameters k_{ns} and k_s experienced by subchains formed at t' ($0 < t' < t$) and surviving till the current time instant t will, in general, depend on the forces experienced by them (Bell, 1978). The effect of forces acting on the subchains on the reaction rate parameters can be qualitatively understood using a simple analysis similar to the work of Mayumi and co-workers (Mayumi et al., 2013). The subchains formed at $t=0$ and surviving till t would experience the largest stretch levels equal to the total

⁵PBS without entanglements (synthesized using PDMS melts with molecular weights below the entanglement weight of PDMS) show oscillatory shear response similar to the results in Figure 1.2(A) (Tang et al., 2016; Kurkin et al., 2021). This indicates that the relaxation timescale of 0.35 s^{-1} is related to the macroscopic relaxation of B:O coordinate-bond dynamic crosslinks.

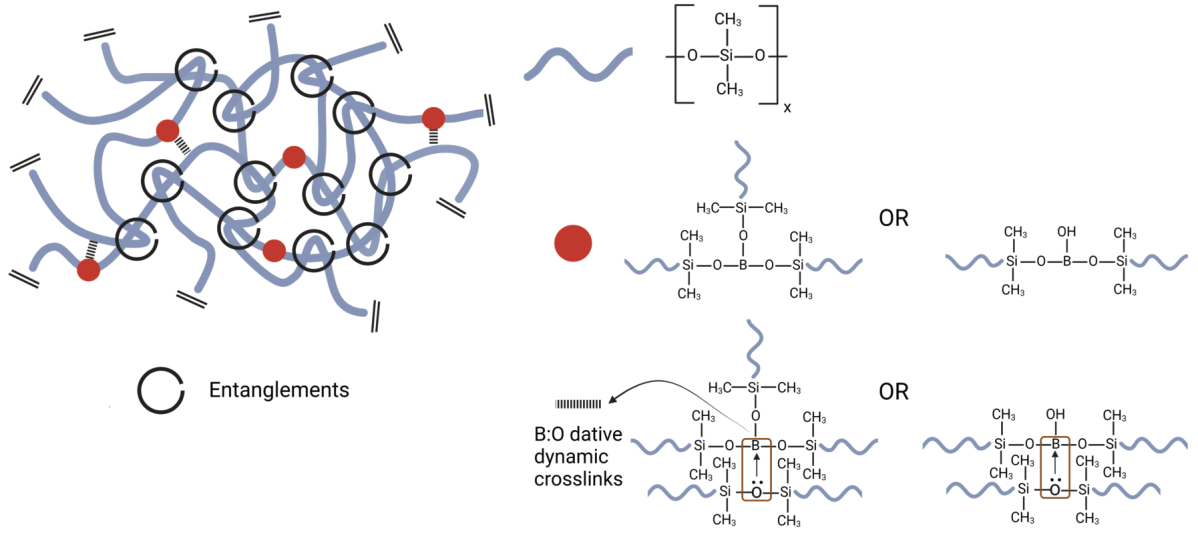


Figure 1.7: **Schematic of B:O coordinate-bond dynamic crosslinks and entanglements in PBS.**

imposed stretch and hence the largest forces. With the Neo-Hookean model as an example, a reduced stress measure under uniaxial loading for the subchains formed at $t = 0$ and surviving till t is defined as $\sigma_r = \sigma/(\lambda^2 - \lambda^{-1})$. Here σ is the true stress, λ is the total imposed stretch, and σ_r is a function of the reaction rate parameters. Figure 1.8(A) and (B) are $\sigma_r - \lambda$ and $\sigma_r - t$ curves obtained using the compression experimental data in Section 1.2.3. Figure 1.8(A) shows that σ_r is approximately constant with λ indicating a weak dependence of the reaction rate parameters on force for the experimental stretch range in this chapter. This translates to $\sigma_r - t$ curves for strain rates from 0.001 s^{-1} to 1 s^{-1} in Figure 1.8(B) being close to overlapping each other till moderate stretch levels before finite extensibility of the chains not accounted for by the Neo-Hookean model comes into the picture. Consequently, we make a reasonable assumption of the reaction rate parameters being approximately constant in the stretch range of our experiments.

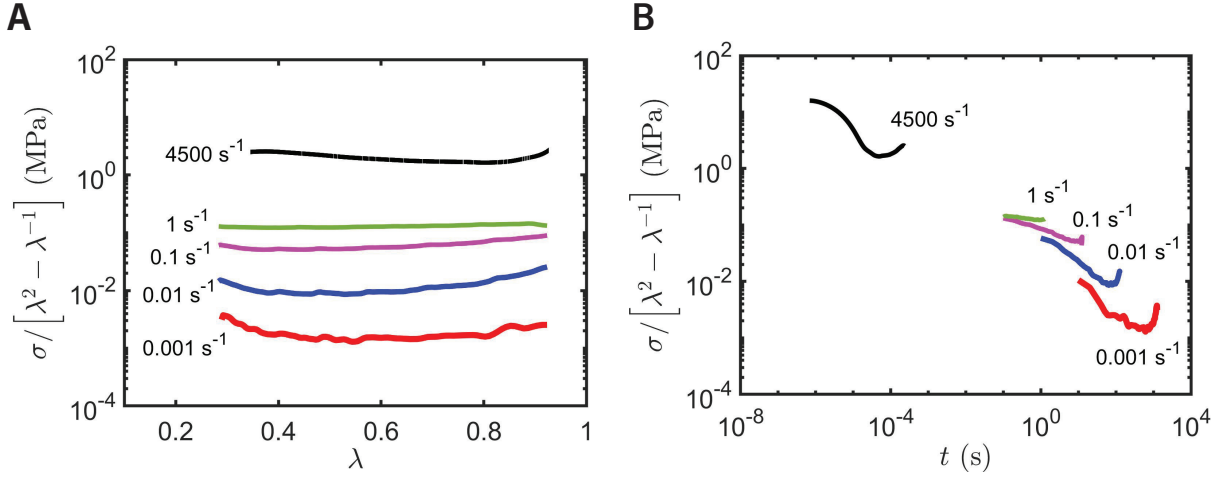


Figure 1.8: **Approximate force independence of reaction rate parameters through a reduced stress analysis.** **(A)** Reduced stress, $\sigma_r = \frac{\sigma}{\lambda^2 - \lambda^{-1}}$ vs compressive stretch, λ curves for strain rates from 0.001 s^{-1} to 4500 s^{-1} obtained using the compression experimental data in Section 1.2.3. σ_r is approximately constant with λ indicating weak dependence of the reaction rate parameters on force for the experimental stretch range in this study. **(B)** $\sigma_r - t$ curves for the wide range of strain rates. The curves are close to overlapping each other for the strain rates of 0.001 s^{-1} to 1 s^{-1} till moderate stretch levels, reflecting the approximate force independence of the reaction rate parameters.

1.3.2 Mechanisms of deformation

We propose *three parallel mechanisms* of deformation (A , $B1$, and $B2$) to describe the experimentally observed response of PBS over the strain rate range of 10^{-3} s^{-1} to 10^3 s^{-1} .

(a) **Intermolecular Resistance (A)**: Elastic and dissipative resistance due to intermolecular interactions between neighboring molecules is represented by mechanism A . It can be considered as a non-linear spring in series with a dashpot.

The network resistances are modeled by mechanism B .

(b) **Network Resistance (B)**: Resistance due to changes in the free energy upon stretching of the elastically active networks, i.e., stretching of constituent subchains within crosslinks corresponding to

- **B:O coordinate-bond dynamic crosslinks** represented by mechanism ($B1$), and
- **Temporary entanglement lockups at high strain rates acting as crosslinks** represented by mechanism ($B2$).

At very low strain rates, when the crosslinks between long chains are mechanically ineffective, PBS behaves as an entangled polymer melt, and dissipative molecular resistance (mechanism A) due to chains sliding past each other provides the primary contribution to the mechanical response. As the strain rate increases, and networks due to crosslinks begin to form, the *rate-dependent* network resistance (networks $B1$ and $B2$) starts to contribute dominantly to the mechanical response, and the contribution of mechanism A to the overall rate-dependent stress response becomes less significant.

The total referential free energy density can be additively decomposed into contributions from each mechanism of deformation as

$$\psi_R = \underbrace{\psi^{(A)}}_{\text{Intermolecular resistance}} + \underbrace{\psi^{(B1)} + \psi^{(B2)}}_{\text{Network resistance}}. \quad (1.13)$$

The total Cauchy stress $\mathbf{T}$ following the parallel multi-mechanism generalization and additive decomposition of the total free energy density in (1.13) is the sum of the contributions from each of the parallel mechanisms, i.e.

$$\mathbf{T} = \mathbf{T}^{(A)} + \mathbf{T}^{(B1)} + \mathbf{T}^{(B2)}, \quad (1.14)$$

where $\mathbf{T}^{(A)}$, $\mathbf{T}^{(B1)}$ and $\mathbf{T}^{(B2)}$ are the Cauchy stress contributions from mechanisms A , $B1$ and $B2$, respectively. A rheological schematic of the model and the mechanisms of deformation is shown in Figure 1.9.

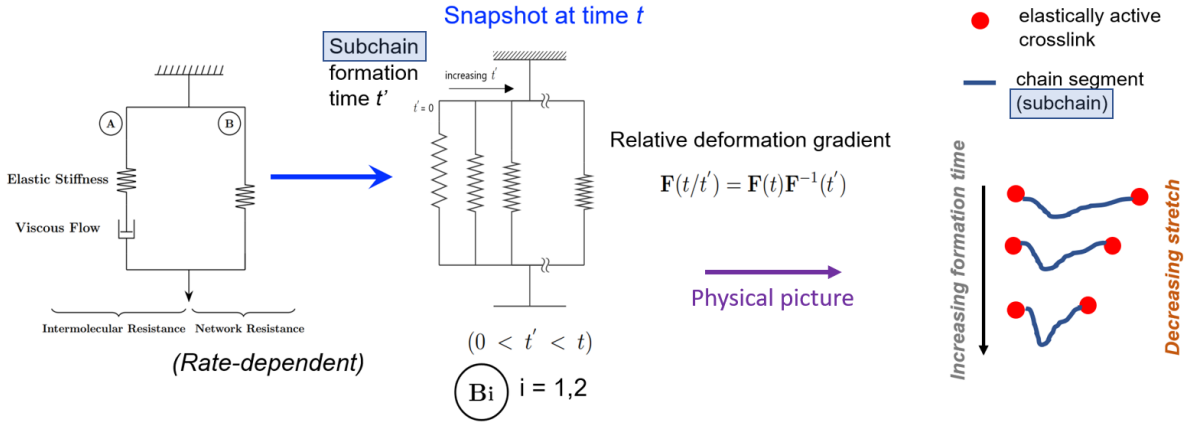


Figure 1.9: **Rheological schematic of model and mechanisms of deformation.**

1.3.3 Constitutive equations for intermolecular mechanism A

Free energy density, Cauchy stress and Mandel stress:

With $\mathbf{E}^{e(A)} = \ln \mathbf{U}^{e(A)}$ denoting an elastic logarithmic strain measure, where $\mathbf{U}^{e(A)}$ is the right elastic stretch tensor ($\mathbf{F}^{e(A)} = \mathbf{R}^{e(A)} \mathbf{U}^{e(A)}$, $\mathbf{R}^{e(A)}$ is the rotation tensor), we adopt for $\psi^{(A)}$ the following special form for moderately large elastic stretches (Anand, 1979, 1986)

$$\psi^{(A)} = \bar{\psi}^{(A)}(\mathbf{E}^{e(A)}) = G \left| \mathbf{E}_0^{e(A)} \right|^2 + \frac{1}{2} K (\text{tr} \mathbf{E}^{e(A)})^2, \quad (1.15)$$

where $G > 0$, $K > 0$ are the shear and bulk modulus respectively. The contribution $\mathbf{T}^{(A)}$ corresponding to the special free energy function considered in (1.15) is given by

$$\mathbf{T}^{(A)} = J^{e(A)-1} \mathbf{R}^{e(A)} \mathbf{M}^{e(A)} \mathbf{R}^{e(A)\top}, \quad \mathbf{M}^{e(A)} = \frac{\partial \bar{\psi}^{(A)}(\mathbf{E}^{e(A)})}{\partial \mathbf{E}^{e(A)}} = 2G \mathbf{E}_0^{e(A)} + K (\text{tr} \mathbf{E}^{e(A)}) \mathbf{1}. \quad (1.16)$$

where $\mathbf{M}^{e(A)}$ is the Mandel stress driving the plastic flow and is defined with respect to the local intermediate structural space for mechanism A .

Flow rule

The evolution equation and initial condition for $\mathbf{F}^{p(A)}$ with the assumption of isotropy and resulting plastic irrotationality, as well as the standard assumption of the body initially being in its virgin state, are given as

$$\dot{\mathbf{F}}^{p(A)} = \mathbf{D}^{p(A)} \mathbf{F}^{p(A)}, \quad \mathbf{F}(\mathbf{X}, 0) = \mathbf{F}^{e(A)}(\mathbf{X}, 0) = \mathbf{F}^{p(A)}(\mathbf{X}, 0) = \mathbf{1}. \quad (1.17)$$

Assuming codirectionality of plastic flow, the plastic stretching tensor $\mathbf{D}^{p(A)}$ can be

⁶ $\mathbf{A}_0$ denotes the deviatoric part of a tensor $\mathbf{A}$.

expressed as

$$\mathbf{D}^{p(A)} = \nu^{p(A)} \left(\frac{\mathbf{M}_0^{e(A)}}{2\bar{\tau}^{(A)}} \right), \quad \nu^{p(A)} \stackrel{\text{def}}{=} \sqrt{2} |\mathbf{D}^{p(A)}|, \quad \bar{\tau}^{(A)} \stackrel{\text{def}}{=} \frac{1}{\sqrt{2}} |\mathbf{M}_0^{e(A)}|, \quad (1.18)$$

where $\nu^{p(A)}$ is an equivalent plastic shear strain rate and $\bar{\tau}^{(A)}$ is an equivalent shear stress. We also define $\nu \stackrel{\text{def}}{=} \sqrt{2} |\mathbf{D}_0|$ as the total equivalent shear strain rate and $\bar{p}^{(A)} \stackrel{\text{def}}{=} -\frac{1}{3} \text{tr} \mathbf{M}^{e(A)}$ as the mean normal pressure. The distortional parts of $\mathbf{F}$, $\mathbf{F}_{dis}$, the right Cauchy green deformation tensor ($\mathbf{C}$), $\mathbf{C}_{dis}$ and the effective total distortional stretch, $\bar{\lambda}$ are defined as

$$\mathbf{F}_{dis} = J^{-1/3} \mathbf{F}, \quad \det \mathbf{F}_{dis} = 1; \quad \mathbf{C}_{dis} = (\mathbf{F}_{dis})^\top \mathbf{F}_{dis} = J^{-2/3} \mathbf{C}; \quad \bar{\lambda} \stackrel{\text{def}}{=} \sqrt{\frac{\text{tr} \mathbf{C}_{dis}}{3}}. \quad (1.19)$$

We choose the flow function for the evaluation of $\nu^{p(A)}$ in the following (similar to (Srivastava et al., 2010a)) form

$$\nu^{p(A)} = \begin{cases} 0 & \text{if } \bar{\tau}_e^{(A)} \leq 0, \\ \nu_0 \left[\sinh \left(\frac{\bar{\tau}_e^{(A)}}{S} \right) \right]^{1/m} & \text{if } \bar{\tau}_e^{(A)} > 0, \end{cases} \quad (1.20)$$

where

$$\bar{\tau}_e^{(A)} \stackrel{\text{def}}{=} \bar{\tau}^{(A)} - \alpha_p \bar{p}^{(A)}, \quad (1.21)$$

denotes a net equivalent shear stress for plastic flow. Here $\alpha_p > 0$ is a pressure sensitivity parameter, ν_0 is a reference strain rate, and m is a strain rate sensitivity parameter. The internal variable S models the dissipative friction-like resistance due to the sliding chains. The evolution of S is taken to be

$$\dot{S} = h (\bar{\lambda} - 1) \nu, \quad \text{with initial value } S(\mathbf{X}, 0) = S_0 > 0, \quad (1.22)$$

where h is a stress-dimensioned material parameter.

1.3.4 Constitutive relations for network stretching mechanisms

$B1$ and $B2$

Free energy and Cauchy stress

The rate-dependent network stretching mechanisms $B1$ and $B2$ contribute dominantly to the overall polymer response as the strain rates start to increase. Since we have already accounted for volumetric elastic energy in $\psi^{(A)}$, we do not allow for volumetric energy in $\psi^{(B1)}$ and $\psi^{(B2)}$. We consider free energy functions of the special form

$$\psi^{(B1)} = \hat{\psi}^{(B1)}(\mathbf{C}_{dis}), \quad \psi^{(B2)} = \tilde{\psi}^{(B2)}(\mathbf{C}_{dis}). \quad (1.23)$$

With the effective total distortional stretch, $\bar{\lambda} \stackrel{\text{def}}{=} \sqrt{\frac{\text{tr} \mathbf{C}_{dis}}{3}}$, we utilize (with modification) the eight-chain free energy density function denoted by ψ (Arruda and Boyce, 1993; Bergström and Boyce, 1998; Boyce and Arruda, 2000; Qi and Boyce, 2004) for this purpose

$$\psi = \mu \lambda_L^2 \left[\left(\frac{\bar{\lambda}}{\bar{\lambda}_L} \right)^\beta + \ln \left(\frac{\beta}{\sinh \beta} \right) \right], \quad (1.24)$$

where

$$\mu > 0 \quad \text{and} \quad \bar{\lambda}_L > 0, \quad (1.25)$$

with μ representing the ground state shear modulus, $\bar{\lambda}_L$ being the network locking stretch

parameter accounting for finite chain extensibility, $\beta = \mathcal{L}^{-1} \left(\frac{\bar{\lambda}}{\bar{\lambda}_L} \right)$ and $\mathcal{L}(\cdot) = \coth(\cdot) - \frac{1}{(\cdot)}$ denoting the Langevin function.

However, with new subchains being formed in their stress-free, relaxed configurations, we need to reformulate the free energy density function in (1.24). With the reasonable assumption of the reaction rate parameters being approximately constant from Section 1.3.1, we make a few additional assumptions that enable the following analysis : (i) We only account for subchains between two B:O coordinate-bond dynamic crosslinks or two entanglement lockups and assume decoupled first-order kinetics for these two types of subchains. (ii) We start from a spatially homogenous, stress-free, equilibrium state of the kinetics at $t=0$, i.e. $n_s(0)$ and $n_{ns}(0)$ related through $n_{ns}(0) = [k_{ns}/k_s]n_s(0)$. As a result, n_s , n_{ns} will be spatially homogeneous and constant with values equal to $n_s(0)$, $n_{ns}(0)$ respectively. The subchains formed at t' ($0 < t' < t$) and surviving till the current time t are stretched with respect to their stress-free, relaxed configurations corresponding to t' . This can be quantitatively denoted by the relative deformation gradient: $\mathbf{F}(t/t') = \mathbf{F}(t)\mathbf{F}^{-1}(t')$, a temporal decomposition of $\mathbf{F}$ which maps the deformed configuration at t' to the deformed configuration at t . Hence, it is necessary to keep track of the subchain formation times. The development of an expression, which tracks the subchain formation times, and holds individually for both types of subchains, is outlined below:

After the first time step Δt , with the first order kinetics in (1.5), we have

$$n_s(\Delta t) = n_s(0) \left[1 - k_{ns} \Delta t \right] + n_{ns}(0) k_s \Delta t. \quad (1.26)$$

For $\Delta t \rightarrow 0$, using Taylor series expansion of e^x to simplify (1.26), we get

$$n_s(\Delta t) = n_s(0) e^{-k_{ns} \Delta t} + n_{ns}(0) k_s \Delta t. \quad (1.27)$$

Consequently, after the second time step, we have

$$n_s(2\Delta t) = n_s(0)e^{-k_{ns}\Delta t} \left[1 - k_{ns}\Delta t \right] + n_{ns}(0)k_s\Delta t \left[1 - k_{ns}\Delta t \right] + n_{ns}(0)k_s\Delta t. \quad (1.28)$$

Again, simplifying (1.28) for small $\Delta t \rightarrow 0$, we get

$$n_s(2\Delta t) = n_s(0)e^{-2k_{ns}\Delta t} + n_{ns}(0)k_s\Delta te^{-k_{ns}\Delta t} + n_{ns}(0)k_s\Delta t. \quad (1.29)$$

After N time steps, following (1.27) and (1.29), $n_s(N\Delta t)$ can be expressed as

$$n_s(N\Delta t) = n_s(0)e^{-Nk_{ns}\Delta t} + \sum_{i=1}^N n_{ns}(0)k_s\Delta te^{-(N-i)k_{ns}\Delta t}. \quad (1.30)$$

This evolution pattern can be written concisely in an integral form for $\Delta t \rightarrow 0$ as

$$n_s(t) = \underbrace{n_s(0)e^{-k_{ns}t}}_{n_s(t/0)} + \int_0^t \underbrace{n_{ns}(0)k_se^{-k_{ns}(t-t')}}_{n_s(t/t')} dt'. \quad (1.31)$$

The first term on the right-hand side of (1.31) denoted by $n_s(t/0)$ corresponds to subchains surviving from $t = 0$ till current time t , and the integrand in the second term denoted by $n_s(t/t')$ corresponds to the newly formed subchains at t' ($0 < t' < t$) and surviving till t . The stretch levels of $n_s(t/0)$ and $n_s(t/t')$ are quantified with $\bar{\lambda}(t/0)$ and $\bar{\lambda}(t/t')$ respectively. We use the notations n_s, n_{ns} for $n_s(0), n_{ns}(0)$ respectively in the following discussion for conciseness. With

$$\mu^{bd} = \mu^{bd}(0) = n_s^{bd}k_B\theta, \quad \mu^{el} = \mu^{el}(0) = n_s^{el}k_B\theta, \quad (1.32)$$

denoting the ground state shear modulus corresponding to a network with only B:O coordinate-bond dynamic crosslinks and the ground state shear modulus corresponding to a network with only entanglement lockups, respectively, $\psi^{(B1)}$ following (1.24) and (1.31)

is given as

$$\begin{aligned}
\psi^{(B1)} &= \underbrace{\left(\frac{n_s^{bd}(t/0)}{n_s^{bd}}\right) \mu^{bd} \bar{\lambda}_L^{-2} \zeta^{bd}(t/0)}_{\text{For subchains surviving from } t=0} + \underbrace{\int_0^t \left(\frac{n_s^{bd}(t/t')}{n_s^{bd}}\right) \mu^{bd} [\bar{\lambda}_L^{-bd}]^2 \zeta^{bd}(t/t') dt'}_{\text{For subchains formed at } t'} \\
&= e^{-k_{ns}^{bd} t} \mu^{bd} \bar{\lambda}_L^{-2} \zeta^{bd}(t/0) + \int_0^t \frac{n_{ns}^{bd}}{n_s^{bd}} k_s^{bd} e^{-k_{ns}^{bd}(t-t')} \mu^{bd} [\bar{\lambda}_L^{-bd}]^2 \zeta^{bd}(t/t') dt',
\end{aligned} \tag{1.33}$$

where

$$\zeta^{bd}(t/t') = \left[\left(\frac{\bar{\lambda}(t/t')}{\bar{\lambda}_L^{-bd}} \right) \beta^{bd}(t/t') + \ln \left(\frac{\beta^{bd}(t/t')}{\sinh \beta^{bd}(t/t')} \right) \right]. \tag{1.34}$$

The first term on the right-hand side of the free energy equation (1.33) corresponds to the free energy contribution from subchains surviving from $t = 0$ till current time t , and the integrand in the second term corresponds to free energy contribution from the newly formed subchains at t' and surviving till t . The Cauchy stress follows from standard calculations. $\mathbf{T}^{(B1)}$ corresponding to $\psi^{(B1)}$ is given as

$$\mathbf{T}^{(B1)} = e^{-k_{ns}^{bd} t} \mu^{bd} \mathbf{T}^{bd}(t/0) + \int_0^t \frac{n_{ns}^{bd}}{n_s^{bd}} k_s^{bd} e^{-k_{ns}^{bd}(t-t')} \mu^{bd} \mathbf{T}^{bd}(t/t') dt', \tag{1.35}$$

where

$$\mathbf{T}^{bd}(t/t') = J^{-1}(t/t') \frac{\bar{\lambda}_L^{-bd}}{\bar{\lambda}(t/t')} \mathcal{L}^{-1} \left[\frac{\bar{\lambda}(t/t')}{\bar{\lambda}_L^{-bd}} \right] \left(\mathbf{B}_{dis}(t/t') \right)_0, \tag{1.36}$$

and

$$\mathbf{B}_{dis} = \mathbf{F}_{dis} (\mathbf{F}_{dis})^\top = J^{-2/3} \mathbf{B}, \tag{1.37}$$

denotes the distortional part of the left Cauchy Green deformation tensor $\mathbf{B}$ with $\mathbf{F}_{dis}$ as defined in (1.19).

The twists and knots which result in temporary entanglement lockups at high strain rates will have a similar mechanical effect as the B:O coordinate-bond dynamic crosslinks. Hence, $\psi^{(B2)}$ is similarly given as

$$\psi^{(B2)} = e^{-k_{ns}^{el}t} \mu^{el} \bar{\lambda}_L^2 \zeta^{el}(t/0) + \int_0^t \frac{n_{ns}^{el}}{n_s^{el}} k_s^{el} e^{-k_{ns}^{el}(t-t')} \mu^{el} [\bar{\lambda}_L^{el}]^2 \zeta^{el}(t/t') dt', \quad (1.38)$$

with

$$\zeta^{el}(t/t') = \left[\left(\frac{\bar{\lambda}(t/t')}{\bar{\lambda}_L^{el}} \right) \beta^{el}(t/t') + \ln \left(\frac{\beta^{el}(t/t')}{\sinh \beta^{el}(t/t')} \right) \right]. \quad (1.39)$$

$\mathbf{T}^{(B2)}$ corresponding to $\psi^{(B2)}$ is

$$\mathbf{T}^{(B2)} = e^{-k_{ns}^{el}t} \mu^{el} \mathbf{\Gamma}^{el}(t/0) + \int_0^t \frac{n_{ns}^{el}}{n_s^{el}} k_s^{el} e^{-k_{ns}^{el}(t-t')} \mu^{el} \mathbf{\Gamma}^{el}(t/t') dt', \quad (1.40)$$

where

$$\mathbf{\Gamma}^{el}(t/t') = J^{-1}(t/t') \frac{\bar{\lambda}_L^{el}}{\bar{\lambda}(t/t')} \mathcal{L}^{-1} \left[\frac{\bar{\lambda}(t/t')}{\bar{\lambda}_L^{el}} \right] \left(\mathbf{B}_{dis}(t/t') \right)_0. \quad (1.41)$$

1.4 Material parameters in the model

1.4.1 Calibration

The set of model parameters in the constitutive model to be calibrated is

$$\{k_s^{bd}, k_{ns}^{bd}, n_s^{bd}, k_s^{el}, k_{ns}^{el}, n_s^{el}, G, K, \nu_0, m, \alpha_p, S_0, h, \bar{\lambda}_L^{bd}, \bar{\lambda}_L^{el}\}. \quad (1.42)$$

k_{ns}^{bd} and n_s^{bd} were obtained in Section 1.3.1 using the shear stress relaxation response. Following the conclusion in Section 1.3.1, we choose $k_s^{bd} \approx k_{ns}^{bd}$. n_s^{el} was estimated from the ground state shear modulus μ^{el} needed to achieve the stress magnitudes at 4500 s^{-1} and 8500 s^{-1} . λ_L^{bd} and λ_L^{el} were evaluated using the calibrated model parameters n_s^{bd} , n_s^{el} and standard physical arguments discussed in Section 1.4.2. We assume $k_s^{el} \approx k_{ns}^{el}$ and Poisson's ratio relating G and K equal to 0.49. The list in (1.42) then reduces to

$$\{k_{ns}^{el}, G, \nu_0, m, \alpha_p, S_0, h\}. \quad (1.43)$$

A one-dimensional version of the model was numerically implemented in MATLAB for further calibration. Given the substantially different orders of stress magnitudes over the wide range of strain rates under consideration, it is difficult to obtain unique values for the model parameters in (4.2). k_{ns}^{el} was estimated such that the contributions from mechanism $B2$ using n_s^{el} and λ_L^{el} evaluated earlier are insignificant in the strain rate range of 10^{-3} s^{-1} - 10^0 s^{-1} , where mechanisms A and $B1$ are sufficient to capture the response following the discussion in Section 1.3.1. The contribution from Mechanism $B1$ using n_s^{bd} , k_{ns}^{bd} , k_s^{bd} and λ_L^{bd} obtained earlier is negligible at the lowest strain rate of 10^{-3} s^{-1} . This is consistent with the discussion in Section 1.3.2, i.e., Mechanism A makes the primary contribution at very low strain rates. Initial estimates for the remaining parameters $G, \nu_0, m, \alpha_p, S_0, h$ from (4.2) related to Mechanism A were obtained by fitting the stress contribution from

Mechanism *A* to the experimental response at 10^{-3} s^{-1} using the lsqcurvefit function (non-linear curve-fitting in least-squares sense) with the trust-region-reflective algorithm in MATLAB. The obtained initial estimates for the model parameters in (4.2) were fine-tuned manually to best capture the experimental stress-strain curves from 10^{-3} s^{-1} to 8500 s^{-1} . The values of model parameters after calibration are listed in Table 1.1.⁷

Parameter	PBS
$k_s^{bd} (\text{s}^{-1})$	0.35
$k_{ns}^{bd} (\text{s}^{-1})$	0.35
$n_s^{bd} (\text{m}^{-3})$	6.4×10^{24}
$k_s^{el} (\text{s}^{-1})$	2×10^3
$k_{ns}^{el} (\text{s}^{-1})$	2×10^3
$n_s^{el} (\text{m}^{-3})$	2×10^{26}
$G (\text{MPa})$	0.4
$K (\text{MPa})$	20
$\nu_0 (\text{s}^{-1})$	2×10^{-3}
$m (-)$	0.95
$\alpha_p (-)$	0.11
$S_0 (\text{kPa})$	0.6
$h (\text{kPa})$	37.7
$\bar{\lambda}_L^{bd} (-)$	37.4
$\bar{\lambda}_L^{el} (-)$	6.7
$\rho (\text{kg m}^{-3})$	1100

Table 1.1: **Deformation model parameters for PBS.**

The fit of the constitutive model to the experimental compression stress-strain curves for PBS over 0.001 s^{-1} to 8500 s^{-1} strain rate is shown in Figure 1.10. The constitutive model reasonably fits the non-linear large strain loading-unloading, and substantial rate-dependent stiffening observed in PBS. In Section 1.3.1, the small strain shear stress relaxation and oscillatory shear response supported the assumption of B:O coordinate-bond dynamic crosslink relaxation being the dominant relaxation mechanism in the

⁷The one-dimensional parameters obtained from the calibration procedure, except for the list of parameters $\{\nu_0, \alpha_p, S_0, h\}$ remain unchanged when used in the three-dimensional equations of the constitutive model presented. Noting that $\tau\nu = \sigma\dot{\epsilon}$, $\sigma = \sqrt{3}\tau$, $\dot{\epsilon} = \frac{\dot{\nu}}{\sqrt{3}}$, the parameters in the above list can be converted from their one-dimensional form to their three-dimensional shear form using $\nu_0^{(shear)} = \sqrt{3}\nu_0^{(comp)}$, $\alpha_p^{(shear)} = \frac{1}{\sqrt{3}}\alpha_p^{(comp)}$, $S_0^{(shear)} = \frac{1}{\sqrt{3}}S_0^{(comp)}$, $h^{(shear)} = \frac{1}{3}h^{(comp)}$.

quasi-static (10^{-3} s^{-1}) to moderate (10^0 s^{-1}) strain rate range. The model predicts the stress relaxation response for a large strain case (compressively loaded at a strain rate of 0.1 s^{-1} to 20% true strain, and then the strain was held constant) well as shown in Figure 1.10(C). This further indicates that the B:O coordinate-bond dynamic crosslink is the dominant mechanism for slow to moderate rate loading for both small and large strains. The model predictions for some intermediate strain rates (10, 100, 500, 1000 s^{-1}) are shown in Figure 1.10(D) for completeness. It can be noted here that material response characterization in this intermediate strain rate regime for soft polymers is a challenging and active area of research. The comparison of these intermediate strain rate model predictions with corresponding experimental results in the future can validate the model further.

1.4.2 Estimates of subchain lengths in PBS

With the average number of monomers in the subchains as N_s and monomer length as b , the root mean squared (rms) end-to-end distance is given as $\vec{r}_s = \sqrt{\langle \mathbf{r}_s^2 \rangle} = \sqrt{N_s}b$. Length of the dimethylsiloxane monomer, b can be estimated using the Si-O bond length ($\approx 0.164 \text{ nm}$) and O-Si-O bond angle ($\approx 110^\circ$) as $b \approx 2 \times 0.164 \times \sin(55^\circ) \text{ nm}$, i.e., $b \approx 0.27 \text{ nm}$ (González Calderón et al., 2020). Estimates of the average molecular mass of subchains can be attained using $M_s = \rho R / (k_B n_s)$, where R is the gas constant. M_s^{bd} and M_s^{el} corresponding to B:O coordinate-bond dynamic crosslinks and entanglement lockups evaluated using model parameters n_s^{bd} , n_s^{el} , and the earlier relation are approximately 103.55 kg/mol and 3.3 kg/mol , respectively. Molecular mass ($\approx 0.074 \text{ kg/mol}$) and length of the dimethylsiloxane monomer, b , can be used to approximate the average subchain contour lengths, L_s^{bd} and L_s^{el} . Scaling the dimethylsiloxane monomer length with the ratio of M_s and dimethylsiloxane monomer molecular mass, results in $L_s^{bd} \approx 378 \text{ nm}$, $L_s^{el} \approx 12 \text{ nm}$ and $N_s^{bd} \approx 1400$, $N_s^{el} \approx 45$.⁸ Using $\vec{r}_s = \sqrt{N_s}b$, we get rms end-to-end distance for

⁸Note that the network locking stretch parameters $\bar{\lambda}_L^{bd}$ and $\bar{\lambda}_L^{el}$ can be calculated using the physical arguments that the stretch locking parameter is the ratio of the fully extended subchain length ($\approx N_s b$)

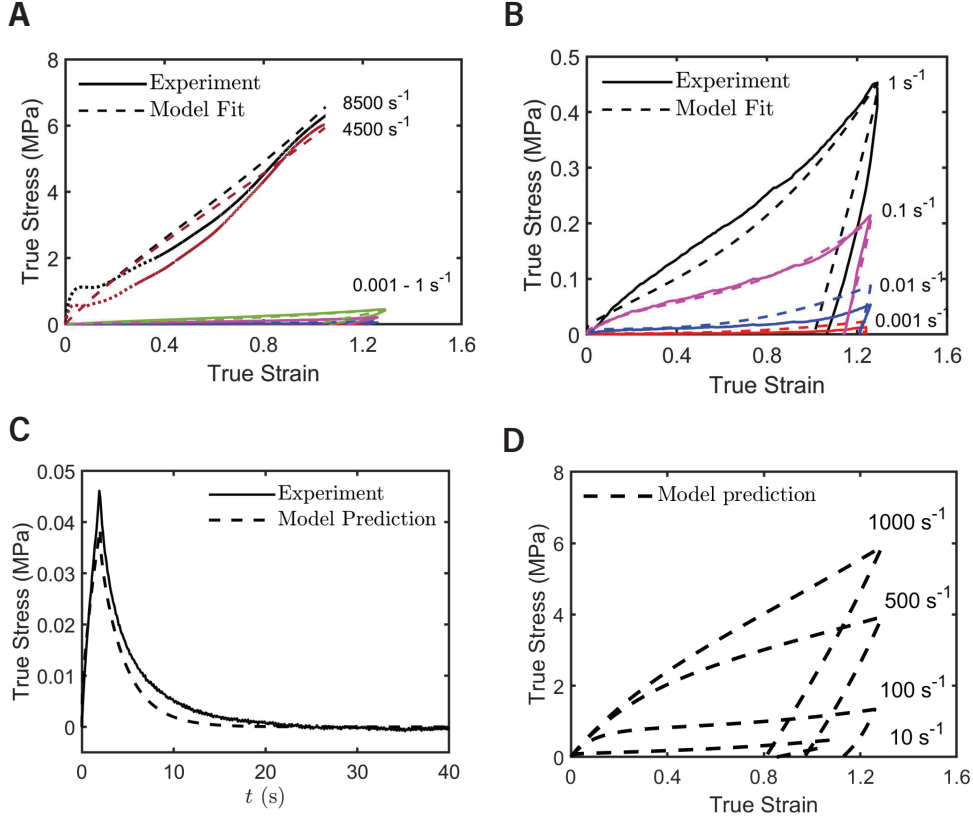


Figure 1.10: **Model fit to compression stress-strain curves and large strain compression stress relaxation prediction.** (A) Fit of the model (dashed lines) to experimental compression stress-strain curves (solid lines) for PBS over strain rates from 0.001 s^{-1} to 8500 s^{-1} . (B) Zoomed-in view of the fit to the experimental compression response for relatively lower strain rates from 0.001 s^{-1} to 1 s^{-1} . (C) Comparison of the model predicted large strain compression stress relaxation response (compressed at a strain rate of 0.1 s^{-1} to 20% true strain, then the strain was held constant) against the experimental result. The good agreement suggests that B:O coordinate-bond dynamic crosslink relaxation is the dominant relaxation mechanism in the quasi-static (10^{-3} s^{-1}) to moderate (10^0 s^{-1}) strain rate range, even at large strains. (D) Model predictions for some intermediate strain rates (10, 100, 500, 1000 s^{-1}). Material response characterization of soft polymers in this regime is a challenging and active area of research. Comparison of these model predictions with experiments in the future can enable further model validation.

subchains between B:O coordinate-bond dynamic crosslinks to be $\bar{r}_s^{bd} \approx 10 \text{ nm}$, and for the subchains between entanglement lockups as $\bar{r}_s^{el} \approx 1.8 \text{ nm}$.

to the corresponding initial root mean squared length ($\bar{r}_s = \sqrt{N_s}b$). Therefore, $\bar{\lambda}_L^{bd} \approx \sqrt{N_s^{bd}} \approx 37.4$ and $\bar{\lambda}_L^{el} \approx \sqrt{N_s^{el}} \approx 6.7$.

1.5 Validation experiments and numerical simulations

We conducted a few validation experiments not used to determine the parameters of our model and compared the experimental results against those from the corresponding three-dimensional numerical simulations. Indentation experiments resulting in inhomogeneous deformations were carried out on PBS rectangular slabs (blocks). Two distinct types of indenters were considered: (i) a conical indenter causing local deformation at the center of the block and (ii) a long, triangular prismatic indenter deforming blocks at two different loading rates. The experiments were conducted using UniVert Mechanical Test System (CellScale). The bottom surfaces of the blocks were completely fixed using a cyanoacrylate adhesive. The total external force required along the direction of indentation for a prescribed indenter displacement profile was measured in each case. The indenter noses were designed to have tip radii of 2 mm to prevent the tearing of PBS. The constitutive model was numerically implemented in ABAQUS/Explicit (2018)⁹ by writing a user-defined material subroutine (VUMAT). The finite element software ABAQUS has been applied for a variety of engineering simulations to solve problems involving complex geometry, loading, and materials behavior (Niu and Srivastava, 2022a,b; Bai et al., 2021; Zhong and Srivastava, 2021; Srivastava et al., 2011). ABAQUS returns only the deformation gradients from the previous and current time steps at each material point. Numerical implementation of the analytical expressions for $\mathbf{T}^{(B1)}$ in (1.35) and $\mathbf{T}^{(B2)}$ in (1.40) will require the storage of the entire deformation gradient history (the integrands in (1.35), (1.40) involve $\mathbf{\Gamma}(t/t') : 0 < t' < t$, a function of the relative deformation gradient $\mathbf{F}(t/t')$) along with many state-dependent variables at each material point. The resulting storage requirement will be huge, making numerical implementation extremely challenging.

⁹Explicit implementation procedure was chosen considering the short timescales of loadings involved in the applications of PBS. ABAQUS/Explicit uses the Green-Naghdi objective stress rate and the symmetric part of the velocity gradient as the objective deformation rate for the mechanical behavior of material defined using VUMAT in scenarios involving finite deformation and/or finite strains.

We have developed an approximate numerical method that allows for stress updates using only deformation gradients at the previous and current time steps. This eliminates the need for storage of the entire deformation gradient history at each material point and the associated large data storage requirements. The mathematical development is presented below:

Focusing on $\mathbf{T}^{(B1)}$ in (1.35), we have

$$\mathbf{T}^{(B1)}(t) = e^{-k_{ns}^{bd}t} \mu^{bd} \mathbf{\Gamma}^{bd}(t/0) + \int_0^t \frac{n_{ns}^{bd}}{n_s^{bd}} k_s^{bd} e^{-k_{ns}^{bd}(t-t')} \mu^{bd} \mathbf{\Gamma}^{bd}(t/t') dt'. \quad (1.44)$$

For the next time step, i.e. $t+\Delta t$, (1.44) can be rewritten as

$$\mathbf{T}^{(B1)}(t + \Delta t) = e^{-k_{ns}^{bd}(t+\Delta t)} \mu^{bd} \mathbf{\Gamma}^{bd}(t + \Delta t/0) + \int_0^{t+\Delta t} \frac{n_{ns}^{bd}}{n_s^{bd}} k_s^{bd} e^{-k_{ns}^{bd}(t+\Delta t-t')} \mu^{bd} \mathbf{\Gamma}^{bd}(t + \Delta t/t') dt'. \quad (1.45)$$

Further, $\mathbf{F}(t + \Delta t/t')$ and $J(t + \Delta t/t')$ can be split as

$$\mathbf{F}(t + \Delta t/t') = \mathbf{F}(t + \Delta t/t) \mathbf{F}(t/t') ; \quad J(t + \Delta t/t') = J(t + \Delta t/t) J(t/t'). \quad (1.46)$$

With (1.46), and (1.19), $\mathbf{C}_{dis}(t + \Delta t/t')$ can be expressed as

$$\mathbf{C}_{dis}(t + \Delta t/t') = J^{-2/3}(t/t') \mathbf{F}^T(t/t') \mathbf{C}_{dis}(t + \Delta t/t) \mathbf{F}(t/t'). \quad (1.47)$$

Using (1.47) and $\bar{\lambda} \stackrel{\text{def}}{=} \sqrt{\frac{\text{tr} \mathbf{C}_{dis}}{3}}$, $\bar{\lambda}(t + \Delta t/t')$ is given as

$$\bar{\lambda}(t + \Delta t/t') = \sqrt{\frac{\text{tr} \left[J^{-2/3}(t/t') \mathbf{F}^T(t/t') \mathbf{C}_{dis}(t + \Delta t/t) \mathbf{F}(t/t') \right]}{3}}. \quad (1.48)$$

For $\Delta t \rightarrow 0$, with (1.19), it follows that

$$\mathbf{C}_{dis}(t + \Delta t/t) \rightarrow \mathbf{I}. \quad (1.49)$$

Under the limit of $\Delta t \rightarrow 0$, (1.48) can be approximated using (1.49), (1.19) as

$$\bar{\lambda}(t + \Delta t/t') \rightarrow \bar{\lambda}(t/t'). \quad (1.50)$$

Following the definition of the deviatoric part of a tensor, $\left[\mathbf{B}_{dis}(t + \Delta t/t') \right]_0$ is given as

$$\left[\mathbf{B}_{dis}(t + \Delta t/t') \right]_0 = \mathbf{B}_{dis}(t + \Delta t/t') - \frac{1}{3} \text{tr} \left[\mathbf{B}_{dis}(t + \Delta t/t') \right]. \quad (1.51)$$

$\mathbf{B}_{dis}(t + \Delta t/t')$ can be expressed following (1.47) as

$$\mathbf{B}_{dis}(t + \Delta t/t') = J^{-2/3}(t + \Delta t/t) \mathbf{F}(t + \Delta t/t) \mathbf{B}_{dis}(t/t') \mathbf{F}^T(t + \Delta t/t). \quad (1.52)$$

Further, using (1.52), we get

$$\frac{1}{3} \text{tr} \left[\mathbf{B}_{dis}(t + \Delta t/t') \right] = \frac{1}{3} \text{tr} \left[J^{-2/3}(t + \Delta t/t) \mathbf{F}(t + \Delta t/t) \mathbf{B}_{dis}(t/t') \mathbf{F}^T(t + \Delta t/t) \right]. \quad (1.53)$$

(1.53) can be rewritten using the identity $\text{tr}(\mathbf{MN}) = \text{tr}(\mathbf{NM})$ as

$$\frac{1}{3}\text{tr}\left[\mathbf{B}_{dis}(t + \Delta t/t')\right] = \frac{1}{3}\text{tr}\left[\mathbf{B}_{dis}(t/t')\mathbf{C}_{dis}(t + \Delta t/t)\right]. \quad (1.54)$$

Again, for $\Delta t \rightarrow 0$, using (1.49), (1.54) can be approximated as

$$\frac{1}{3}\text{tr}\left[\mathbf{B}_{dis}(t + \Delta t/t')\right] \rightarrow \frac{1}{3}\text{tr}\left[\mathbf{B}_{dis}(t/t')\right]. \quad (1.55)$$

Finally, for $\Delta t \rightarrow 0$, substituting in (1.52), (1.55) in (1.51) and using (1.49), we get the approximation

$$\left[\mathbf{B}_{dis}(t + \Delta t/t')\right]_0 \rightarrow J^{-2/3}(t + \Delta t/t)\mathbf{F}(t + \Delta t/t)\left[\mathbf{B}_{dis}(t/t')\right]_0\mathbf{F}^T(t + \Delta t/t). \quad (1.56)$$

Denoting the second term in (1.45) as $\mathbf{T}_2^{(B1)}$, we have

$$\mathbf{T}_2^{(B1)}(t + \Delta t) = \int_0^{t+\Delta t} \frac{n_{ns}^{bd}}{n_s^{bd}} k_s^{bd} e^{-k_{ns}^{bd}(t+\Delta t-t')} \mu^{bd} \mathbf{\Gamma}^{bd}(t + \Delta t/t') dt'. \quad (1.57)$$

(1.57) can be decomposed into two integrals from (0 to t) and (t to $t + \Delta t$) as

$$\begin{aligned} \mathbf{T}_2^{(B1)}(t + \Delta t) &= \int_0^t \frac{n_{ns}^{bd}}{n_s^{bd}} k_s^{bd} e^{-k_{ns}^{bd}(t+\Delta t-t')} \mu^{bd} \mathbf{\Gamma}^{bd}(t + \Delta t/t') dt' \\ &+ \int_t^{t+\Delta t} \frac{n_{ns}^{bd}}{n_s^{bd}} k_s^{bd} e^{-k_{ns}^{bd}(t+\Delta t-t')} \mu^{bd} \mathbf{\Gamma}^{bd}(t + \Delta t/t') dt'. \end{aligned} \quad (1.58)$$

Focusing on $\mathbf{\Gamma}^{bd}(t + \Delta t/t')$, from (1.36), we have

$$\mathbf{\Gamma}^{bd}(t + \Delta t/t') = J^{-1}(t + \Delta t/t') \frac{\bar{\lambda}_L^{bd}}{\bar{\lambda}(t + \Delta t/t')} \mathcal{L}^{-1}\left(\frac{\bar{\lambda}(t + \Delta t/t')}{\bar{\lambda}_L^{bd}}\right) \left[\mathbf{B}_{dis}(t + \Delta t/t')\right]_0. \quad (1.59)$$

For $\Delta t \rightarrow 0$, using (1.46), (1.50) and (1.56), (1.59) can be approximated as

$$\mathbf{\Gamma}^{bd}(t + \Delta t/t') \rightarrow J^{-5/3}(t + \Delta t/t) \mathbf{F}(t + \Delta t/t) \mathbf{\Gamma}^{bd}(t/t') \mathbf{F}^T(t + \Delta t/t). \quad (1.60)$$

Substituting (1.60) in (1.58), we get the approximation

$$\begin{aligned} \mathbf{T}_2^{(B1)}(t + \Delta t) &\rightarrow e^{-k_{ns}^{bd} \Delta t} J^{-5/3}(t + \Delta t/t) \mathbf{F}(t + \Delta t/t) \mathbf{T}_2^{(B1)}(t) \mathbf{F}^T(t + \Delta t/t) \\ &+ \int_t^{t+\Delta t} \frac{n_{ns}^{bd}}{n_s^{bd}} k_s^{bd} e^{-k_{ns}^{bd}(t+\Delta t-t')} \mu^{bd} \mathbf{\Gamma}^{bd}(t + \Delta t/t') dt'. \end{aligned} \quad (1.61)$$

Evaluating the second integral in (1.61) using the trapezoidal rule and noting that $\mathbf{\Gamma}(t + \Delta t/t + \Delta t) = \mathbf{0}$, (1.61) can be rewritten as

$$\begin{aligned} \mathbf{T}_2^{(B1)}(t + \Delta t) &\rightarrow e^{-k_{ns}^{bd} \Delta t} J^{-5/3}(t + \Delta t/t) \mathbf{F}(t + \Delta t/t) \mathbf{T}_2^{(B1)}(t) \mathbf{F}^T(t + \Delta t/t) \\ &+ \frac{1}{2} \frac{n_{ns}^{bd}}{n_s^{bd}} k_s^{bd} e^{-k_{ns}^{bd} \Delta t} \mu^{bd} \mathbf{\Gamma}^{bd}(t + \Delta t/t) \Delta t. \end{aligned} \quad (1.62)$$

Finally, the update procedure for $\mathbf{T}^{(B1)}(t + \Delta t)$ under the limit $\Delta t \rightarrow 0$ by combining (1.45) and (1.62) can be summarized as

$$\mathbf{T}^{(B1)}(t + \Delta t) \rightarrow e^{-k_{ns}^{bd}(t+\Delta t)} \mu^{bd} \mathbf{\Gamma}^{bd}(t + \Delta t/0) + \mathbf{T}_2^{(B1)}(t + \Delta t). \quad (1.63)$$

Noting from (1.35) and (1.40) that $\mathbf{T}^{(B2)}(t + \Delta t)$ can be obtained by replacing the superscript “ bd ” with “ el ” in (1.45), the corresponding update procedure under the limit $\Delta t \rightarrow 0$ following (1.62) and (1.63) is given as

$$\mathbf{T}^{(B2)}(t + \Delta t) \rightarrow e^{-k_{ns}^{el}(t+\Delta t)} \mu^{el} \mathbf{\Gamma}^{el}(t + \Delta t/0) + \mathbf{T}_2^{(B2)}(t + \Delta t), \quad (1.64)$$

with

$$\begin{aligned} \mathbf{T}_2^{(B2)}(t + \Delta t) \rightarrow & e^{-k_{ns}^{el}\Delta t} J^{-5/3}(t + \Delta t/t) \mathbf{F}(t + \Delta t/t) \mathbf{T}_2^{(B2)}(t) \mathbf{F}^T(t + \Delta t/t) \\ & + \frac{1}{2} \frac{n_{ns}^{el}}{n_s^{el}} k_s^{el} e^{-k_{ns}^{el}\Delta t} \mu^{el} \mathbf{\Gamma}^{el}(t + \Delta t/t) \Delta t. \end{aligned} \quad (1.65)$$

The standard implementation procedure in ABAQUS/Explicit using VUMAT has been followed. It can be summarized as using the deformation gradient for the current time step provided by ABAQUS at each material point to perform explicit integration of the differential equations involved. This allows for the straightforward calculation of the total Cauchy stress using (1.14), (1.16), (1.63), (1.64), and an update of the state variables. The relevant codes can be accessed at https://github.com/SrivastavaResearchLab/Polymer_Deformation_2023.git. Polylactic acid indenters were modeled as rigid analytical surfaces. Contact between the indenters and PBS blocks was assumed to be frictionless as the interfaces were lubricated during the experiments. The dimensions of the blocks used were 20 ± 2 mm x 20 ± 2 mm x 8 ± 1 mm.

1.5.1 Verification of VUMAT and numerical update procedure for $\mathbf{T}^{(Bi)}$

The convolution-like integrals for $\mathbf{T}^{(B1)}$ and $\mathbf{T}^{(B2)}$ can be directly evaluated in the one-dimensional MATLAB model implementation because the entire deformation history can be stored. Furthermore, the MATLAB implementation was shown to capture the experimental results presented in Section 1.4, and hence, it can be used as a benchmark.

The VUMAT subroutine and $\mathbf{T}^{(Bi)}$ numerical update procedure were verified by comparing results from one-element (unit cube) simple compression simulations with MATLAB predictions and experimental data. Figure 1.11 shows a comparison of the true stress-strain curves for 1 s^{-1} and 4500 s^{-1} strain rates, which show good agreement.

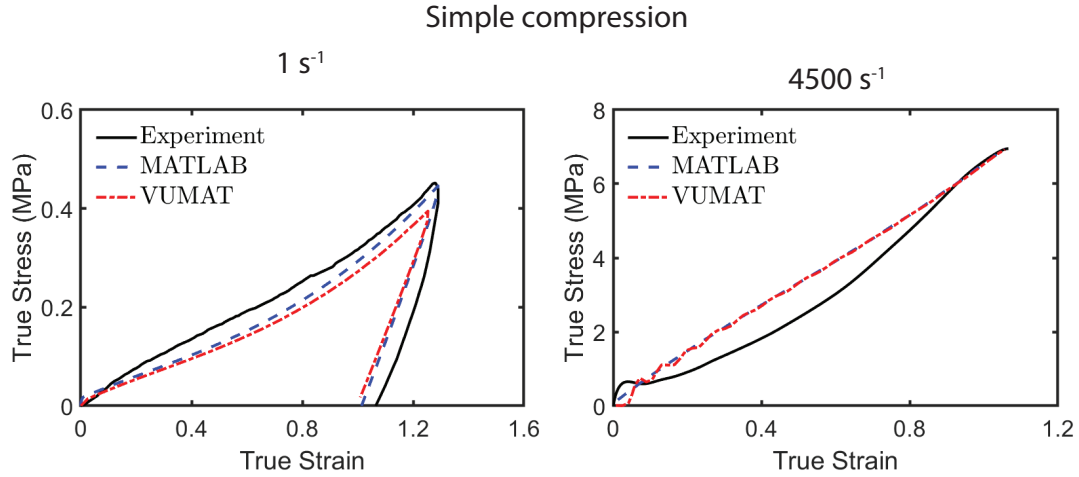


Figure 1.11: **VUMAT and $\mathbf{T}^{(Bi)}$ numerical update procedure verification.** The one-dimensional MATLAB model implementation can store the entire deformation history and directly evaluate the convolution-like integrals for $\mathbf{T}^{(Bi)}$. An approximate numerical update procedure was developed for the model implementation in ABAQUS (Section 1.5). The MATLAB implementation captures the experimental response (Section 1.4) and serves as a reference. Comparison of one-element (unit cube) simple compression simulation (using VUMAT) generated true stress-strain curves with the MATLAB predictions and experimental results for strain rates of 1 s^{-1} and 4500 s^{-1} , showing good agreement.

1.5.2 Conical indenter

The conical indenter was prescribed a linear displacement ramp to 3 mm displacement with a speed of 10 mm/s. Figure 1.12(A) displays the experiment schematic and dimensions in mm of the conical indenter. Considering the symmetries of the experiment about the planes P_1 and P_2 in Figure 1.12(B), only a quarter of the block needs to be simulated with appropriate boundary conditions. The quarter block was meshed using 76960 ABAQUS C3D8R three-dimensional, reduced integration stress elements as shown in Figure 1.12(B). The indenter initially touching the vertex 1 of the representative numbered quarter block in Figure 1.12(B) moves along $-\mathbf{e}_Y$ direction. The bottom surface 2-6-7-3 is fixed while

the surfaces 1-2-3-4 on P_2 and 1-5-6-2 on P_1 are prescribed the boundary conditions $u_X = u_Y^R = u_Z^R = 0$ and $u_Z = u_X^R = u_Y^R = 0$ respectively accounting for the symmetries discussed earlier. u , u^R denote the translational and rotational degrees of freedom of a point about the axis indicated in the subscript. Figure 1.13(A) shows the contours of maximum principal logarithmic strain on the YZ projection of the fully deformed quarter block, showing the complex three-dimensional, inhomogeneous deformations involved. The peak value of strain rate obtained from the simulation is in the order of 10 s^{-1} . Numerical prediction for the indentation force agrees well with the experimental result, as seen in Figure 1.13(B).

1.5.3 Triangular prismatic indenter

Two linear ramp displacement profiles to 3 mm at speeds of (i) 0.7 mm/s and (ii) 2.5 mm/s were prescribed to a triangular prismatic indenter. Figure 1.14(B) shows the schematic for the experiment and dimensions for the indenter in mm. The experiment is symmetric about the planes P_1 and P_2 in Figure 1.14(C), and hence only a quarter of the block needs to be simulated with appropriate boundary conditions. The quarter block was uniformly meshed using 29403 (27 along the thickness) ABAQUS C3D8R three-dimensional, reduced integration stress elements as shown in Figure 1.14(C). The indenter initially touching the edge 1-4 of the representative numbered quarter block in Figure 1.14(C) moves along $-\mathbf{e}_Y$ direction. The bottom surface 2-6-7-3 is fixed while the surfaces 1-2-3-4 on P_2 and 1-5-6-2 on P_1 are prescribed the boundary conditions $u_X = u_Y^R = u_Z^R = 0$ and $u_Z = u_X^R = u_Y^R = 0$, respectively. Figure 1.15(A) shows the contours of maximum principal logarithmic strain on the isometric view and XY projection of the completely deformed full block for the indentation speed of 0.7 mm/s, showing the inhomogeneous deformations. The peak values of strain rates evaluated from the simulations are in the order of 0.1 s^{-1} and 1 s^{-1} for the indenter speeds of 0.7 mm/s and 2.5 mm/s, respectively. Good agreement between the predictions from simulations and experimental results for

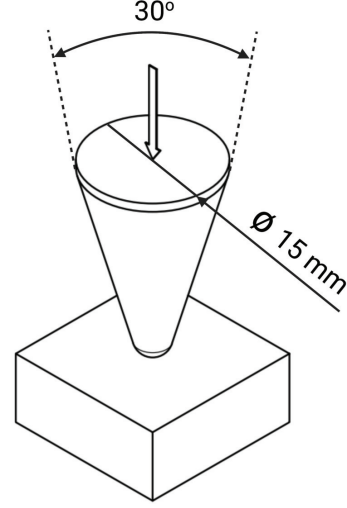
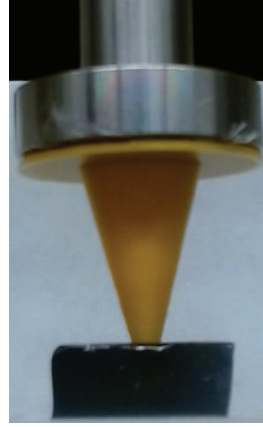
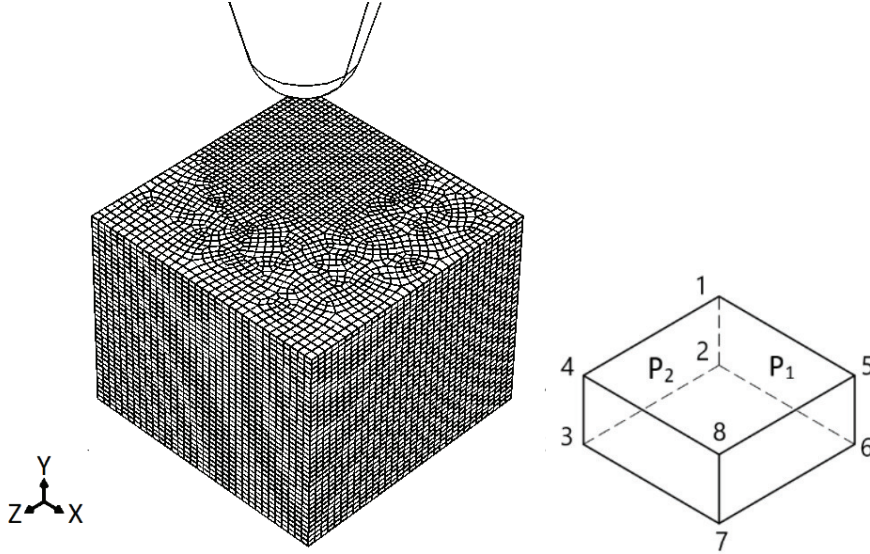
A**B**

Figure 1.12: **Model validation experiment: conical indentation of a PBS block.** (A) Schematic and dimensions in mm for the conical indenter experiment. The PBS block was colored for easy visualization. (B) The experiment is symmetric about the planes denoted by P_1 , and P_2 , and as a result, only a quarter block needs to be considered with appropriate boundary conditions. Finite element mesh used for the quarter block simulation, the conical indenter is modeled as a rigid surface. The bottom surface 2-6-7-3 of the quarter block is fixed while the surfaces 1-2-3-4 on P_2 and 1-5-6-2 on P_1 are prescribed the boundary conditions $u_X = u_Y^R = u_Z^R = 0$ and $u_Z = u_X^R = u_Y^R = 0$, respectively.

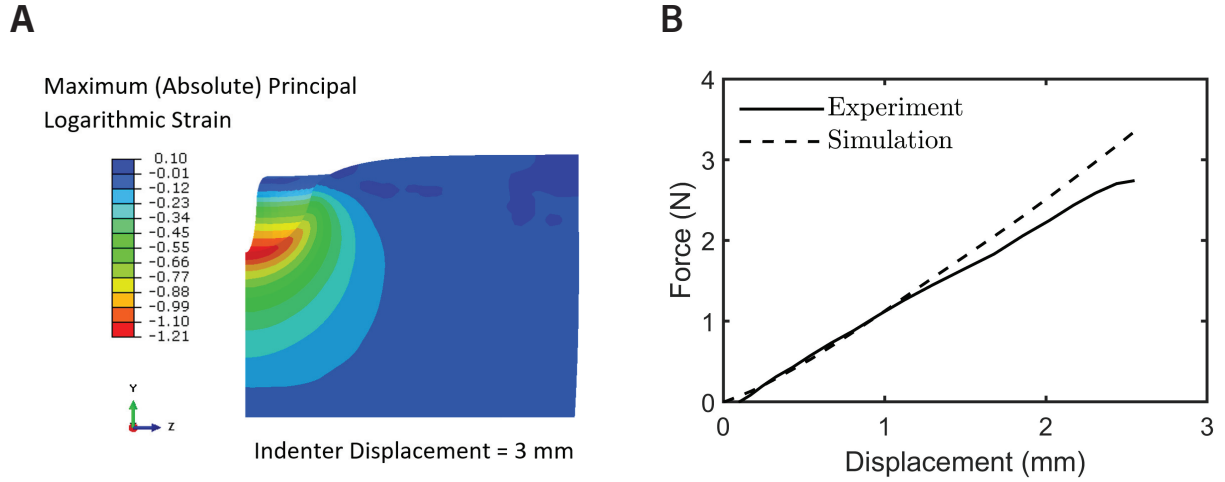


Figure 1.13: **Model predictions for conical indentation of a PBS block.** (A) Contours of maximum principal logarithmic strain on the YZ plane of the deformed quarter PBS block. (B) Comparison of the numerically predicted indentation force with the corresponding experimental result.

the indentation forces can be seen in Figure 1.15(B).

1.5.4 Impact protection provided by PBS through force distribution

A numerical experiment was conducted to understand PBS's impact protection capability. Specifically, an annealed 316 stainless steel cylinder (20 mm x 4 mm) was considered to be impacted by a conical projectile (assumed to be rigid) with an initial kinetic energy of 10 mJ. A thin, 1 mm-thick PBS cylinder (with the same diameter as the steel cylinder) was then bonded to the steel cylinder. The Mises stress contours (at zero projectile velocity) in the steel cylinder with and without the thin PBS cylinder are compared in Figure 1.16. The projectile was modeled as a rigid analytical surface in ABAQUS. The mating surfaces of the PBS and steel cylinders were constrained completely. The maximum Mises stress in the steel cylinder was significantly reduced when the thin PBS cylinder was bonded (478 MPa $\rightarrow$ 10 MPa). The stress contours in the steel were less localized. This can be attributed to PBS's ability to distribute force upon impact. The significantly rate-stiffening behavior of PBS implies that a rapidly applied local loading is

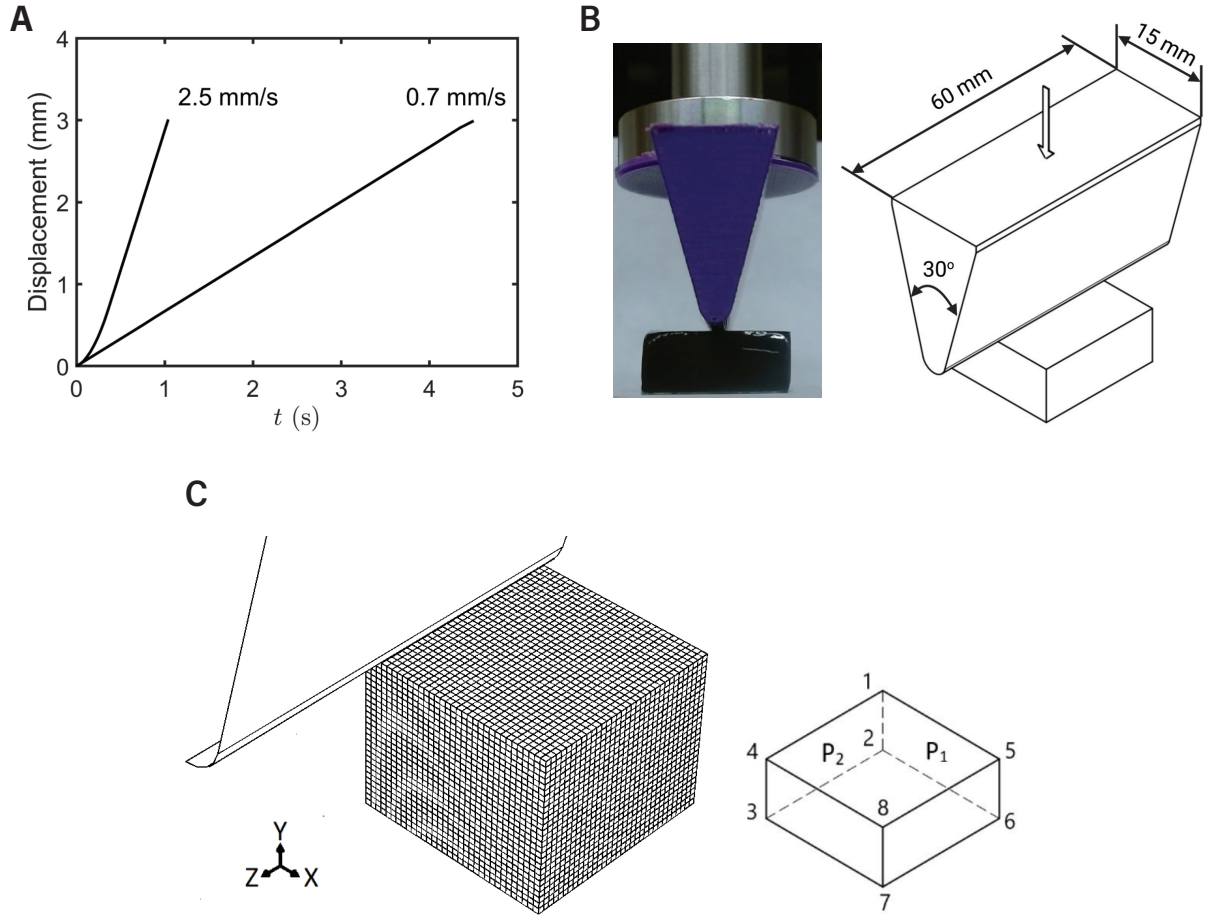


Figure 1.14: **Model validation experiment: prismatic indentation of a PBS block.** (A) Linear ramp displacement profiles of the prismatic indenter. (B) Schematic for the experiment and dimensions in mm for the triangular prismatic indenter. The PBS block was colored for easy visualization. (C) The experiment is symmetric about the planes denoted by P_1 , and P_2 , and as a result, only a quarter block needs to be considered with appropriate boundary conditions. Finite element mesh used for the quarter block simulations. The triangular prismatic indenter was modeled as a rigid surface. The bottom surface 2-6-7-3 of the quarter block is fixed while the surfaces 1-2-3-4 on P_2 and 1-5-6-2 on P_1 are prescribed the boundary conditions $u_X = u_Y = u_Z = 0$ and $u_X = u_Y = u_Z = 0$ respectively.

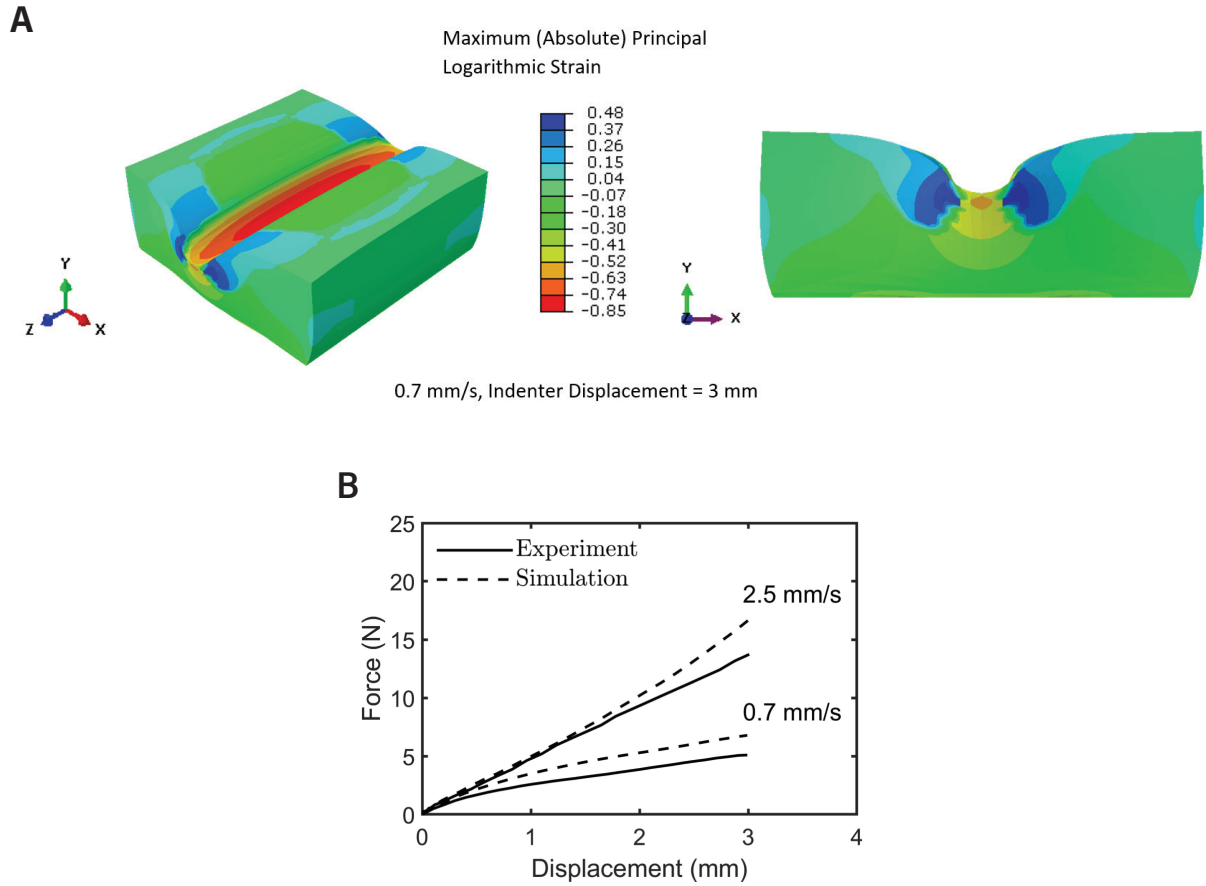


Figure 1.15: **Model predictions for prismatic indentation of a PBS block.** (A) Contours of maximum principal logarithmic strain in the isometric view and XY projection of the deformed PBS block for the indentation speed of 0.7 mm/s. (B) Comparison of the numerically predicted indentation forces for triangular prismatic indenter speeds of 0.7 mm/s and 2.5 mm/s with corresponding experimental results. The model and simulation predicted results are very close to the experimental results.

distributed over a larger region, thereby providing impact protection.

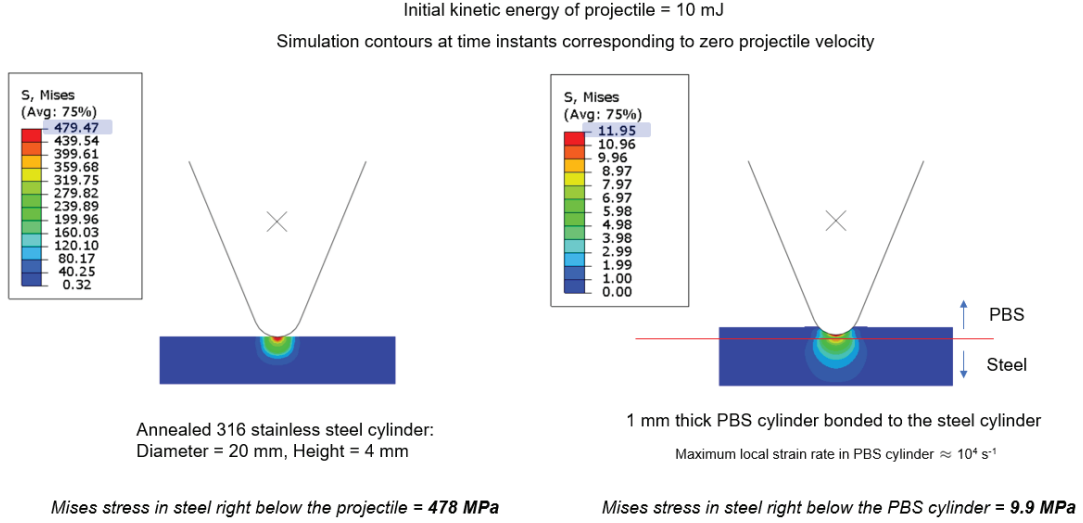


Figure 1.16: PBS: force distribution upon impact shown through a numerical experiment. Mises stress contour comparison in annealed 316 stainless steel cylinder with and without thin PBS coating impacted by a rigid projectile. The steel cylinder had dimensions of 20 mm $\times$ 4 mm, and the thin cylindrical PBS coating had dimensions of 20 mm $\times$ 1 mm. The projectile had an initial kinetic energy of 10 mJ, and the stress contours shown correspond to the condition in which the projectile velocity was zero. The maximum Mises stress in the steel reduced from 478 MPa to 10 MPa with the thin PBS coating. Furthermore, the stress contours were less localized. PBS's rate-stiffening behavior enables force distribution and hence protection upon local impact.

As a qualitative experimental example, eggs were dropped with and without a thin PBS coating from a height of 30 cm. Figure 1.17 shows that the egg with the thin PBS coating did not crack upon impact and rebounded, while the one without it cracked.

To further understand the impact protection through force distribution, this experiment was modeled by considering only the egg shell and the following assumptions: (i) the egg shell geometry as a thin hollow prolate spheroid (an ellipse rotated about its major axis) with thickness = 0.3 mm and outer semi major and minor axes lengths of 30 mm and 20 mm, respectively, (ii) the egg shell as a linear elastic isotropic material with $E = 20$ GPa, $\nu = 0.3$ (Kemps et al., 2004) and $\rho = 2000 \text{ kg/m}^3$ (Carter, 1968), (iii) PBS coating thickness of 1 mm, and (iv) the base on which the egg impacts as a rigid analytical surface. An axisymmetric FE model was used, as shown in Figure 1.18, where the dropped parts

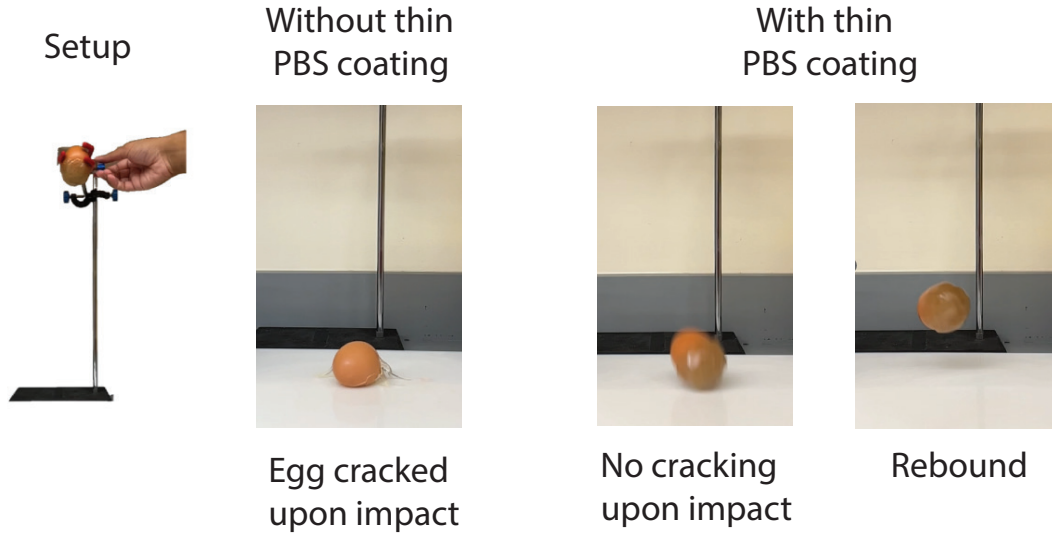


Figure 1.17: **Impact protection of an egg with a thin PBS coating.** To demonstrate PBS's impact protection capability, eggs with and without a thin PBS coating were dropped from a height of 30 cm. The egg with a thin PBS coating did not crack upon impact and rebounded. The uncoated specimen cracked upon impact.

were assigned an initial velocity based on the drop height. We also considered a coating of PDMS [Sylgard 184] with the same dimensions and modeled as a Mooney-Rivlin solid (Gao et al., 2024) to compare PBS's impact mitigation with an elastomer. The mating surfaces of the egg shell and the coating were constrained completely. Figure 1.18 shows the Mises stress contours (MPa) on the egg shell near the impact location for the uncoated, PBS-coated, and PDMS-coated cases at time instants corresponding to zero kinetic energy. Upon impact, the thin PBS coating distributed the locally applied impact force over a larger region of the egg shell, thereby preventing cracking. A similar observation is also made for the PDMS coating numerically. While elastomers can mitigate impact similar to PBS, the higher malleability and conformity, significantly rate-dependent response, as well as autonomous self-healing capabilities of PBS, give it an advantage for practical applications.

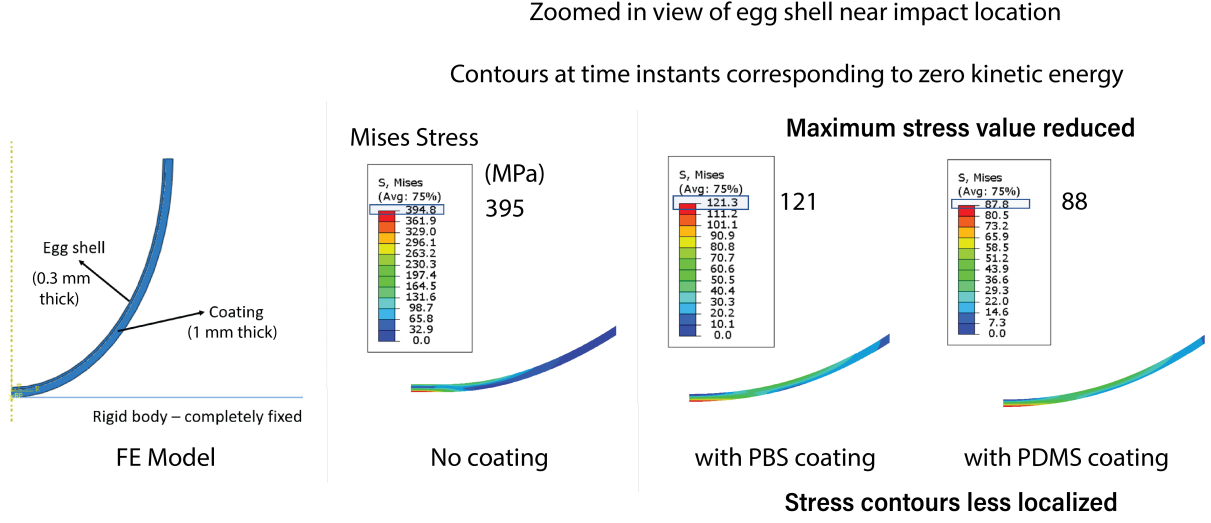


Figure 1.18: **Modeling of the dropped uncoated and coated eggs: impact protection through force distribution.** The qualitative egg drop experiment (with and without PBS coating) was modeled to further understand the impact protection through force distribution. Schematic of the axisymmetric FE model. A PDMS [Sylgard 184] coating with the same dimensions was also considered to compare PBS’s impact mitigation with an elastomer. Mises stress contours (MPa) on the egg shell near the impact location for the three cases at time instants corresponding to zero kinetic energy. The thin PBS and PDMS coatings distributed the locally applied impact force over a larger region of the egg shell, preventing cracking as observed experimentally for the PBS case. While elastomers can mitigate impact similar to PBS, the higher malleability and conformity, significantly rate-dependent response, as well as autonomous self-healing capabilities of PBS, give it an advantage for practical applications.

1.6 Conclusion

We have conducted experiments and developed a finite deformation elastic-viscoplastic constitutive model for polyborosiloxane (PBS), an important rate stiffening soft polymer with dynamic, reversible crosslinks and impact protection applications for quasi-static (10^{-3} s^{-1}) to high (10^3 s^{-1}) strain rates. Using the macroscopic observations from our experiments and the model framework, we suggest two crosslink mechanisms in PBS and their distinct macroscopic relaxation timescales (τ). The dynamic network-forming reversible crosslink mechanisms are boron-oxygen coordinate-bond crosslinks ($\tau \approx 3 \text{ s}$) and temporary entanglement lockups at high strain rates acting as crosslinks ($\tau \approx 0.5 \text{ ms}$). The microstructure physics-based continuum model suggests possible microstructural mechanisms and provides estimations of the subchain lengths between crosslinks from

the macroscopic experiments. We estimate subchains between B:O coordinate-bond dynamic crosslinks to have root mean squared end-to-end distance $\overrightarrow{r}_s^{bd} \approx 10 \text{ nm}$, and the subchains between entanglement lockups to have $\overrightarrow{r}_s^{el} \approx 1.8 \text{ nm}$ in PBS under the undeformed state. The average subchain contour lengths are estimated to be $L_s^{bd} \approx 378 \text{ nm}$ for subchains between B:O coordinate-bond dynamic crosslinks, and $L_s^{el} \approx 12 \text{ nm}$ for subchains between entanglement lockups. To analyze real structures, it is essential to perform new material model-based computational simulations for three-dimensional geometries. We have numerically implemented the model in the finite element software ABAQUS and have outlined a numerical update procedure to evaluate the convolution-like time integrals arising from dynamic crosslink kinetics. The approximate predictive capabilities of the constitutive model and its numerical implementation were verified by comparing the simulation results with results from validation experiments involving inhomogeneous deformations. The proposed model and its theoretical framework should extend to other soft polymers and polymer gels with dynamic, reversible crosslinks that are microstructurally similar to PBS.

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CHAPTER 2

Modeling fracture of soft polymers

2.1 Introduction

Soft polymers are a broad class of polymers with chain networks formed through permanent or dynamic chemical or mechanical crosslinks, leading to elastic modulus in the range of a few kilopascals to tens of megapascals. This class encompasses elastomers, viscoelastic and elastic-viscoplastic polymers, polymer gels,¹ and biological tissues. Their physical forms can range from very viscous, slowly flowing, soft solids to elastomeric, rubber-like materials.

Current fracture toughness measures based on energy release rate or crack tip stress intensity are non-unique when evaluated for rate-dependent soft materials. They vary with the loading profile and specimen geometry under consideration (Song et al., 2021; Long and Hui, 2016; Long et al., 2021; Yin et al., 2021). It will be of value to have a general measure in a continuum sense that is not geometry and rate-dependent.

Significant experimental studies have focused on the fracture of elastomers and polymer gels (Lake et al., 1967a; Lake, 1995; Chen et al., 2017; Creton, 2017; Long et al., 2021;

¹The polymer gels here are restricted to those where loading timescales are significantly faster than the solvent diffusion timescale.

Kim et al., 2021; Tang et al., 2017; Shrivastava and Gundiah, 2023). The rate-dependent fracture of polymers has also been of significant interest, and various relevant experimental studies have been published (Rosakis and Ravichandran, 2000; Shukla et al., 2010; Mac Donald and Ravichandran, 2020; LeBlanc et al., 2020, 2018; Srivastava et al., 2000, 2002; Subhash et al., 2006; Subhash and Liu, 2009; Zheng et al., 2022; Malhotra et al., 2021; Brown et al., 2002; Malito et al., 2019; Burla et al., 2020; Liu et al., 2023; Cox et al., 2005; Guzek et al., 1997; Xu and Wang, 2006). The rate-dependent fracture of polymers that contain dynamic crosslinks has been recently experimentally studied (Luo et al., 2014; Liu et al., 2019b; Song et al., 2021; Zhao et al., 2022; Chen and Ravi-Chandar, 2023; Bai et al., 2018; Liu et al., 2021). Mayumi and co-workers (Mayumi et al., 2016) conducted notched tension tests on Polyvinyl alcohol gels and noticed a significant decrease in failure stretch with increasing loading rates. Shen and co-workers (Shen et al., 2021) performed notched pure shear tests on EPS25 vitrimer with multi-step loading profiles. The authors observed loading rate-dependent flow-fracture (crack blunting-advancing) transitions. Qi and co-workers (Qi et al., 2024) presented a method to map the dissipation field during crack propagation in viscous soft polymers and enable individual evaluation of the intrinsic and dissipative components of fracture toughness. The authors evaluated the stress and, consequently, the total energy density provided by mechanical work at each material point using a non-linear constitutive model and deformation field measurements. The dissipation field was then obtained by subtracting the free energy density field evaluated using the constitutive model from the total energy density field.

2.1.1 An overview of damage and fracture modeling approaches

At this stage, it is prudent to present a broad overview of damage and fracture modeling approaches utilizing the finite element method (FEM).

FEM without gradient-damage

A simple method for modeling fracture in FEM involves employing a local damage variable without non-local gradient terms. An element's stiffness is reduced to zero once the damage variable at an integration point/s hits a critical threshold. However, this approach produces solutions that are highly mesh-dependent. The predicted crack propagation path can be nonphysical and inaccurate if the mesh is not aligned with the actual path, which can be difficult to guess beforehand (Song et al., 2008; Pelfrene et al., 2016).

FEM with gradient-damage

The non-local gradient-damage term in mesh-based numerical modeling is needed to obtain mesh-independent solutions. For numerical reasons, FEM with gradient-damage considers a “smeared crack approach”, in which the mathematical framework for damage depends on a damage field variable d along with its gradient ∇d . A length scale parameter l is also used over which d varies between zero and one (Miehe et al., 2010b). FEM with gradient-damage can provide mesh-independent results, provided that the typical element size is less than $0.2l$ (Narayan and Anand, 2019). Numerous works in the literature have used FEM with gradient-damage to model damage initiation, growth, and complete failure in a broad range of materials (Miehe et al., 2010b; Narayan and Anand, 2019, 2021; Kuhl et al., 2000; Konica and Sain, 2021; Yang et al., 2024; Hu et al., 2020; Behera et al., 2022; Du et al., 2024).

At a high level, the modeling frameworks are based on either the (i) *variational method*

- minimization of a non-linear energy function (Bourdin et al., 2000) or the (ii) *method of virtual power* (Narayan and Anand, 2021). A further two-level sub-categorization can be done based on how damage initiation is modeled:

Without explicit damage initiation criterion

Variational phase field models predict crack nucleation through the minimization of a non-linear energy function without explicit criteria. Their drawback of being unable to capture experimentally observed failure surfaces has been noted in (Lopez-Pamies et al., 2025).

With explicit damage initiation criterion

A significant number of recent approaches to damage and fracture modeling fall under this category. The two major ways in which damage initiation is predicted are (a) using critical value/s of free energy density (Mao et al., 2017; Konale and Srivastava, 2025) and (b) using a stress-based failure/material strength surface (Kumar and Lopez-Pamies, 2020; Lopez-Pamies et al., 2025). Although both approaches are fundamentally equivalent, the free energy density approach provides a distinct advantage: it can yield unique critical values under a given loading mode for rate-dependent materials, to which the approach's applications are currently restricted. An experimentally informed free energy density-based failure surface can also be developed, similar to its stress-based counterpart, for broader applications to arbitrary loading modes.²

Perspective

There have been informative discussions in the literature on the limitations of certain damage and fracture computation methods and models (Lopez-Pamies et al., 2025; Nguyen et al., 2020; Bažant et al., 2022). Common themes include misinterpretation of the absence of an explicit damage initiation criterion and neglect of important but overlooked phenom-

²Section 2.10 presents the findings from an attempt to develop generally applicable damage initiation criteria for brittle materials, of which soft polymers are an example.

ena (owing to their often negligible effects), such as the effect of crack-parallel stresses. Although these points are partially valid, when assessing overall numerical predictive performance, it is important to distinguish the limitations of constitutive damage models from those of computational methods. Once a computational method is chosen, the constitutive model governs the predictive capabilities. A constitutive model can be made arbitrarily complex to capture an arbitrary number of physical phenomena. It is always of interest to have an all-encompassing constitutive model, but the mathematical complexity also has to be kept tractable.

Recent modeling efforts on the fracture of soft polymers, which are limited only to elastomers based on the gradient-damage framework, can be broadly separated into three approaches. First, considers the variational approach without explicit damage initiation criteria (Bourdin et al., 2000, 2008). The second approach for elastomers directly considers the underlying Euler-Lagrange equations and uses independent stress-based criteria for damage nucleation from the bulk (Kumar and Lopez-Pamies, 2020; Kumar et al., 2020; Lopez-Pamies et al., 2025). It uses the Griffith energy competition as a descriptor of nucleation from a large pre-existing crack. The third approach is based on the method of virtual power. In this approach, the undamaged free energy density function, which is the driving force for the initiation and evolution of damage, is formulated in two ways. One considers it to be entirely entropic following the ideal chain assumption (Miehe and Schänzel, 2014; Raina and Miehe, 2016; Gültekin et al., 2016; Wu et al., 2016). The other incorporates an energetic contribution to the free energy density function due to backbone bond or crosslink stretching (whichever is weaker), along with the conventional entropic part (Mao et al., 2017; Talamini et al., 2018; Mao and Anand, 2018a,b; Li and Bouklas, 2020). This is motivated by the scaling law for elastomers proposed by Lake and Thomas (Lake et al., 1967b) relating the Griffith energy release rate to the binding energy between monomer units (or of crosslinks (Mao and Anand, 2018a)) among other microscopic parameters. However, all these models are limited to elastomers only, while

the focus of this chapter is to develop a general, unified damage and fracture model for rate-dependent, viscous polymers that is also applicable to elastomers.

The modeling of the rate-dependent fracture in dynamically crosslinked soft polymers and viscous polymers is limited and has been restricted to crack tip stress field analyses (Guo et al., 2018, 2019; Hui et al., 2019; Liu et al., 2019b) or crack tip driving force-based approach (Shen and Vernerey, 2020; Shen et al., 2021; Ciavarella, 2024) for specific pre-cracked geometries. The crack tip stress field asymptotic analyses use the deformation and stress fields near the crack tip to predict fracture. The crack tip driving force approach relates the crack propagation velocity to the energy release rate. Models with the ability to predict crack nucleation, propagation, and coalescence in arbitrary, complex geometries are lacking and are necessary for solving a general class of problems. The gradient-damage approach, as discussed earlier, is a suitable choice for this purpose.

Critical gaps in modeling and predicting damage initiation, growth, and fracture of soft polymers are (i) the establishment of a unique physical parameter to quantify fracture resistance of soft polymers and (ii) the development of a unified model to predict damage initiation and growth which is generally applicable for soft polymers that range from viscous polymers to elastomers and is independent of loading history and specimen geometry.

For rate-dependent soft polymers, we considered polyborosiloxane (PBS). PBS is a dynamically crosslinked, rate-stiffening soft polymer that is highly viscous at long timescales with a significantly rate-dependent mechanical response (Li et al., 2014; Tang et al., 2016; Zhao et al., 2021, 2020a; Kim et al., 2022). PBS flows under its weight over long timescales (minutes) and exhibits a rubber-like response with a significant shape recovery upon deformation when subjected to high loading rates. The rate-dependent fracture response of PBS has not been experimentally studied, and corresponding models have not been developed, which are critical given viscous polymers such as PBS's broad applications, which include self-healing sensing and actuation (Narumi et al., 2019; Wang

et al., 2018; Liu et al., 2019a) and flexible impact mitigation (Tu et al., 2023; Myronidis et al., 2022).

Below are the main contributions of this chapter:

1. We have proposed and shown that a critical value of stress work W_{cr} can be an approximately unique measure of fracture resistance for a certain class of soft polymers.³ It quantifies the total energy dissipation per unit volume for the complete failure of an arbitrary material point under a loading mode when it is subjected to loading rates sufficient to cause damage initiation. Complete failure refers to the inability to support any mechanical stress. W_{cr} can be numerically evaluated for general deformations, of which the homogeneous deformations are special cases, where it can be directly obtained from the experiments.
2. We have developed a model to predict damage initiation, damage growth, and complete failure in soft polymers using a multi-mechanism generalized gradient-damage framework based on the method of virtual power, where the material strength is accounted for.⁴ The free energy density contribution from network resistance is formulated considering crosslink relaxation and formation. As motivated by our previous work (Konale et al., 2023), the number of chain segments that contribute to the free energy and their individual stretches vary with time, as governed by the underlying crosslink kinetics.⁵ The damage initiation criterion ψ_{cr}^+ (*energetic contribution to W_{cr}*) in the model can be evaluated using a one-dimensional numerical implementation of a deformation-only constitutive model

³ W_{cr} is a general definition in the continuum sense. The work of fracture can be considered as a special case of W_{cr} . For a uniaxial tensile loading of an unnotched specimen, W_{cr} is equivalent to the standard modulus of toughness (Roylance, 2001; Brostow et al., 2015) or work of fracture [defined as the area under the engineering stress-strain curve in an unnotched uniaxial tension test till failure] (Chen et al., 2017; Wang et al., 2019; Yang et al., 2019).

⁴This method uses explicit criteria for damage initiation in contrast to the variational phase field method, which minimizes an energy function to predict damage initiation. The limitations of the variational phase field method have recently been discussed in (Lopez-Pamies et al., 2025).

⁵This form of free energy reflects microstructural physics and is thermodynamically consistent. The free energy density functions with direct rate-dependence proposed in some of the literature are thermodynamically inconsistent.

and homogeneous deformation experiment (e.g., simple tension test) results.

3. We validated the predictive capabilities of the fracture model for three elastomers (EPDM, SBR, NR), a viscous soft polymer (EPS25 vitrimer), and PBS.

2.2 Fracture in soft polymers

Notches and cracks in soft polymers undergo a significant amount of blunting before fracture initiation (Miehe and Schänzel, 2014; Luo et al., 2014). Hence, geometric non-linearity due to the large deformation corresponding to blunting has to be accounted for along with material non-linearity. Figure 2.1 depicts key observations during the deformation and fracture of PBS. To fabricate PBS, hydroxy-terminated Polydimethylsiloxane (PDMS, Sigma Aldrich 481963, 750 cSt) and Boric Acid (BA, Sigma Aldrich B0394, ACS reagent, $\geq 99.5\%$) were mixed in the ratio 160:1 by weight. The mixture was stirred and then heated at 120°C for 72 hrs to obtain PBS (Konale et al., 2023). The mass density of PBS was measured to be 1100 kg m⁻³. A notched PBS specimen in Figure 2.1(A) was stretched at slow (6 mm/s) and fast (60 mm/s) rates. All tension tests on PBS in this chapter were conducted using the UniVert Mechanical Test System (CellScale) and at 23°C. Three replicates were performed for each experimental result reported. We applied 3D-printed Polylactic acid (PLA) custom fixtures to avoid creeping of PBS under the clamping pressure of conventional spring-loaded grips. In the custom fixtures shown in Figure 2.1(A), parts of the specimen outside the gauge length are glued to the fixture's interior faces using a cyanoacrylate adhesive.

PBS's rate-dependent fracture response is highlighted in Figure 2.1(B) through a transition from high extensibility at a slow deformation rate without fracture to fracture with low extensibility at a fast loading rate. Figure 2.1(C) shows the specimen geometry evolution with stretching load through damage evolution and complete failure at the 60 mm/s loading rate. The high extent of notch blunting before notch propagation can be

observed in Figure 2.1(C). The presence of viscous effects in soft polymers results in a trumpet-like early fracture profile (formation of a second local notch ahead of the original blunted notch) (de Gennes, 1996; Hui et al., 2022). Figure 2.1(D) shows deformation profiles before complete fracture in notched PBS and notched Polydimethylsiloxane (PDMS) elastomer (Lee et al., 2024) specimens. Compared to the trumpet profile for PBS, the elastomer exhibits a single parabola-like fracture profile. The single parabola-like fracture profile for elastomers has been well documented in literature (Qi et al., 2019; Shrivastava et al., 2023). A fracture model for soft polymers, such as the one proposed in this chapter, in addition to accurately predicting the failure loads, should predict these significantly varying physical phenomena observed in experiments.

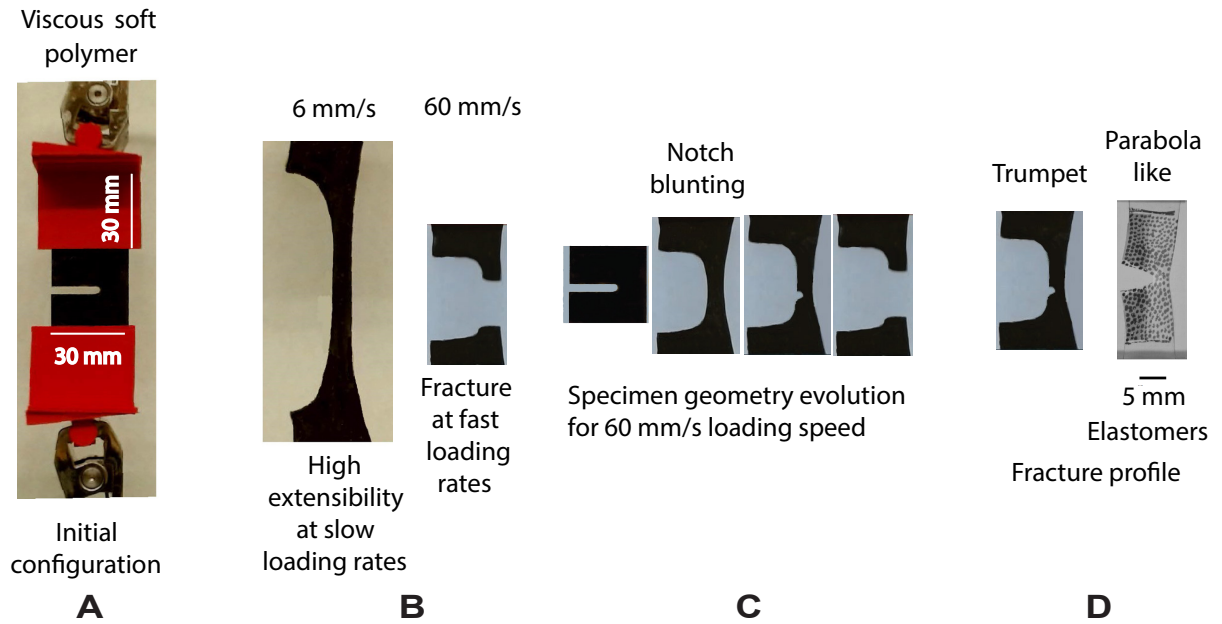


Figure 2.1: **Fracture phenomenon in soft polymers.** (A) The geometry of a notched PBS specimen and the experiment schematic. The thickness of the specimen is 3.2 mm. PBS was colored for visualization. (B) The high extensibility of the notched PBS specimen when stretched at a slow loading speed (6 mm/s). The fracture of the specimen at a fast loading speed (60 mm/s). (C) The specimen geometry evolution till complete failure for the 60 mm/s loading speed. Significant notch blunting occurs before notch propagation. (D) The trumpet fracture profile (formation of a second local notch ahead of the original blunted notch) in the rapidly stretched notched PBS specimen. A single parabola-like fracture profile experimentally observed in PDMS elastomer (image from J. Lee and co-workers (2024) (Lee et al., 2024) reproduced with Creative Commons [CC BY-NC-ND 4.0 - <https://creativecommons.org/licenses/by-nc-nd/4.0/>] copyright license).

2.3 A fracture resistance measure for soft polymers

In permanently crosslinked soft polymers that fracture by crosslink failure, i.e., polymer chain backbone bonds are stronger than crosslinks, it can be assumed that all crosslinks at a material point fail simultaneously (Mao and Anand, 2018a). The weak crosslinks in dynamically crosslinked soft polymers are constantly breaking and reforming, even in an undeformed state. This dissociation rate is amplified by stretching of the corresponding subchains with respect to the stress-free subchain formation configuration (Song et al., 2021; Venkata et al., 2021). Subchains denote parts of chain molecules between two crosslinks connected to the elastically active network. All subchains in a volume element in a dynamically crosslinked soft polymer can be broken simultaneously only when they are stretched equally to the level corresponding to stretch-induced dissociation. This scenario, however, can occur only at infinitely fast rates, which is not practically feasible. At adequate fast rates, a sufficient number of subchains surviving from close to time $t = 0$ will dissociate from the elastically active network, creating voids. These voids will coalesce and lead to the complete failure of a volume element (Shen et al., 2021). A measure of fracture resistance here can thus be thought of as a critical value of input energy density beyond which network stretching and subsequent damage in the form of stress-accelerated subchain dissociation becomes more dominant than network reconfiguration.

Stress work defined as

$$W(X_i, t) = \int_0^t \frac{1}{2} S_{ij} \dot{C}_{ij} dt, \quad (2.1)$$

quantifies the total energy density per unit referential volume supplied to a material point X_i due to the work done by external macroscopic forces in the scenarios where kinetic energy is negligible. S_{ij} , $C_{ij} = F_{ki}F_{kj}$ are the second Piola-Kirchhoff stress and the right Cauchy-Green deformation tensors, respectively, with F_{ij} being the deformation gradient

tensor. W_{cr} is defined as the value of W , i.e., the total energy per unit referential volume that needs to be supplied for the complete failure of an arbitrary material point subjected to sufficiently fast loading rates for damage initiation. Irrespective of the strain rate value under consideration, the same number of bonds have to be broken for the complete failure of a material point. A common loading mode-based response asymmetry observed in soft polymers is tension-compression asymmetry (Drozdov and deC. Christiansen, 2020; Huang et al., 2016). W_{cr} 's value can then be different in tension and compression for these soft polymers. However, for a given loading mode (i.e., tension, compression, etc.), the hypothesis is that W_{cr} is a unique value with respect to loading rates and specimen geometries for a certain class of soft polymers. This hypothesis's motivation comes from the earlier argument, along with the following presented experimental observations. It can be understood mathematically on a high level by considering a material represented by a Maxwell element (see Figure 2.2 for the schematic).



Figure 2.2: **Maxwell element schematic.**

At slow strain rates, the spring (representing the load-bearing bonds) is not stretched significantly as the dashpot sustains most of the deformation. Consider the reasonable assumption of bond energies being independent of strain rate. Then, with increasing strain rate, the springs (bonds) start stretching significantly, and there will be a specific point (quantified by W_{cr}) at which complete failure of the spring (bonds) occurs. Although these arguments may not hold for the Kelvin-Voigt (KV) element, it can be noted that the KV element is not a good representation of most materials. However, the above arguments can be easily extended to the more applicable rheological material idealizations of Standard

Linear Solid (SLS) or multiple Maxwell elements in parallel (generalized Maxwell). This can be attributed to the Maxwell elements (or the spring in SLS) in parallel, deforming independently. Hence, each element and the corresponding mechanism of deformation will have its own unique critical energy density value quantifying its damage. W_{cr} can then be interpreted as the sum of these critical energy density values for all mechanisms.

It can also be noted here that soft polymers can exhibit near incompressibility (Barney et al., 2022; Gurvich and Fleischman, 2003; Kadapa and Hossain, 2022). For the case of pure hydrostatic tension, as incompressibility is approached, the deformation until fracture will be small. But W_{cr} will not be zero under this loading as the ground-state bulk modulus (K) of a nearly incompressible polymer is quite large, resulting in finite W_{cr} . For the volumetric part of free energy density (ψ), if the simple functional form of $\frac{1}{2}K (J - 1)^2$ is considered, then under purely dilatational deformation, $\psi = \frac{1}{2}K (J - 1)^2$, with $J = \det F_{ij}$. For a purely elastic polymer, $W = \psi$. As an example, if the ground state Young's modulus and Poisson's ratio of such a nearly incompressible polymer are taken as 2 MPa and 0.4999, respectively, K will be 3.3 GPa. Further, if J_{cr} under purely tensile dilatational deformation is arbitrarily considered to have a very low value of 1.001, then we get $W_{cr} = 1.6$ kPa, *which is a significant non-zero value*. This can also be attributed to the non-zero energy density needed for the dissociation of a finite number of bonds, leading to the failure of a material point. W_{cr} can be evaluated using relatively simple experiments involving homogeneous deformation and without constitutive models. The unit of W_{cr} is J/m³ in contrast to J/m² for the conventional fracture toughness measures (Yin et al., 2021; Mai and Powell, 1991). The standard energy release rate (Li et al., 1985; Budiansky and Rice, 1973) and the essential work of fracture (Mai and Powell, 1991) are the measures of loss of energy per unit area of crack growth. They usually depend on the constraints created at the crack tip by the amount of material, i.e., specimen dimensions considered (Kang et al., 2005; Zhu and Joyce, 2012; Yin et al., 2021). On the contrary, W_{cr} considers the energy density associated with the complete failure of an arbitrary

material point. Hence, the value of W_{cr} should be independent of specimen dimensions. For very soft materials, feasible experiments often involve inhomogeneous deformations (Duncan et al., 2020; Montanari et al., 2023). W_{cr} 's general definition allows its evaluation in arbitrary specimen geometries, contrary to conventional fracture toughness measures, which require specific geometries.

We now utilize experimental stress-strain curves for a few soft polymers to demonstrate the negligible dependence of W_{cr} on strain rate. Zhao and co-workers (Zhao et al., 2020b) synthesized a rate-dependent soft polymer with dynamic crosslinks [TFPM-PDMS-25000: 4-[tris(4-formylphenyl)methyl]benzaldehyde (TFPM) used as a tetratopic linker to crosslink PDMS]. The mechanical response of TFPM-PDMS-25000 under uniaxial tensile loading at different stretching speeds was also characterized in the paper. Results are shown in Figure 2.3(A). We calculated W_{cr} for each loading rate by evaluating the area under the corresponding engineering stress-strain curve. The variation of W_{cr} with respect to strain rate is shown in Figure 2.3(B). W_{cr} is approximately constant over one decade of strain rate considered. Polyurethane's (2% w/w of Polyethylene glycol with respect to Palm-based polyol content) rate-dependent mechanical response reported by Somarathna and co-workers (Somarathna et al., 2020) is shown in Figure 2.3(C) with uniaxial tensile engineering stress-strain curves over a range of strain rates. We calculated W_{cr} for each strain rate from the corresponding engineering stress-strain curve. We show that W_{cr} is roughly constant over two decades of strain rate through Figure 2.3(D). Over this range of strain rate, energy release rate, and crack tip field measures for rate-dependent soft polymers can vary by an order of magnitude or more (Song et al., 2021; Long and Hui, 2016). We also show a negligible variation in W_{cr} value for different loading modes by evaluating W_{cr} from the stress-strain data for Ecoflex elastomer (material has no loading mode-based response asymmetries) reported by Ahmad and co-workers (Ahmad et al., 2019). Results of homogeneous uniaxial tension (UX), pure shear (PS), and equibiaxial tension (EB) tests performed by the authors at a constant engineering strain rate of

0.3 s^{-1} are shown in Figure 2.3(E). We evaluated W_{cr} for each loading mode from the corresponding engineering stress-strain curves. W_{cr} is approximately constant for the three loading modes considered: 2 MPa (UX), 1.6 MPa (PS), and 1.7 MPa (EB), as shown in Figure 2.3(F). These examples further support our hypothesis that, within a reasonable approximation, W_{cr} can be used as a scalar measure of fracture resistance for a certain class of soft polymers.

2.4 Multi-mechanism fracture modeling framework

We follow the Kroner-Lee (Kröner, 1959; Lee, 1969; Kothari et al., 2019; Niu et al., 2023) multiplicative decomposition of the deformation gradient tensor F_{ij} into elastic and plastic parts F_{ij}^e and F_{ij}^p , respectively and the parallel multi-mechanism generalization of the multiplicative decomposition used in elastic-viscoplastic and viscoelastic constitutive theories for polymers (Boyce et al., 1988, 1989; Vogel et al., 2014; de Rooij and Kuhl, 2016; Bergström and Boyce, 1998; Anand et al., 2009; Srivastava et al., 2010b). We consider an isothermal framework for materials that can be idealized as isotropic. For the general framework, M parallel mechanisms of deformation are considered. Damage variables $d^{(q)}(X_i, t) \in [0, 1]$, $q \in [1, M]$ corresponding to each micro-mechanism are introduced to model damage and subsequent fracture. We set the general criteria to be $d^{(q)}(X_i, t) = 0$ for undamaged and $d^{(q)}(X_i, t) = 1$ for completely damaged mechanism q (when a mechanism q reaches complete damage, it has no contribution to the total stress) at a material point X_i . W_{cr} in a multi-mechanism sense can be interpreted as the value of W when the damage fields for all mechanisms ($d^{(1)}, d^{(2)}, \dots, d^{(M)}$) reach the value of 1 at a material point subjected to loading rates sufficiently fast to cause damage initiation in each mechanism. The method of virtual power is now used to derive the macroscopic and microscopic force balances.

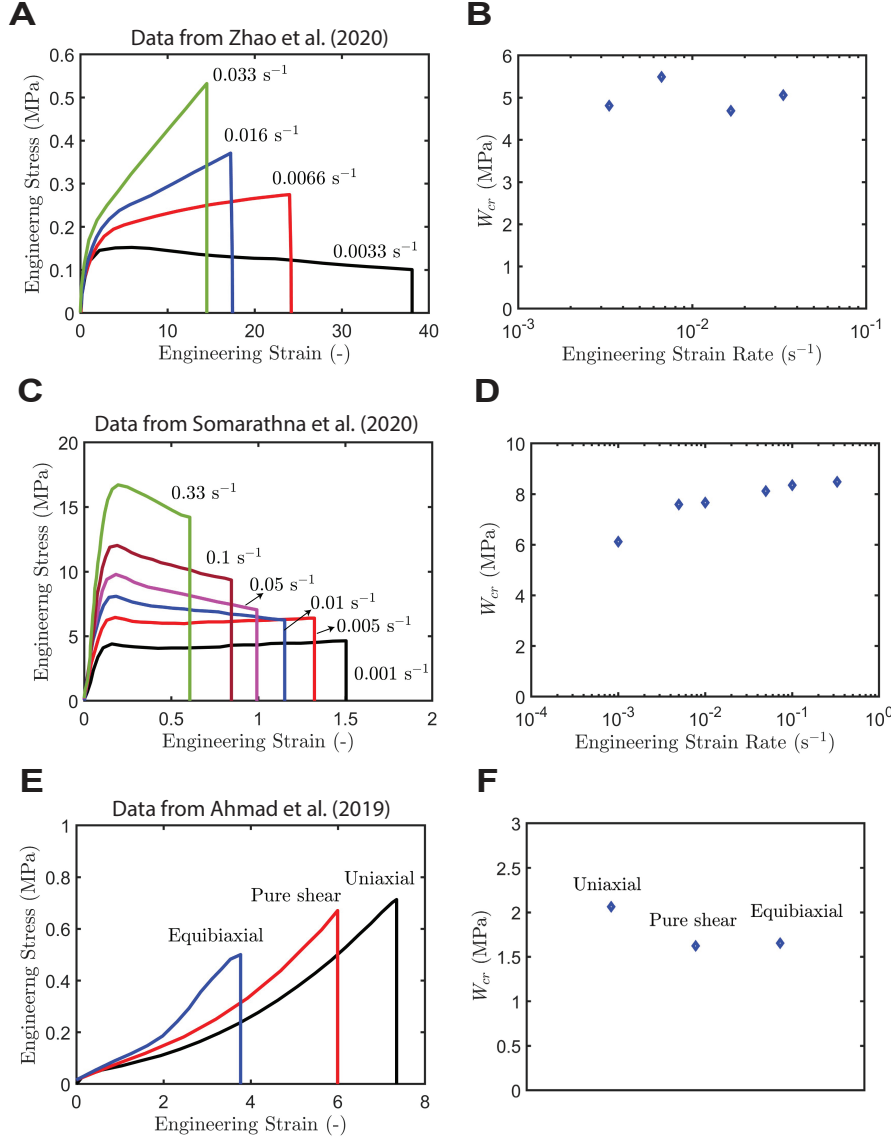


Figure 2.3: **Approximately constant W_{cr} for broad loading rates and loading modes.** (A) The engineering stress-strain curves at different strain rates for a rate-dependent viscous soft polymer with dynamic crosslinks (TFPM-PDMS-25000) from (Zhao et al., 2020b). (B) W_{cr} evaluated as the area under the stress-strain curve is approximately constant over the decade of strain rate considered. (C), (D) The engineering stress-strain curves for polyurethane from (Somarathna et al., 2020) and roughly constant W_{cr} evaluated over two decades of strain rates, respectively. (E) The Uniaxial tension, Pure shear, and Equibiaxial tension engineering stress-strain curves for Ecoflex elastomer at a stretch rate of 0.3 s^{-1} from (Ahmad et al., 2019). (F) Ecoflex's approximately constant W_{cr} evaluated for three different loading modes considered.

2.4.1 Method of virtual power for multi-mechanism deformation and damage

The kinematic rate-like descriptors in the framework, while considering M parallel mechanisms of deformation that capture the mechanical response of a material, are taken as

$$\dot{\chi}_i, \dot{F}_{ij}^{e(1)}, \dot{F}_{ij}^{e(2)}, \dots, \dot{F}_{ij}^{e(M)}, \dot{\epsilon}^{p(1)}, \dot{\epsilon}^{p(2)}, \dots, \dot{\epsilon}^{p(M)}, \dot{d}^{(1)}, \dot{d}^{(2)}, \dots, \dot{d}^{(M)}, \frac{\partial \dot{d}^{(1)}}{\partial X_i}, \frac{\partial \dot{d}^{(2)}}{\partial X_i}, \dots, \frac{\partial \dot{d}^{(M)}}{\partial X_i}. \quad (2.2)$$

χ_i is a smooth one-to-one mapping of a material point X_i to its position in the deformed configuration x_i as $x_i = \chi_i(X_1, X_2, X_3, t)$. $F_{ij} = \frac{\partial \chi_i}{\partial X_j}$ is the deformation gradient tensor. Each mechanism at a material point is assumed to experience the same total deformation quantified by the deformation gradient tensor. $F_{ij}^{e(q)}$ and $F_{ij}^{p(q)}$ are the elastic and plastic parts of F_{ij} in mechanism $q \in [1, M]$ following the multiplicative decomposition $F_{ij} = F_{ik}^{e(q)} F_{kj}^{p(q)}$.

$$\dot{\epsilon}^{p(q)} \stackrel{\text{def}}{=} \sqrt{D_{ij}^{p(q)} D_{ij}^{p(q)}}, \quad D_{ij}^{p(q)} = \frac{1}{2} \left(\dot{F}_{ik}^{p(q)} F_{kj}^{p(q)-1} + F_{ki}^{p(q)-1} \dot{F}_{jk}^{p(q)} \right), \quad \epsilon^{p(q)} \stackrel{\text{def}}{=} \int_0^t \dot{\epsilon}^{p(q)} dt, \\ d^{(q)} \in [0, 1] \quad : q \in [1, M], \quad (2.3)$$

are a measure of plastic strain rate, plastic stretching tensor corresponding to $F_{ij}^{p(q)}$, a measure of plastic strain, and damage variable, respectively, for mechanism q . The model is developed for isotropic materials, and consequently, the assumption of plastic irrotationality is valid. The definition of velocity gradient $L_{ij} (= \dot{F}_{ik} F_{kj}^{-1})$ and the multiplicative decomposition of F_{ij} result in the following constraints

$$\frac{\partial \dot{\chi}_i}{\partial X_k} F_{kj}^{-1} = \dot{F}_{ik}^{e(q)} F_{kj}^{e(q)-1} + \dot{\epsilon}^{p(q)} F_{ik}^{e(q)} N_{kl}^{p(q)} F_{lj}^{e(q)-1}, \quad N_{ij}^{p(q)} = \frac{D_{ij}^{p(q)}}{\left(\sqrt{D_{kl}^{p(q)} D_{kl}^{p(q)}}\right)} : \quad \forall q \in [1, M], \quad (2.4)$$

where $N_{ij}^{p(q)}$ is the plastic flow direction for mechanism q . Evolution of a reference body B and an arbitrary part of it, denoted by P , with outward unit normal n_i^R on its boundary ∂P is associated with macroscopic and microscopic force systems. The macroscopic system is defined by (i) traction t_i^R corresponding to n_i^R and generalized body force per unit referential volume b_i^R expending power over $\dot{\chi}_i$ on ∂P and in P , respectively. Specifically, $b_i^R = b_i^{0R} - \rho_R \ddot{\chi}_i$, with b_i^{0R} being the conventional body force per unit referential volume, $-\rho_R \ddot{\chi}_i$ representing the inertial body force per unit referential volume, ρ_R denoting the referential mass density and (ii) elastic stress $P_{ij}^{e(q)}$ expending power over the elastic distortion rate $\dot{F}_{ij}^{e(q)}$ in P .

The microscopic system is defined with: (i) scalar microscopic stresses $\pi^{(q)}$ expending power over $\dot{\epsilon}^{p(q)}$ in P , (ii) scalar microscopic stresses $\bar{\omega}^{(q)}$ expending power over $\dot{d}^{(q)}$ in P , (iii) vector microscopic stresses $\xi_i^{(q)}$ expending power over $\frac{\partial \dot{d}^{(q)}}{\partial X_i}$ in P and (iv) scalar microscopic tractions $\Upsilon^{(q)}(n_i^R)$ expending power over $\dot{d}^{(q)}$ on ∂P . For a given P , the power expended on P by the material external to it ($\mathcal{W}_{ext}(P)$) and the power expenditure within P ($\mathcal{W}_{int}(P)$) respectively, can be expressed as

$$\begin{aligned} \mathcal{W}_{ext}(P) &= \int_{\partial P} t_i^R \dot{\chi}_i da_R + \int_P b_i^R \dot{\chi}_i dv_R + \int_{\partial P} \sum_{q=1}^M \left(\Upsilon^{(q)} \dot{d}^{(q)} \right) da_R, \\ \mathcal{W}_{int}(P) &= \int_P \left(\sum_{q=1}^M \left[J^{p(q)} P_{ij}^{e(q)} \dot{F}_{ij}^{e(q)} + J^{p(q)} \pi^{(q)} \dot{\epsilon}^{p(q)} + \bar{\omega}^{(q)} \dot{d}^{(q)} + \xi_i^{(q)} \frac{\partial \dot{d}^{(q)}}{\partial X_i} \right] \right) dv_R. \end{aligned} \quad (2.5)$$

Note that the stress powers $P_{ij}^{e(q)} \dot{F}_{ij}^{e(q)}$, $\pi^{(q)} \dot{\epsilon}^{p(q)}$ in (2.5) are per unit volume ($J^{p(q)}$) of the intermediate space corresponding to $F_{ij}^{p(q)}$. Assuming that the fields $\chi_i, F_{ij}^{e(1)}, F_{ij}^{e(2)}, \dots, F_{ij}^{e(M)}$,

$\dots\epsilon^{p(1)}, \epsilon^{p(2)}, \epsilon^{p(M)}, d^{(1)}, d^{(2)}, \dots, d^{(M)}$ are known at an arbitrarily chosen time instant, the fields $\dot{\chi}_i, \dot{F}_{ij}^{e(1)}, \dot{F}_{ij}^{e(2)}, \dots, \dot{F}_{ij}^{e(M)}, \dot{\epsilon}^{p(1)}, \dot{\epsilon}^{p(2)}, \dots, \dot{\epsilon}^{p(M)}, \dot{d}^{(1)}, \dot{d}^{(2)}, \dots, \dot{d}^{(M)}$ need to be specified independently while satisfying the constraints in (2.4). The generalized virtual velocity can be written as

$$\mathcal{V} = (\tilde{\chi}_i, \tilde{F}_{ij}^{e(1)}, \tilde{F}_{ij}^{e(2)}, \dots, \tilde{F}_{ij}^{e(M)}, \tilde{\epsilon}^{p(1)}, \tilde{\epsilon}^{p(2)}, \dots, \tilde{\epsilon}^{p(M)}, \tilde{d}^{(1)}, \tilde{d}^{(2)}, \dots, \tilde{d}^{(M)}). \quad (2.6)$$

The virtual field $\mathcal{V}$ is rigid if it satisfies

$$(\nabla \tilde{\chi}_i) = \tilde{F}_{ij} = \Omega_{ik} F_{kj}, \quad \tilde{F}_{ij}^{e(q)} = \Omega_{ik} F_{kj}^{e(q)}, \quad \tilde{\epsilon}^{p(q)} = 0, \quad \tilde{d}^{(q)} = 0 : \forall q \in [1, M], \quad (2.7)$$

where Ω_{ij} is a spatially constant skew tensor. Here, $(\tilde{\cdot})$ denotes virtual field quantities.

The principle of virtual power requires that

$$\mathcal{W}_{int}(\mathbf{P}, \mathcal{V}) = \mathcal{W}_{ext}(\mathbf{P}, \mathcal{V}) : \forall \mathcal{V} \text{ and } \mathcal{W}_{int}(\mathbf{P}, \mathcal{V}) = 0 : \forall \text{ rigid } \mathcal{V}. \quad (2.8)$$

The total free energy density per unit referential volume ψ_R is assumed to be additively decomposable into contributions from each mechanism of deformation and energetic parts accounting for the effects of gradients in the damage fields $\psi^{d(q)}$: $q \in [1, M]$ as

$$\psi_R = \sum_{q=1}^M \left(J^{p(q)} \psi^{(q)} + \psi^{d(q)} \right), \quad T_{ij} = \sum_{q=1}^M T_{ij}^{(q)}, \quad (2.9)$$

where $\psi^{(q)}$ is the contribution from mechanism q per unit volume of the corresponding intermediate space $(J^{p(q)})$. $T_{ij}, T_{ij}^{(q)}$ are the total Cauchy stress and total Cauchy stress contribution from mechanism q , respectively. $\psi^{d(q)}$ can be interpreted as defect energy due to local elastic incompatibilities caused by gradients in the damage field variable $d^{(q)}$. This is similar to the interpretation of the free energy contributions due to plastic

strain gradients in standard strain-gradient plasticity theories (Lele and Anand, 2009; Reddy, 2011; Gao et al., 1999). The total first Piola-Kirchoff stress P_{ij} , the total second Piola-Kirchoff stress S_{ij} and T_{ij} are related through the following relations

$$P_{ij} = JT_{ik}F_{jk}^{-1} = J\left(\sum_{q=1}^M T_{ik}^{(q)}\right)F_{jk}^{-1} = \sum_{q=1}^M JT_{ik}^{(q)}F_{jk}^{-1} = \sum_{q=1}^M P_{ij}^{(q)}, \quad P_{ij} = F_{ik}S_{kj}, \quad (2.10)$$

$$S_{ij} = JF_{ik}^{-1}T_{kl}F_{jl}^{-1},$$

where $P_{ij}^{(q)}$ is the total first Piola-Kirchoff stress contribution from mechanism q .

2.4.2 Consequences of the method of virtual power: Macroscopic and microscopic stresses for multi-mechanism deformation and damage

The principle of virtual power necessitates that (2.8) is satisfied. To deduce the consequences, any generalized virtual velocity $\mathcal{V}$ consistent with (2.4) can be chosen. Standard procedure and variational arguments to obtain macroscopic and microscopic force balances (Germain, 1973; Gurtin, 1996, 2002; Gurtin et al., 2010; Narayan and Anand, 2019) are followed, and the results are summarized.

Macroscopic stresses

A $\mathcal{V}$ with $\tilde{\epsilon}^{p(q)} \equiv 0$, $\tilde{d}^{(q)} \equiv 0 : \forall q \in [1, M]$ is considered, so that $\frac{\partial \tilde{\chi}_i}{\partial X_j} = \tilde{F}_{ik}^{e(q)} F_{kj}^{p(q)}$. Using (2.10) and standard arguments, (2.8) results in

$$t_i^R = P_{ij}n_j^R, \quad \frac{\partial P_{ij}}{\partial X_j} + b_i^{0R} = \rho_R \ddot{\chi}_i : P_{ij}^{(q)} \stackrel{\text{def}}{=} J^{p(q)} P_{ik}^{e(q)} F_{jk}^{p(q)-1} \forall q \in [1, M]. \quad (2.11)$$

(2.11) represents the standard referential forms of macroscopic traction condition and force balance, respectively. The second requirement of the principle of virtual power, as given in (2.8), leads to

$$P_{ik}F_{jk} = F_{ik}P_{jk}, \quad (2.12)$$

which is the standard result obtained from macroscopic moment balance.

Microscopic stresses

(i) Scalar microstresses $\pi^{(q)}$

Next we consider a $\mathcal{V}$ with $\tilde{\chi}_i \equiv 0_i$, $\tilde{d}^{(q)} \equiv 0 : \forall q \in [1, M]$ and arbitrarily chosen $\tilde{\epsilon}^{p(q)} : q \in [1, M]$. Two additional stress measures are defined, (i) Symmetric elastic second Piola-Kirchhoff stress and (ii) Mandel stress, respectively, as

$$S_{ij}^{e(q)} \stackrel{\text{def}}{=} J^{e(q)} F_{ik}^{e(q)-1} T_{kl}^{(q)} F_{jl}^{e(q)-1}, \quad M_{ij}^{e(q)} \stackrel{\text{def}}{=} C_{ik}^{e(q)} S_{kj}^{e(q)} = J^{e(q)} F_{ki}^{e(q)} T_{kl}^{(q)} F_{jl}^{e(q)-1} : \forall q \in [1, M]. \quad (2.13)$$

$C_{ij}^{e(q)} = F_{ki}^{e(q)} F_{kj}^{e(q)}$ is the elastic right Cauchy-Green deformation tensor for mechanism q . The first requirement of the principle of virtual power given in (2.8) results in the microscopic force balance

$$\pi^{(q)} = \frac{1}{2} \left(M_{ij}^{e(q)} + M_{ji}^{e(q)} \right) N_{ij}^{p(q)} \quad \forall q \in [1, M]. \quad (2.14)$$

(ii) Scalar microstresses $\bar{\omega}^{(q)}$ and vector microstresses $\xi_i^{(q)}$

To obtain $\bar{\omega}^{(q)}$ and $\xi_i^{(q)}$, a $\mathcal{V}$ with $\tilde{\epsilon}^{p(q)} \equiv 0 : \forall q \in [1, M]$ and $\tilde{\chi}_i \equiv 0_i$ is considered. Choosing $\tilde{d}^{(q)} : q \in [1, M]$ arbitrarily, (2.8) yields the microscopic traction conditions and force balances respectively as

$$\Upsilon^{(q)} = \xi_i^{(q)} n_i^R, \quad \frac{\partial \xi_i^{(q)}}{\partial X_i} - \bar{\omega}^{(q)} = 0 : \forall q \in [1, M]. \quad (2.15)$$

A set of thermodynamically consistent constitutive equations, along with the above macroscopic and microscopic force balances, gives the complete set of governing mechanical equations for the model. The internal power expenditure in (2.5) can now be rewritten using (2.13) as

$$\mathcal{W}_{int}(\mathbf{P}) = \int_{\mathbf{P}} \left(\sum_{q=1}^M \left[\frac{1}{2} J^{p(q)} S_{ij}^{e(q)} \dot{C}_{ij}^{e(q)} + J^{p(q)} \pi^{(q)} \dot{\epsilon}^{p(q)} + \bar{\omega}^{(q)} \dot{d}^{(q)} + \xi_i^{(q)} \frac{\partial \dot{d}^{(q)}}{\partial X_i} \right] \right) dv_{\mathbf{R}}. \quad (2.16)$$

2.4.3 Free energy imbalance

Under isothermal conditions, the local form of free energy imbalance can be finally expressed as

$$\dot{\psi}_{\mathbf{R}} - \sum_{q=1}^M \left(\frac{1}{2} J^{p(q)} S_{ij}^{e(q)} \dot{C}_{ij}^{e(q)} + J^{p(q)} \pi^{(q)} \dot{\epsilon}^{p(q)} + \bar{\omega}^{(q)} \dot{d}^{(q)} + \xi_i^{(q)} \frac{\partial \dot{d}^{(q)}}{\partial X_i} \right) \leq 0. \quad (2.17)$$

We note here that all the terms involved in (2.17) are frame invariant.

2.4.4 Constitutive equations for deformation and damage

Using (2.9), the free energy imbalance in (2.17) can be written as

$$\begin{aligned} \sum_{q=1}^M \left(J^{p(q)} \left(\frac{\partial \hat{\psi}^{(q)}(\Lambda^{(q)})}{\partial C_{ij}^{e(q)}} - \frac{1}{2} S_{ij}^{e(q)} \right) \dot{C}_{ij}^{e(q)} - J^{p(q)} \left(\pi^{(q)} - \frac{\partial \hat{\psi}^{(q)}(\Lambda^{(q)})}{\partial \epsilon^{p(q)}} \right) \dot{\epsilon}^{p(q)} - \left(\bar{\omega}^{(q)} - \frac{\partial \tilde{\psi}_R(\Lambda)}{\partial d^{(q)}} \right) \dot{d}^{(q)} \right. \\ \left. - \left(\xi_i^{(q)} - \frac{\partial \tilde{\psi}_R(\Lambda)}{\partial (\frac{\partial d^{(q)}}{\partial X_i})} \right) \frac{\partial \dot{d}^{(q)}}{\partial X_i} \right) \leq 0. \end{aligned} \quad (2.18)$$

where

$$\Lambda^{(q)} = \left(C_{ij}^{e(q)}, \epsilon^{p(q)}, d^{(q)}, \frac{\partial d^{(q)}}{\partial X_i} \right) \quad q \in [1, M], \quad \Lambda = (\Lambda^{(1)}, \Lambda^{(2)}, \dots, \Lambda^{(M)}), \quad (2.19)$$

are generalized lists of variables. $\psi^{d(q)}$ is taken to be dependent on $d^{(q)}, \frac{\partial d^{(q)}}{\partial X_i}$. The following state relations are assumed

$$S_{ij}^{e(q)} = 2 \frac{\partial \hat{\psi}^{(q)}(\Lambda^{(q)})}{\partial C_{ij}^{e(q)}}, \quad \xi_i^{(q)} = \frac{\partial \tilde{\psi}_R(\Lambda)}{\partial (\frac{\partial d^{(q)}}{\partial X_i})}, \quad \pi_{en}^{(q)} = \frac{\partial \hat{\psi}^{(q)}(\Lambda^{(q)})}{\partial \epsilon^{p(q)}}, \quad \bar{\omega}_{en}^{(q)} = \frac{\partial \tilde{\psi}_R(\Lambda)}{\partial d^{(q)}} : \forall q \in [1, M], \quad (2.20)$$

with $\pi^{(q)} = \pi_{en}^{(q)} + \pi_{dis}^{(q)}$, $\bar{\omega}^{(q)} = \bar{\omega}_{en}^{(q)} + \bar{\omega}_{dis}^{(q)}$. The subscripts *en* and *dis* denote energetic and dissipative parts, respectively. To satisfy the dissipation inequality in (2.18), it is assumed that

$$\pi_{dis}^{(q)} \dot{\epsilon}^{p(q)} \geq 0, \quad \bar{\omega}_{dis}^{(q)} \dot{d}^{(q)} \geq 0 \quad \forall q \in [1, M]. \quad (2.21)$$

The constitutive dependence of ψ_R , $\psi^{(q)}$, and $\psi^{d(q)}$ on the generalized lists of variables in (2.19) can be simplified without significant loss of generality. Specifically,

$$\psi_R = \sum_{q=1}^M \left[g^{(q)}(d^{(q)}) \psi_o^{(q)+} + \psi_o^{(q)-} + \underbrace{\psi_*^{(q)} [l^{(q)}]^2 \left(\frac{\partial d^{(q)}}{\partial X_i} \frac{\partial d^{(q)}}{\partial X_i} \right)}_{\psi^{d(q)}} \right], \quad \psi_o = \sum_{q=1}^M \psi_o^{(q)}(C_{ij}, C_{ij}^{e(q)}), \quad (2.22)$$

where $0 \leq g^{(q)}(d^{(q)}) \leq 1$ are monotonically decreasing functions which degrade the energy storage with damage growth at a material point. $\psi_*^{(q)} > 0$ and $l^{(q)} > 0$ are coefficients with units of energy per unit volume and length scale parameters that control the spread of the diffuse damage zone corresponding to $d^{(q)}$, respectively. ψ_o is the total undamaged elastic free energy, which is the additive decomposition of contributions $(\psi_o^{(q)})$ from the M mechanisms of deformation. $\psi_o^{(q)-}$ is the part of $\psi_o^{(q)}$ which is compressive dilatational. $\psi_o^{(q)+} = \psi_o^{(q)} - \psi_o^{(q)-}$ is the damage driving part of $\psi_o^{(q)}$. This split accounts for damage growth being driven only by distortional and tensile dilatational deformation (Amor et al., 2009; Hesammokri et al., 2023).

Further, using (2.22) and (2.20), we get

$$\pi_{en}^{(q)} = 0, \quad \bar{\omega}_{en}^{(q)} = \frac{\partial g^{(q)}(d^{(q)})}{\partial d^{(q)}} \psi_o^{(q)}, \quad \xi_i^{(q)} = 2\psi_*^{(q)} [l^{(q)}]^2 \frac{\partial d^{(q)}}{\partial X_i}. \quad (2.23)$$

Also, $\pi_{dis}^{(q)}, \bar{\omega}_{dis}^{(q)}$ can be taken in their simplified general forms as

$$\pi_{dis}^{(q)} = g^{(q)}(d^{(q)}) \mathcal{S}^{(q)}(C_{ij}^{e(q)}, \dot{\epsilon}^{p(q)}) \text{ when } \dot{\epsilon}^{p(q)} > 0, \quad \bar{\omega}_{dis}^{(q)} = \hat{\bar{\omega}}_{dis}^{(q)}(d^{(q)}, \dot{d}^{(q)}) \text{ when } \dot{d}^{(q)} > 0, \quad (2.24)$$

with $\mathcal{S}^{(q)}(C_{ij}^{e(q)}, \dot{\epsilon}^{p(q)})$, $\hat{\bar{\omega}}_{dis}^{(q)}(d^{(q)}, \dot{d}^{(q)})$ being non-negative so that the reduced dissipation

inequalities in (2.21) are satisfied.

2.5 Framework specialization to model soft polymers

The functional forms for the deformation-related quantities during specialization, specifically ψ_o and $\mathbf{T}_o$, are taken from our previous work (Konale et al., 2023). In the following sub-sections, the multi-mechanism fracture modeling framework developed in Section 2.4 is specialized along with the ψ_o , $\mathbf{T}_o$ expressions to model damage initiation and evolution. ψ_o^+ is introduced and discussed as a damage driving force with ψ_{cr}^+ (the energetic part of W_{cr}) as a damage initiation criterion.

2.5.1 Free energy and Cauchy stress

To specialize the fracture modeling framework for soft polymers, we restrict the number of micro-mechanisms M to 2 in (2.22). As an approximation, only one damage variable is used. This reflects the commonly observed single load drop corresponding to fracture in experiments for a broad class of soft polymers in the literature. The single load drop is also shown in our experiments on notched PBS specimens (Figure 2.6(B)). The resistance to deformation results from the underlying bonds and their networks. For a material point to be completely damaged, both mechanisms A and B have to be completely damaged, i.e., all bonds resisting deformation must be broken. In the case of multiple load drop observation (King, 2022; Imaoka et al., 2023), different damage variables for each mechanism need to be used. Some works on fracture modeling of composites have incorporated multiple damage variables to account for matrix and filler damage separately (Zhang et al., 2022; Jihai Yuan and Chen, 2022; Li et al., 2024).

Constitutive model for viscous soft polymer PBS

The two mechanisms of deformation considered are: (a) **Intermolecular Resistance** (A): Elastic and dissipative resistance due to intermolecular interactions between neighboring molecules (Ames et al., 2009; Srivastava et al., 2010a; Lamont et al., 2025). (b) **Network Resistance** (B): stretching of constituent subchains within Boron(B):Oxygen(O) coordinate-bond dynamic crosslinks (Konale et al., 2023).

ψ_o is given as

$$\begin{aligned}
 \psi_o &= \underbrace{\psi_o^{(A)}}_{\text{Intermolecular resistance}} + \underbrace{\psi_o^{(B)}}_{\text{Network resistance}}, \\
 &= \underbrace{G \left(E_{ij}^{e(A)\prime} E_{ij}^{e(A)\prime} \right) + \frac{1}{2} K \left(E_{kk}^{e(A)} \right)^2}_{\psi_o^{(A)}} \\
 &\quad + \underbrace{\mu \bar{\lambda}_L^2 \left(e^{-k_{ns}t} \zeta(t/0) + \int_0^t \frac{n_{ns}}{n_s} k_s e^{-k_{ns}(t-t')} \zeta(t/t') dt' \right)}_{\psi_o^{(B)}}, \tag{2.25}
 \end{aligned}$$

with $\zeta(t/t') = \left[\left(\frac{\bar{\lambda}(t/t')}{\bar{\lambda}_L} \right) \beta(t/t') + \ln \left(\frac{\beta(t/t')}{\sinh \beta(t/t')} \right) \right]$.

if $E_{kk}^{e(A)} < 0$: $\psi_o^{(A)+} = G \left(E_{ij}^{e(A)\prime} E_{ij}^{e(A)\prime} \right)$, $\psi_o^{(A)-} = \frac{1}{2} K \left(E_{kk}^{e(A)} \right)^2$,

else : $\psi_o^{(A)+} = \psi_o^{(A)}$, $\psi_o^{(A)-} = 0$.

We note that $\psi_o^{(B)}$ is purely distortional. Any suitable free energy density base function can be used in (2.25) for a specific soft polymer under consideration. $G > 0$, $K > 0$ are the shear and bulk moduli respectively. $E_{ij}^{e(A)} = \ln U_{ij}^{e(A)}$ is an elastic logarithmic strain measure, where $U_{ij}^{e(A)}$ is the right elastic stretch tensor following the polar decomposition of $F_{ij}^{e(A)}$. $A_{ij}^{\prime}$ denotes the deviatoric part of a tensor A_{ij} . Plastic incompressibility is assumed for mechanism A . $\bar{\lambda}_L$ is the network locking stretch parameter accounting for finite chain extensibility. $\bar{\lambda} \stackrel{\text{def}}{=} \sqrt{\frac{C_{kk}^*}{3}}$ is the effective total distortional stretch with C_{ij}^* being the distortional part of C_{ij} . $\mathcal{L}(\cdot) = \coth(\cdot) - \frac{1}{(\cdot)}$ denotes the Langevin function

and $\beta = \mathcal{L}^{-1} \left(\frac{\bar{\lambda}}{\bar{\lambda}_L} \right)$. The referential number density of chain segments currently in their subchain state (S) and that of chain segments that can be in the subchain state but are not currently (NS) are denoted by n_s and n_{ns} , respectively. The underlying dynamic, reversible reactions can be represented as



where k_s and k_{ns} are the forward and backward reaction rate parameters, respectively. $\mu = n_s k_B \theta$ denotes the ground state shear modulus with k_B , θ denoting the Boltzmann constant and temperature respectively. Kinematic quantities with notation (t/t') use the relative deformation gradient: $F_{ij}(t/t') = F_{ik}(t)F_{kj}^{-1}(t')$, a temporal decomposition of F_{ij} , which maps the deformed configuration at t' to the deformed configuration at t as the input. This accounts for subchains formed at t' ($0 < t' < t$) and surviving till the current time t being stretched with respect to their stress-free, relaxed configurations corresponding to t' . $\psi_o^{(B)}$ in (2.25) is the summation of the stretching energies of these subchains with different stretch levels corresponding to their formation time instants. This proposed free energy formulation avoids having a thermodynamically inconsistent, explicitly rate-dependent free energy density function that is sometimes proposed in the literature.

The degradation function $g(d)$ is taken as (Miehe et al., 2010a; Mao et al., 2017; Anand et al., 2019; Konica and Sain, 2021; Narayan and Anand, 2021)

$$g(d) = (1 - d)^2. \quad (2.27)$$

The total Cauchy stress is given as

$$\begin{aligned}
T_{ij} &= g(d) \left[\underbrace{T_{ij}^{(A)o+} + T_{ij}^{(B)o}}_{T_{ij}^{o+}} \right] + \underbrace{T_{ij}^{(A)o-}}_{T_{ij}^{o-}}, \\
\text{if } E_{mm}^{e(A)} < 0 : T_{ij}^{(A)o+} &= J^{-1} R_{ik}^{e(A)} \left(2G E_{kl}^{e(A)\prime} \right) R_{jl}^{e(A)}, \\
T_{ij}^{(A)o-} &= J^{-1} R_{ik}^{e(A)} \left(K E_{mm}^{e(A)} \delta_{kl} \right) R_{jl}^{e(A)}, \\
\text{else : } T_{ij}^{(A)o+} &= J^{-1} R_{ik}^{e(A)} \left(2G E_{kl}^{e(A)\prime} + K E_{mm}^{e(A)} \delta_{kl} \right) R_{jl}^{e(A)}, \\
T_{ij}^{(A)o-} &= 0, \\
T_{ij}^{(B)o} &= e^{-k_{ns}t} \mu \Gamma_{ij}(t/0) + \int_0^t \frac{n_{ns}}{n_s} k_s e^{-k_{ns}(t-t')} \mu \Gamma_{ij}(t/t') dt', \\
\Gamma_{ij}(t/t') &= J^{-1}(t/t') \frac{\bar{\lambda}_L}{\bar{\lambda}(t/t')} \mathcal{L}^{-1} \left[\frac{\bar{\lambda}(t/t')}{\bar{\lambda}_L} \right] B_{ij}^*(t/t').
\end{aligned} \tag{2.28}$$

$T_{ij}^o = T_{ij}^{o+} + T_{ij}^{o-}$ is the undamaged total Cauchy stress. T_{ij}^{o+} , T_{ij}^{o-} are the undamaged total Cauchy stress contributions corresponding to ψ_o^+ , ψ_o^- respectively. $T_{ij}^{(A)o} = T_{ij}^{(A)o+} + T_{ij}^{(A)o-}$ and $T_{ij}^{(B)o}$ are the undamaged total Cauchy stress contributions from mechanisms A ($\psi_o^{(A)} = \psi_o^{(A)+} + \psi_o^{(A)-}$) and B ($\psi_o^{(B)}$) respectively (Konale et al., 2023). $B_{ij} = F_{ik} F_{jk}$ is the total left Cauchy-Green deformation tensor.

Broader applicability of the general constitutive model for PBS

Setting $G = k_{ns} = k_s = 0$ in (2.25) results in $\psi_o = \mu \bar{\lambda}_L^{-2} \zeta(t/0)$, which is the eight-chain free energy density function (Arruda and Boyce, 1993) used for elastomers. The effect of k_{ns} 's value on the mechanical response can be understood easily by setting $G = 0$ in (2.25) to get $\psi_o = \psi_o^{(B)}$. At any given time instant, smaller values of k_{ns} will result in ψ_o tending to the eight-chain form $\mu \bar{\lambda}_L^{-2} \zeta(t/0)$, i.e., elastomer-like behavior with insignificant network reconfiguration. On the other hand, higher values of k_{ns} will lead to the departure of ψ_o from the elastic behavior to significantly viscous behavior with appreciable network reconfiguration. Hence, *the proposed constitutive model can be used as a general model for a broad class of soft polymers with response varying from elastic to significantly viscous.*

2.5.2 Viscous flow rule for Mechanism A

Assuming plastic irrotationality, plastic incompressibility, and co-directionality of plastic flow, the plastic stretching tensor $D_{ij}^{p(A)}$ can be expressed as

$$D_{ij}^{p(A)} = \nu^{p(A)} \left(\frac{M_{ij}^{e(A)\prime}}{2\bar{\tau}^{(A)}} \right), \quad \nu^{p(A)} \stackrel{\text{def}}{=} \sqrt{2} \sqrt{D_{ij}^{p(A)} D_{ij}^{p(A)}}, \quad \bar{\tau}^{(A)} \stackrel{\text{def}}{=} \frac{1}{\sqrt{2}} \sqrt{M_{ij}^{e(A)\prime} M_{ij}^{e(A)\prime}}, \quad (2.29)$$

where $\nu^{p(A)}$, $\bar{\tau}^{(A)}$ are the equivalent plastic shear strain rate and the equivalent shear stress, respectively. With $\gamma^{p(A)} \stackrel{\text{def}}{=} \int_0^t \nu^{p(A)} dt$ being an equivalent plastic shear strain rate, we have $\nu^{p(A)} = \sqrt{2}\dot{\epsilon}^{p(A)}$, $\gamma^{p(A)} = \sqrt{2}\epsilon^{p(A)}$. Instead of $\dot{\epsilon}^{p(A)}$, we use $\nu^{p(A)}$ as the rate-like kinematic descriptor. The expression for $\pi^{(A)}$ in (2.14) will have to be divided by $\sqrt{2}$ to account for the change of $\dot{\epsilon}^{p(A)}$ to $\nu^{p(A)}$ as the kinematic descriptor. Using (2.14), (2.23) and (2.29), we have $\bar{\tau}^{(A)} = \pi^{(A)} = \pi_{dis}^{(A)}$.

Further, it is assumed that $\mathcal{S}^{(A)}$ in (2.24) depends on $C_{ij}^{e(A)}$ only through $\hat{p}^{(A)} = -K E_{kk}^{e(A)} = -K \left(\frac{1}{2} \text{trace}(\ln C_{ij}^{e(A)}) \right)$. Also, $\hat{p}^{(A)} = \frac{\bar{p}^{(A)}}{g(d)}$ with $\bar{p}^{(A)} \stackrel{\text{def}}{=} -\frac{1}{3} M_{kk}^{e(A)}$. $\mathcal{S}^{(A)}(C_{ij}^{e(A)}, \nu^{p(A)})$ is chosen to follow a specific form as

$$\mathcal{S}^{(A)}(C_{ij}^{e(A)}, \nu^{p(A)}) = \alpha_p \hat{p}^{(A)} + S \sinh^{-1} \left[\left(\frac{\nu^{p(A)}}{\nu_0} \right)^m \right], \quad (2.30)$$

where $\alpha_p \geq 0$ is a pressure sensitivity parameter. The last term in (2.30) represents a rate-dependent resistance to plastic flow. S is an internal variable that models the dissipative friction-like resistance due to the sliding chains. $\nu_0 > 0$ is a reference strain rate and $m \in (0, 1]$ is a strain rate sensitivity parameter. The evolution equation for S is taken as $\dot{S} = h (\bar{\lambda} - 1) \nu$ with initial value $S(X_i, 0) = S_0 > 0$. h is a stress-dimensioned material parameter and $\nu \stackrel{\text{def}}{=} \sqrt{2} \sqrt{D_{ij}^{e(A)\prime} D_{ij}^{e(A)\prime}}$ is the total equivalent shear strain rate. The strength relation in (2.24) for mechanism A can now be concisely expressed as

$$\nu^{p(A)} = \begin{cases} 0 & \text{if } \bar{\tau}_e^{(A)} \leq 0, \\ \nu_0 \left[\sinh \left(\frac{\bar{\tau}_e^{(A)}}{g(d)S} \right) \right]^{1/m} & \text{if } \bar{\tau}_e^{(A)} > 0, \end{cases} \quad (2.31)$$

where $\bar{\tau}_e^{(A)} \stackrel{\text{def}}{=} \bar{\tau}^{(A)} - \alpha_p \bar{p}^{(A)}$ denotes a net equivalent shear stress for plastic flow.

2.5.3 Evolution equation for damage variable d

The microstructural changes leading to damage in soft polymers like PBS that exhibit dynamic crosslinking are thought to be reversible, i.e., failure of weak dynamic, reversible crosslinks (Song et al., 2021; Shen et al., 2021; Shen and Vernerey, 2020). However, *short timescales for complete damage growth* with respect to the dynamic crosslink macroscopic formation timescale (τ_f) would suggest that damage, if initiated, will grow monotonically. Specimen geometry and experiment schematic for a notched tension test performed at 60 mm/s loading speed are shown in Figure 2.6(B). The sharp load drop corresponding to fracture in Figure 2.6(B) for the notched PBS specimen (τ_f is in the order of seconds for PBS (Konale et al., 2023)) highlights the rapid damage growth in PBS. Therefore, we adhere to the modeling approach for irreversible damage growth (Narayan and Anand, 2019; Konica and Sain, 2021).

Following Narayan and Anand (Narayan and Anand, 2019), the scalar microstress $\bar{\omega}_{dis}$ in (2.24) is taken in the following specific form

$$\bar{\omega}_{dis} = \alpha + \zeta \dot{d} : \alpha = 2(1-d)\psi_{cr}^+ + 2\psi_* d > 0 \text{ with } \psi_{cr}^+ > 0, \psi_* > 0, \quad (2.32)$$

where ψ_* , ψ_{cr}^+ are coefficients with units of energy per unit volume, and $\zeta > 0$ is a constant kinetic modulus so that the dissipation inequality is satisfied when $\dot{d} > 0$. The timescale for damage growth is determined by ζ , with decreasing values of ζ leading towards rate-independent damage evolution. $\psi_{cr}^+ + \psi_*$ represents a part of the total energy

dissipated per unit volume as d grows from 0 to 1. ψ_* denotes a part of the referential energy density dissipated after damage initiation till complete material point failure. The microforce balance in (2.15) along with (2.20) and (2.32) gives the evolution equation for d as

$$\zeta \dot{d} = 2(1 - d)\mathcal{H} - 2\psi_* \left(d - l^2 \frac{\partial^2 d}{\partial X_i \partial X_i} \right), \quad (2.33)$$

where $\zeta > 0$ is a constant kinetic modulus. ζ determines the timescale of damage growth with decreasing values of ζ leading towards rate-independent damage evolution. $\mathcal{H}$ is a monotonically increasing history function expressed as

$$\mathcal{H}(t) \stackrel{\text{def}}{=} \max_{s \in [0, t]} \left[\langle \psi_o^+(s) - \psi_{cr}^+ \rangle \right], \quad (2.34)$$

$$\langle \psi_o^+(s) - \psi_{cr}^+ \rangle = \begin{cases} 0 & \text{if } \psi_o^+(s) - \psi_{cr}^+ < 0, \\ \psi_o^+(s) - \psi_{cr}^+ & \text{if } \psi_o^+(s) - \psi_{cr}^+ \geq 0 \text{ and } \psi_o^{(B)} \geq \psi_{cr}^{(B)}. \end{cases}$$

Damage initiates only when ψ_o^+ reaches the critical value ψ_{cr}^+ (the energetic contribution to W_{cr}) along with $\psi_o^{(B)} = \psi_{cr}^{(B)}$. Here, material strength is explicitly accounted for. The additional condition of $\psi_o^{(B)} = \psi_{cr}^{(B)}$ ($\psi_o^{(B)}$ is purely distortional, $\psi_o^{(B)+} = \psi_o^{(B)}$) necessary to ensure consistency due to two mechanisms of deformation involved is motivated by (i) mechanism B being the dominant mechanism of deformation in the timescales of interest, and (ii) the experimentally observed single load drop corresponding to fracture. A physical interpretation can be that the high stresses resulting from the failure of mechanism B lead to the subsequent rapid failure of mechanism A .

It can be noted here that a macroscopic stretch measure, e.g., $\bar{\lambda}(t/0)$ by itself, cannot be used as a criterion for damage initiation. This can be understood by considering a loading rate much slower than the dynamic crosslink macroscopic relaxation timescale (τ_r). In this case, the polymer network can easily reconfigure. Even when a specimen is subjected

to large global deformation, the subchains, due to continuous network reconfigurations, will be stretched by an insignificant amount. For loading rates much faster than τ_r , the polymer network will not have sufficient time to reconfigure, and the subchains will undergo a significant amount of stretching upon large specimen deformation. Hence, to capture the extent of subchain stretching, $\bar{\lambda}(t/0)$ will have to be further supplemented with a measure of loading rate, e.g., $\dot{\bar{\lambda}}(t/0)$. $\bar{\lambda}(t/0)_{cr}$ will then not be a unique value but dependent on $\dot{\bar{\lambda}}(t/0)$. In contrast, ψ_{cr}^+ and $\psi_{cr}^{(B)}$ are unique values.

In this chapter, we have focused on fracture under an overall tensile loading. For soft polymers with tension-compression response asymmetry, different values for the parameter set $\{\psi_{cr}^+, \psi_*, l, \zeta\}$ can be used under compressive loading. This change of parameter values can be triggered using a field variable (e.g., hydrostatic pressure), which governs the tension-compression response asymmetry in the constitutive model.

The complete set of model parameters for deformation, damage initiation, and damage growth is

$$\underbrace{\{k_s, k_{ns}, n_s, G, K, \nu_0, m, \alpha_p, S_0, h, \bar{\lambda}_L\}}_{\text{deformation}}, \underbrace{\{\psi_{cr}^+, \psi_*, l, \zeta\}}_{\text{damage}}. \quad (2.35)$$

For incompressible elastomers, the list of model parameters is reduced to

$$\underbrace{\{n_s, \bar{\lambda}_L\}}_{\text{deformation}}, \underbrace{\{\psi_{cr}^+, \psi_*, l, \zeta\}}_{\text{damage}}. \quad (2.36)$$

2.6 Numerical procedure

2.6.1 Finite element implementation of the damage model

The model was implemented in the finite element software ABAQUS, which has been acceptably used for engineering simulations involving complex loadings, inhomogeneous deformations, and non-linear material responses (Konica et al., 2024; Niu and Srivastava, 2022a; Vaishakh et al., 2024; Niu and Srivastava, 2022b; Bai et al., 2021; Zhong and Srivastava, 2021; Srivastava et al., 2011). The damage variable d is an additional degree of freedom at each node, along with displacement. Therefore, implementation in ABAQUS requires the development of user-defined elements. Since the timescales involved are short, an explicit formulation was used, and correspondingly, a VUEL (user-defined element with explicit formulation) was developed. Following the recent works outlining the numerical implementation of phase field models using VUEL (Wang et al., 2020; Yuan et al., 2022; Hai et al., 2024), we split the problem computationally into (i) a mechanical problem (VUMAT, user-defined mechanical material behavior with inbuilt elements) and (ii) a problem corresponding to the additional degree of freedom d (VUEL). The evolution of d at a node is dependent upon quantities evaluated in the mechanical problem (VUMAT), specifically the damage driving history function $\mathcal{H}$ at the corresponding material (integration) points. Further, to update the total Cauchy stress T_{ij} at a material point, d , evaluated from the damage problem (VUEL) at the corresponding nodes is required. This information is passed between VUMAT and VUEL using global variables and the VUSDFLD subroutine. The finite element implementation workflow⁶ is summarized in Figure 2.4.⁷

⁶Pseudo input file and subroutines are presented in Appendix A.1. The relevant codes can be accessed at https://github.com/SrivastavaResearchLab/Polymer_Fracture_2025.git.

⁷Chapter 6 presents an additional application of the VUMAT-VUEL-VUSDFLD approach along with damage initiation modeling ideology in Section 2.5.3 for modeling damage and fracture in polymeric architected materials.

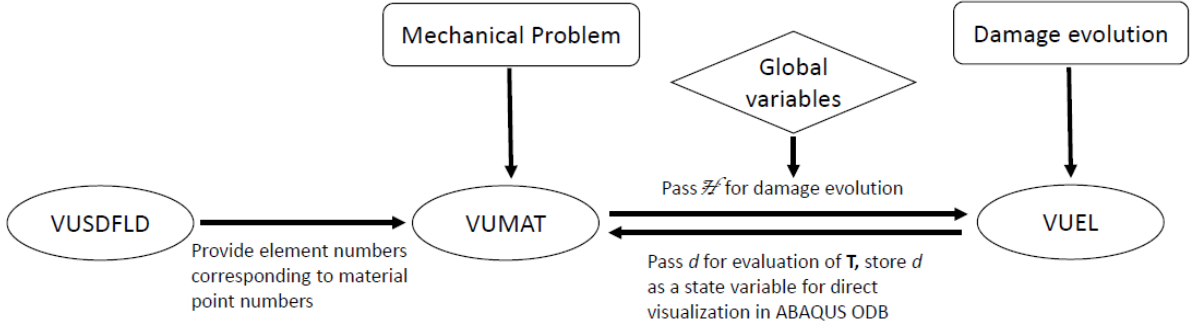


Figure 2.4: **Summary of the finite element implementation workflow.** The total Cauchy stress tensor T_{ij} is denoted by $\mathbf{T}$.

2.6.2 Numerical update procedure for $T_{ij}^{(B)o}$ and $\psi_o^{(B)}$

The finite element program ABAQUS returns only the deformation gradients from the previous and current time steps at each material point. Direct numerical implementation of the analytical expressions for $T_{ij}^{(B)o}$ and $\psi_o^{(B)}$ in ABAQUS will require the storage of the entire deformation gradient history along with many state-dependent variables at each material point. The resulting storage requirement will be huge, making it challenging. A numerical update procedure for $T_{ij}^{(B)o}$ developed in the previous chapter is applied, which requires deformation gradients only from the previous and current time steps at each material point. It is summarized as

$$\begin{aligned}
 T_{ij}^{(B)o}(t + \Delta t) &\rightarrow e^{-k_{ns}(t+\Delta t)} \mu \Gamma_{ij}(t + \Delta t/0) + T_{ij}^{(B)o2}(t + \Delta t), \\
 T_{ij}^{(B)o2}(t + \Delta t) &\rightarrow e^{-k_{ns}\Delta t} J^{-5/3}(t + \Delta t/t) F_{ik}(t + \Delta t/t) T_{kl}^{(B)o2}(t) F_{jl}(t + \Delta t/t) \\
 &+ \frac{1}{2} \frac{n_{ns}}{n_s} k_s e^{-k_{ns}\Delta t} \mu \Gamma_{ij}(t + \Delta t/t) \Delta t.
 \end{aligned} \tag{2.37}$$

We apply the procedure to $\psi_o^{(B)}$ which can be summarized as

$$\begin{aligned}
 \psi_o^{(B)}(t + \Delta t) &\rightarrow e^{-k_{ns}(t+\Delta t)} \mu \left[\bar{\lambda}_L \right]^2 \zeta(t + \Delta t/0) + \psi_o^{(B)2}(t + \Delta t) - \mu \left[\bar{\lambda}_L \right]^2 \zeta(0), \\
 \psi_o^{(B)2}(t + \Delta t) &\rightarrow e^{-k_{ns}\Delta t} \psi_o^{(B)2}(t) + \frac{1}{2} \left[\left(\frac{n_{ns}}{n_s} k_s e^{-k_{ns}\Delta t} \mu \left[\bar{\lambda}_L \right]^2 \zeta(t + \Delta t/t) \right) \right. \\
 &\left. + \left(\frac{n_{ns}}{n_s} k_s \mu \left[\bar{\lambda}_L \right]^2 \zeta(t + \Delta t/t + \Delta t) \right) \right] \Delta t.
 \end{aligned} \tag{2.38}$$

2.6.3 Verification of VUMAT-VUEL-VUSDFLD and numerical update procedure for $\psi_o^{(B)}$

For homogeneous deformation, $\Delta d = 0$ and (2.33) simplifies to

$$\zeta \dot{d} = 2(1 - d) \max_{s \in [0, t]} \left[\langle \psi_o^+(s) - \psi_{cr}^+ \rangle \right] - 2\psi_* d. \quad (2.39)$$

A one-dimensional implementation of the damage model in MATLAB can then serve as a benchmark for verifying VUMAT-VUEL-VUSDFLD. Following the discussion in Chapter 1 (Section 1.5.1), the MATLAB implementation will also serve as a benchmark for the $\psi_o^{(B)}$ numerical update procedure verification. Figure 2.5 shows the comparison of one-element (unit cube) simple compression (at 1 s^{-1} true strain rate) simulation results for ψ_o and d ($\psi_{cr}^+ = 0.025 \text{ MPa}$, $\psi_* = 0.025 \text{ MPa}$, $\zeta = 1 \text{ kPa.s}$) with the MATLAB predictions, showing good agreement.

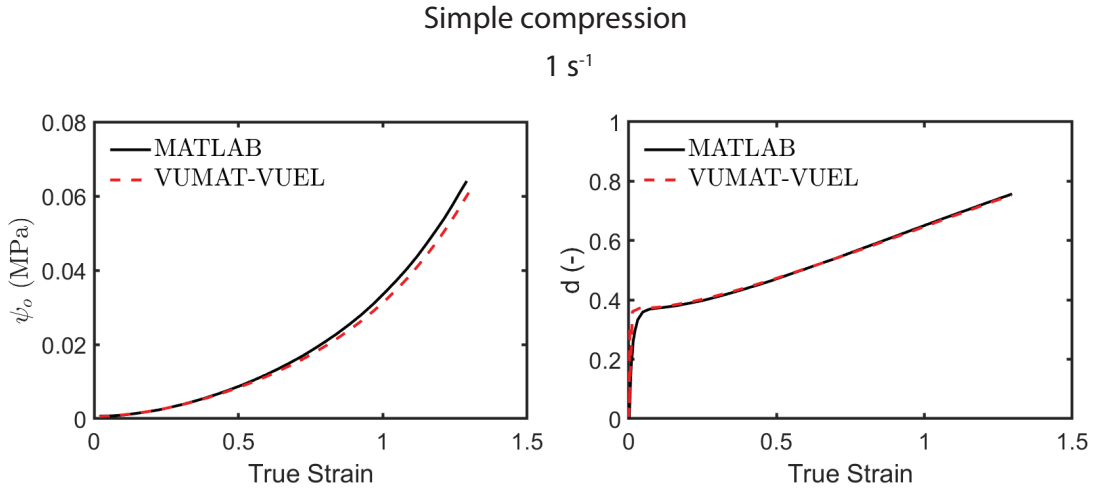


Figure 2.5: **VUMAT-VUEL-VUSDFLD and $\psi_o^{(B)}$ numerical update procedure verification.** Good agreement of one-element (unit cube) simple compression (at 1 s^{-1} true strain rate) simulation (using VUMAT-VUEL-VUSDFLD) results for ψ_o and d ($\psi_{cr}^+ = 0.025 \text{ MPa}$, $\psi_* = 0.025 \text{ MPa}$, $\zeta = 1 \text{ kPa.s}$) with the benchmark MATLAB predictions.

2.6.4 Finite element model reduction using symmetries

For all simulations in this chapter, the bottom surface of the specimen is completely fixed, i.e., $u_X = u_Y = u_Z = 0$. u denotes the translational degrees of freedom of a material point about the axis indicated in the subscript. The Z and Y axes are parallel to the thickness and height of the specimen, respectively. For quarter block simulations, $u_X = 0$ is prescribed on the symmetry surface with normal along the X axis. For both quarter and half block simulations, $u_Z = 0$ is prescribed on the symmetry surface with normal along the Z axis. Finally, for all simulations, u_Y corresponding to the loading profile is prescribed along with $u_X = u_Z = 0$ on the specimen's top surface.

2.7 Model parameter calibration for PBS

Values for all the deformation-associated parameters in (2.35) are taken from the previous chapter. Uniaxial homogeneous tension tests can be used for evaluation of W_{cr} , ψ_{cr}^+ , and $\psi_{cr}^{(B)}$.⁸ Dogbone tensile PBS specimens of ASTM D638 Type V standard [gauge dimensions: 9.5 ± 1 mm x 3.2 ± 1 mm x 3.2 ± 1 mm] (International, 2015) were subjected to a stretching speed of 60 mm/s. Displacement was monitored through markers at the two extremes of the gauge length and standard image processing. (2.1) for this loading can be written as

$$W(X_i, t) = \int_0^t s \dot{\lambda} dt \quad (2.40)$$

where s , λ are the engineering stress and gauge section stretch in the direction of loading, respectively.⁹ $\dot{\lambda}$ was evaluated to be 4 s^{-1} and the corresponding σ vs. t curve is shown

⁸Soft polymers exhibit fast damage evolution through sharp load drops corresponding to fracture (Liu et al., 2019b; Zhao et al., 2020b). Hence, W_{cr} (note that it accounts for both energetic and dissipative contributions) for soft polymers will be approximately equal to the value of W at damage initiation ($\psi_0^+ = \psi_{cr}^+$). For hyperelastic materials, $W_{cr} = \psi_{cr}^+$ (the energetic contribution) since there are no viscous contributions to W_{cr} . $W_{cr} > \psi_{cr}^+$ for materials with viscous effects.

⁹Analytical expressions for W_{cr} in the case of uniaxial tension can be obtained for hyperelastic materials

in Figure 2.6(A). $W_{cr} = 0.6$ MPa was calculated using (2.40) with the integration carried till the time instant of complete failure. A one-dimensional version of the model without considering damage was implemented in MATLAB to simulate this experiment and calculate ψ_o^+ and $\psi_o^{(B)}$. $\psi_{cr}^+ = 0.27$ MPa, $\psi_{cr}^{(B)} = 0.12$ MPa were obtained as the values of ψ_o^+ , $\psi_o^{(B)}$ at complete failure.

Although the parameters ψ_* , l , ζ can be fit to the uniaxial tensile test, sensitivity to damage gradients can only be obtained with inhomogeneous deformations, e.g., the notched tension test in Figure 2.6(B). The finite element mesh with a total of 32,468 C3D8R (three-dimensional, reduced integration) stress elements used for simulating the notched tension experiment is shown in Figure 2.6(B). In all the simulations in this chapter, a finer mesh was applied in the regions surrounding the notch to capture the stress and strain gradients. Several works in literature have studied failure in soft polymers experimentally (Yin et al., 2021; Li et al., 2021) and theoretically (Long and Hui, 2015; Qi et al., 2018) by considering a macroscopic fracture process zone. The l value based on the length scale of the fracture process zone in a gradient-damage-based model for elastomers (similar to the gradient-damage model in this chapter but limited to hyperelastic materials) has been shown to predict size-dependent fracture (Lee et al., 2024). The actual value of the length scale associated with the macroscopic fracture process zone in PBS has not been experimentally measured so far. Hence, the parameter l is considered a regularization parameter here. $l = 2.5$ mm was chosen to result in a computationally tractable mesh given the specimen dimensions while following the restriction of $l \gtrsim 5 \times$ typical element size (h_e) (Narayan and Anand, 2021) and being in the range of fracture process zone length scales reported for soft polymers in literature (Matsuda et al., 2020; Long et al., 2021; Sloodman et al., 2022). ψ_* and ζ were calibrated to fit the load-displacement curve after the load maximum in the notched tension calibration experiment. The model fit to the experimental

using (2.40). E.g., if neo-Hookean model is used: $W_{cr} = \mu(\frac{\lambda_{cr}^2}{2} + \frac{1}{\lambda_{cr}} - \frac{3}{2})$, if Mooney-Rivlin model is used: $W_{cr} = 2C_1(\frac{\lambda_{cr}^2}{2} + \frac{1}{\lambda_{cr}} - \frac{3}{2}) + 2C_2(\lambda_{cr} + \frac{1}{2\lambda_{cr}^2} - \frac{3}{2})$ where C_1 , C_2 are material constants and the ground state shear modulus $\mu = 2(C_1 + C_2)$.

extension force-displacement curve corresponding to the notched tension test is shown in Figure 2.6(B). Values of the model parameters are listed in Table 2.1. For PBS, we assume $k_{ns} = k_s = k$, shown to work reasonably well in the previous chapter.

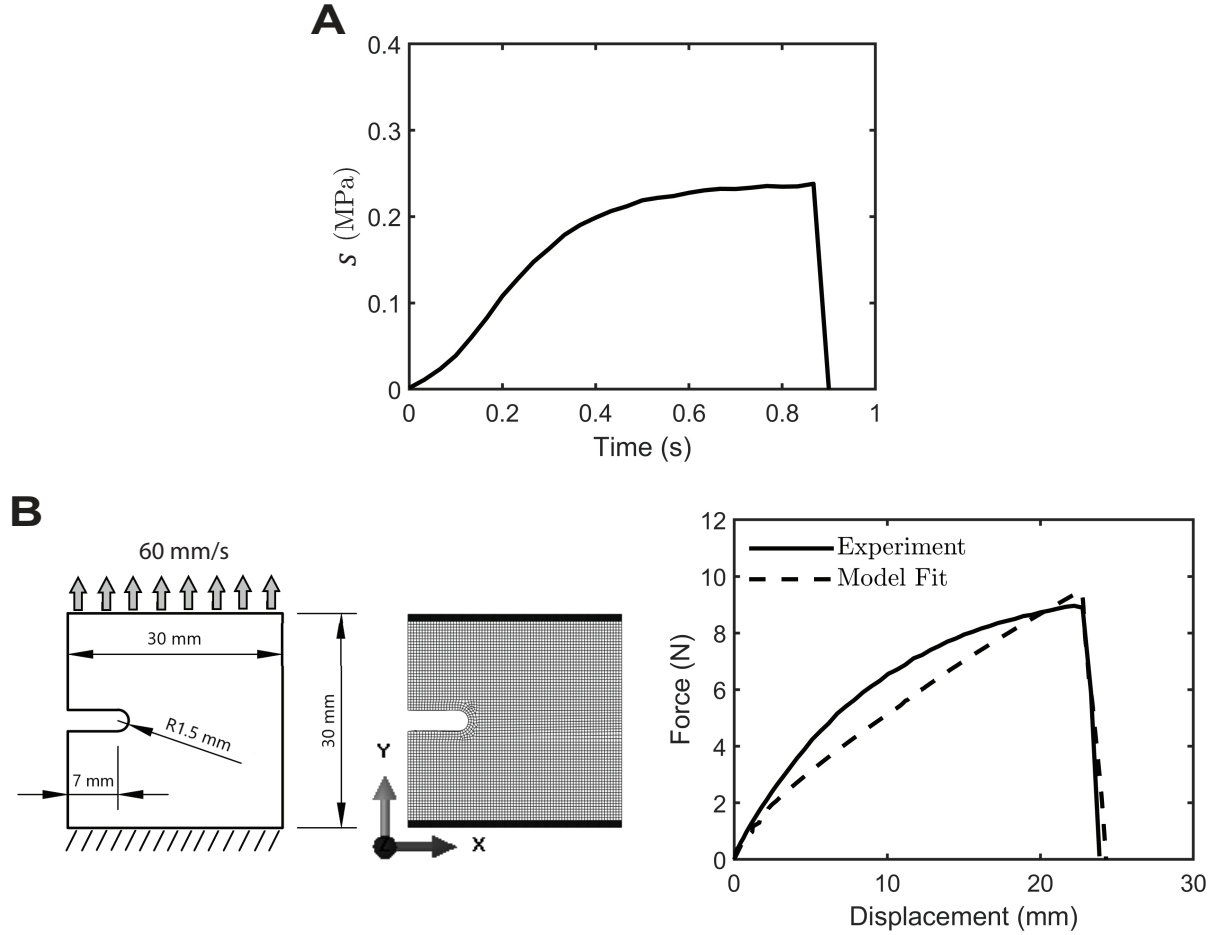


Figure 2.6: **Fracture model fit to experiments for PBS.** (A) The engineering stress (s) vs. time curve for the uniaxial tension experiment on PBS. The gauge section of the specimen undergoes stretching at an engineering strain rate of 4 s^{-1} . (B) The schematic for the notched PBS tension experiment. The specimen has a thickness of 3.2 mm and was subjected to a loading speed of 60 mm/s. The finite element mesh used for the simulation. The fit of the model (dashed lines) to the experimental extension force-displacement curve.

Deformation		Damage	
Parameter	PBS	Parameter	PBS
k (s ⁻¹)	0.35	ψ_{cr}^+ (MPa)	0.27
n_s (m ⁻³)	6.4 x 10 ²⁴	ψ_* (kPa)	4
G (MPa)	0.4	l (mm)	2.5
K (MPa)	20	ζ (kPa.s)	1.1
ν_0 (s ⁻¹)	2 x 10 ⁻³		
m (-)	0.95		
α_p (-)	0.11		
S_0 (kPa)	0.6		
h (kPa)	37.7		
$\bar{\lambda}_L$ (-)	37.4		

Table 2.1: **Fracture model parameters for PBS.**

2.8 Model predictions for fracture in a variety of soft polymers

The model is applied to predict fracture in select soft polymers - a permanently crosslinked rubber [ethylene propylene diene monomer (EPDM)], PBS, and dynamically crosslinked EPS25 vitrimer. Also, we show that macroscopic model parameters for PBS can estimate subchain dissociation energy, which is a fracture microstructural process-related quantity. Three independent experiments were performed on PBS with different specimen geometries and loading configurations subjected to a range of stretching speeds. The previously calibrated model parameters for PBS obtained using the notched tension experiment in Figure 2.6(B) were kept fixed for independent validations. This allowed for an independent, true assessment of the model's predictive capabilities for the three validation experiments on PBS. Experimental data reported in (Benvidi et al., 2019) and (Shen et al., 2021) is used to test the predictive abilities of the proposed model for EPDM rubber and EPS25 vitrimer, respectively. Based on the experimental material response, intermolecular resistance (mechanism *A*) is neglected for modeling the deformation of EPDM rubber and EPS25 vitrimer in (Benvidi et al., 2019; Shen et al., 2021), i.e., $\psi_o^{(A)} = 0$ and $\psi_o = \psi_o^{(B)}$. For the network resistance (mechanism *B*), the free energy

density functions with the dynamic crosslink formulation for the hyperelastic function part were the same as used in (Benvidi et al., 2019; Shen et al., 2021) for EPDM rubber, EPS25 vitrimer. This also allowed us to test the dynamic crosslink formulation's predictive capabilities for different hyperelastic free energy density base functions. Deformation-related material parameters for EPDM and EPS25 vitrimer are taken from (Benvidi et al., 2019; Shen et al., 2021).

2.8.1 PBS: Estimate of subchain dissociation energy using ψ_{cr}^+

The referential number density of subchains in PBS surviving from $t=0$ till any time instant t ($n_s(t/0)$) can be obtained using the first order kinetics of underlying dynamic reactions (Konale et al., 2023) as equal to $n_s e^{-k_{ns}t}$. At the time instant corresponding to the load drop in Figure 2.6(A) for the uniaxial tension test, with $k_{ns} = 0.35 \text{ s}^{-1}$ and $n_s = 6.4 \times 10^{24} \text{ m}^{-3}$, $n_s(t/0)$ is equal to $4.7 \times 10^{24} \text{ m}^{-3}$. These subchains experience the highest level of stretch and, hence, will be most likely to undergo stretch-induced dissociation. Hence, it can be assumed that the energy associated with the dissociation of $4.7 \times 10^{24} \text{ m}^{-3}$ subchains at a material point is $\psi_{cr}^{(B)}$. The subchain dissociation energy can then be estimated as 15 kJ/mol. We independently conducted oscillatory rheology experiments on PBS at temperatures of 25 °C, 37 °C, and 50 °C, shown in Figure 2.7(A) till 1 % strain. The experiments were performed using the ARES G2 Rheometer and the cone and plate geometry (40 mm diameter and 0.04 radians cone angle). Three repetitions were performed for each experimental result reported. In the previous chapter, it was established that the crossover frequency in small-strain frequency sweep experiments is proportional to the backward reaction rate parameter (k_{ns}) for the reversible reaction involving subchain dissociation in (2.26). Assuming Arrhenius temperature dependence of k_{ns} , the activation energy for the backward reaction, i.e., subchain dissociation, can be obtained by fitting to the crossover frequencies in Figure 2.7(B) as 24 kJ/mol, which is in the same order of magnitude as the subchain dissociation

energy (15 kJ/mol). This same order of magnitude similarity between activation energy and subchain dissociation energy is reasonable considering (a) the relative simplicity of the chemical reaction under consideration - only one type of bond (B:O coordinate-bond) is involved, and (b) the assumptions made during the estimation analyses. Further, the subchain dissociation energy value lies in the range of weak, non-covalent bond energies reported in the literature (Burley and Petsko, 1988; Wang et al., 2017; Buaksuntear et al., 2022). A physical understanding of the macroscopic fracture model parameters can thus provide reasonable approximations for fracture-related microstructural processes.

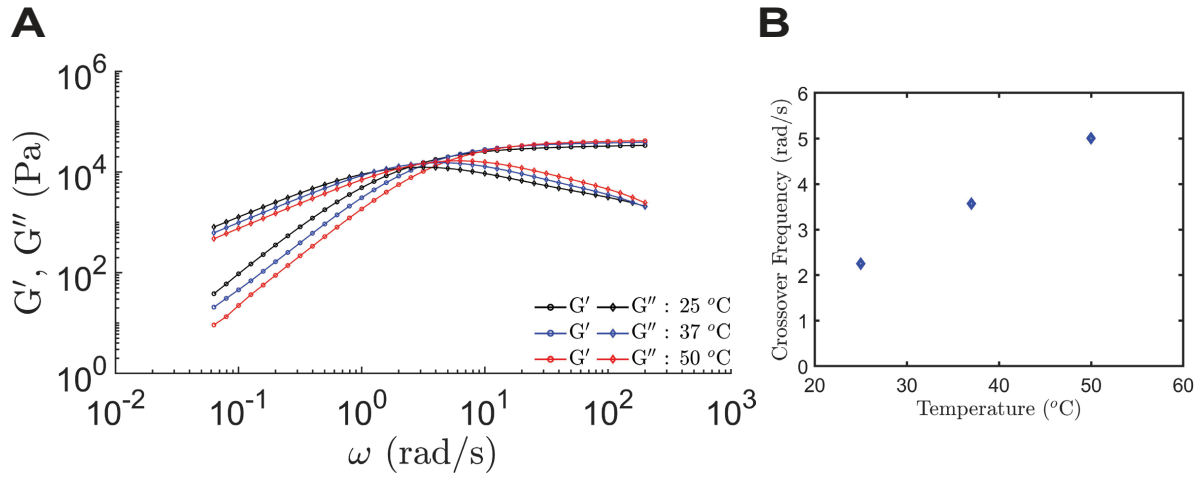


Figure 2.7: **Oscillatory rheology experiments on PBS to validate subchain dissociation energy estimate obtained using ψ_{cr}^+ .** (A) The oscillatory shear response of PBS (1% shear strain) at 25 °C, 37 °C and 50 °C. (B) The crossover frequency vs. temperature plot for PBS.

2.8.2 PBS: Single edge notch test for validation

A U-shaped edge notch with depth and radius of 18.5 mm and 3 mm, respectively, was created equidistant from the gripping fixtures in the gauge section with dimensions 30 ± 2 mm x 30 ± 2 mm x 3.2 ± 1 mm. This is a much deeper notch than what was used for calibration. The specimen geometry for the experiment schematic shown in Figure 2.8(A) was stretched at 60 mm/s. The finite element mesh (31,892 C3D8R elements) used for simulation is shown in Figure 2.8(A). The specimen was stretched until failure. Good agreement between the prediction from simulation and experimental result for the

extension force can be seen in Figure 2.8(B). Specimen geometry changes at different displacement levels from the experiment and simulation are shown in Figure 2.8(C), which also shows very good agreement between the experimental and simulation results. Particularly, the trumpet fracture profile (formation of a second local notch ahead of the original blunted notch) observed experimentally in Figure 2.8(C)(iv) is predicted by the model. The model correctly predicts the two cases of rupture with low extensibility at a fast loading rate (60 mm/s) and high extensibility at a slow loading rate (6 mm/s), as shown in Figure 2.8(C). The high extensibility at the relatively slower loading speed of 6 mm/s is due to ψ_o^+ , $\psi_o^{(B)}$ at the notch tip not being able to exceed ψ_{cr}^+ and $\psi_{cr}^{(B)}$, respectively.

Superposition of experimental and model-predicted specimen shapes

The developed deformation (in Chapter 1) and fracture models can be further validated by directly comparing model-predicted specimen shapes with experimental results. Specifically, the experimental and model-predicted edge profiles for the two single-edge notched geometries (notch depths of 8.5 and 18.5 mm) considered in this chapter are superposed at the deformation-only and complete rupture stages. Figure 2.9 shows the good agreement between the model predictions and experimental results, providing additional validation for the deformation and fracture models.

Energy release rate: evaluation path and loading rate dependence

The energy release rate (ERR) for soft polymers can depend on the evaluation path and loading rate, as discussed in Section 2.3. To demonstrate this, the referential integral

$$ERR = \int_{\Gamma} \left(W n_1 - \frac{\partial u_i}{\partial X_1} P_{ij} n_j \right) d\Gamma \quad (2.41)$$

was evaluated over various closed paths Γ enclosing the notch and stretching speeds (loading rates) for the PBS single-edge notch geometry in Figure 2.6(B) [notch depth = 8.5

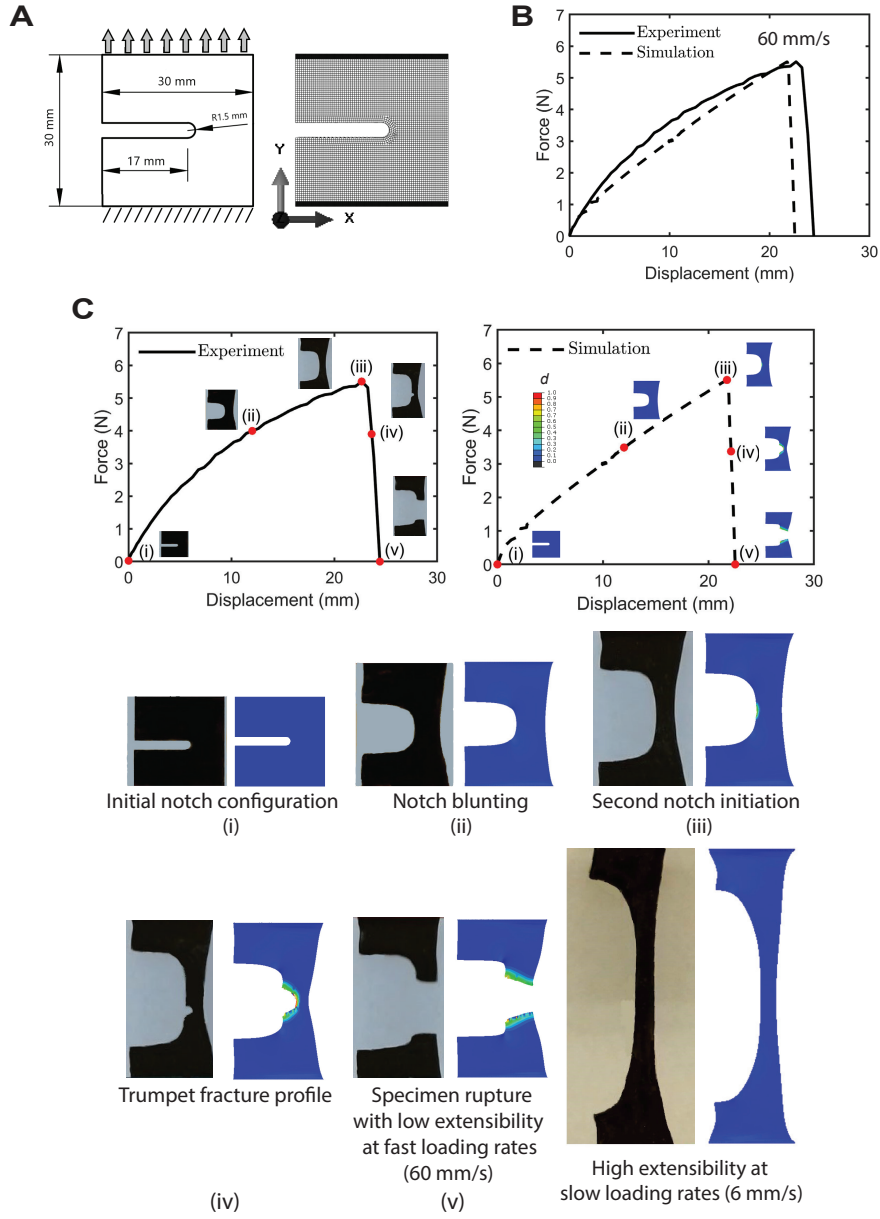


Figure 2.8: **Predictive ability of the fracture model for PBS (forces and rate-dependent physical response).** (A) The edge notch PBS stretch experiment schematic. The finite element mesh used for the simulations. (B) A comparison between the numerically predicted force-displacement curve and the experimental result for 60 mm/s loading speed shows good agreement. (C) The specimen shapes at different levels of stretch from the experiment (left) and the simulation (right) show good agreement for the 60 mm/s stretching speed. The model predicts the experimentally observed trumpet fracture profile. Also, the model predicts rupture with low extensibility at a fast loading rate (60 mm/s) and high extensibility at a slow loading rate (6 mm/s). Elements with $d > 0.95$ are hidden, and the experimental specimen was colored for visualization.

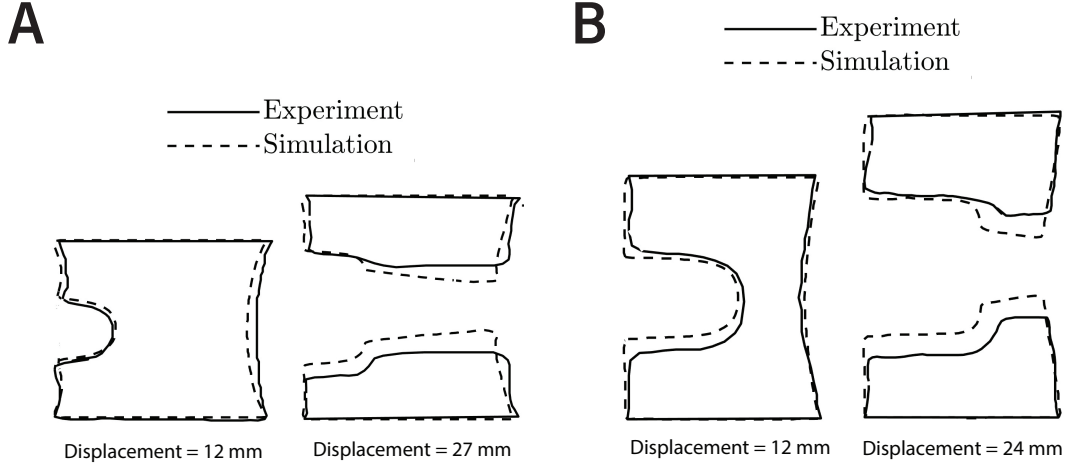


Figure 2.9: **Direct comparison of experimental and model-predicted specimen shapes.** Superposition of experimental and model-predicted edge profiles at deformation-only and completely ruptured stages for the single-edge notched geometries with notch depths **(A)** 8.5 mm and **(B)** 18.5 mm. Good agreement of model predictions with experimental results further validates the developed deformation and fracture models.

mm] using simulations. It can be noted that for traction-free edges (e.g., notch edges), the second term in the integrand in (2.41) is equal to zero. Except for the notch arc, the path segments were chosen to align with the x ($n_1 = 0, n_2 = \pm 1$) or y ($n_1 = \pm 1, n_2 = 0$) axes. For the integral evaluation, W , $(F_{11} - 1)P_{11} + (F_{21} - 1)P_{21}$ and $(F_{11} - 1)P_{12} + (F_{21} - 1)P_{22}$ were stored as state-dependent variables (SDVs). The values of these SDVs were extracted from the nodes along the evaluation path to calculate the ERR. Since the values of these SDVs were approximately uniform across the thickness, ERR was evaluated on the mid-plane for this exercise. The time instants at which damage initiates for each loading speed are considered.

Figure 2.10 shows the ERR values for four different paths (dashed lines) and a stretching speed of 60 mm/s. The path dependence of ERR values is evident. For path 2, ERR was also evaluated at loading speeds of 30 mm/s and 15 mm/s to highlight its rate dependence. An interesting observation is that for paths big enough to encompass stress concentration, ERR is approximately equal to W_{cr} (0.6 MPa) times the specimen thickness (3.18 mm). Because ERR quantifies the energy released per unit crack area growth, dividing it by the

thickness approximately quantifies the energy released per unit volume of crack extension. Although not directly equal to W_{cr} , ERR divided by thickness has a similar physical meaning. Hence, the observation is not surprising.

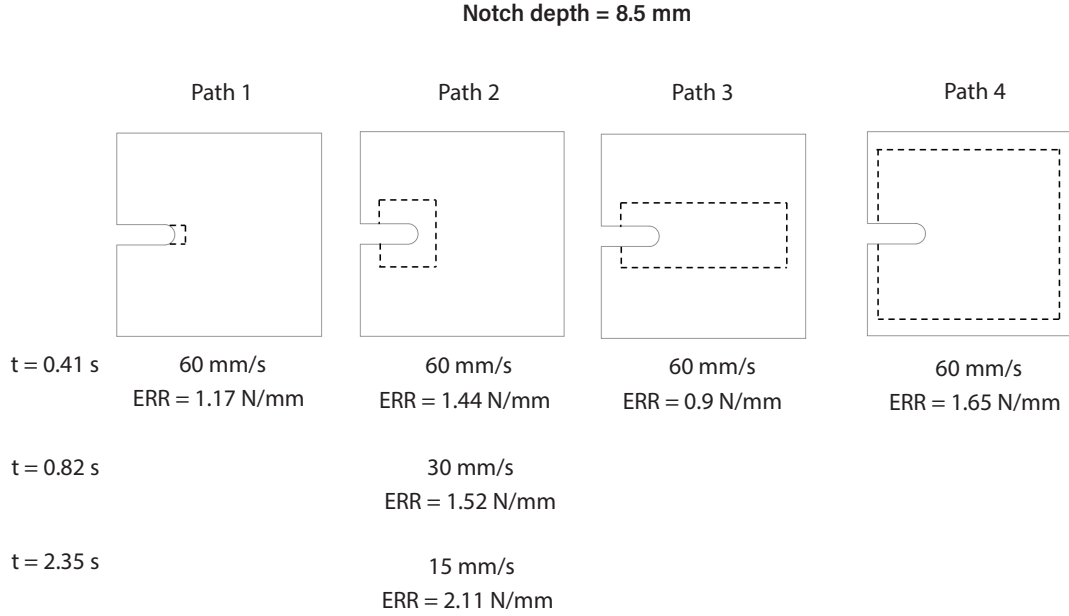


Figure 2.10: **Energy release rate (ERR): evaluation path and loading rate dependence.** ERR values for four different paths (dashed lines) and three stretching speeds (15 mm/s, 30 mm/s, 60 mm/s) of PBS single-edge notch geometry (notch depth = 8.5 mm) specimens using simulations. SDVs for ERR calculation were approximately constant with thickness; hence, ERR was evaluated on the mid-plane. ERR varies significantly with both evaluation path and loading rate. ERR values for paths large enough to encompass stress concentrations are approximately equal to W_{cr} (0.6 MPa) times the thickness (3.18 mm), consistent with the physical interpretations of ERR and W_{cr} .

2.8.3 PBS: Center notch for validation

To validate the model predictions for PBS in a scenario involving a different notch geometry (same gauge section dimensions and boundary conditions as the single edge notch geometry), a 10 mm x 3 mm slot was created at the center of the gauge section, as shown in Figure 2.11(A). Further, relatively low loading speeds of 2 mm/s and 6 mm/s were used for this geometry. The finite element mesh used for the simulations is shown in Figure 2.11(B). One-quarter of the specimen (25,832 C3D8R elements) was simulated with appropriate boundary conditions considering the symmetries of the problem. For these

relatively slower speeds, the experimental results exhibit necking instability in the form of the non-positive slope of force-displacement curves after a certain level of displacement. This can be attributed to significant network reconfiguration occurring over these long timescales (relative to the B:O coordinate bond dynamic crosslink macroscopic relaxation timescale) due to dynamic crosslink relaxation and reformation. Model predictions for the force-displacement curves, particularly the displacement levels for the onset of necking instability, agree well with the experimental results as seen in Figure 2.11(C). Fracture does not occur for these slow loading speeds which can be attributed to ψ_o^+ and $\psi_o^{(B)}$, not being able to exceed ψ_{cr}^+ and $\psi_{cr}^{(B)}$, respectively at the notch tip. The specimens exhibited high extensibility in the experiments, as shown in Figure 2.11(D) by comparing the undeformed and deformed (at $t = 45$ s) shapes from the experiment for the 2 mm/s loading speed. Figure 2.11(E) shows the simulation-predicted deformed shapes at different levels of stretch corresponding to the stretching speed of 2 mm/s.

2.8.4 PBS: Stretching using inserts for validation

The fracture model is now applied to a different loading configuration. The PBS specimen has two 5 mm diameter symmetric holes in the gauge section (same dimensions as the single edge notch geometry) as shown in Figure 2.12(A). 3D-printed hard plastic (PLA) inserts of the same diameter placed in the holes are then used to stretch the specimen. The bottom insert is held fixed while the top insert is displaced at 60 mm/s. Figure 2.12(A) shows the experiment schematic. The finite element mesh used for the simulation is shown in Figure 2.12(B). One-quarter of the specimen (29,016 C3D8R elements) was simulated with appropriate boundary conditions considering the symmetries of the problem. The specimen was stretched until failure. The inserts were modeled as rigid analytical surfaces. Contact between the inserts and the specimen was assumed to be frictionless as the interfaces were lubricated in the experiments. Model prediction for the extension force agrees reasonably with the experimental result, as shown in Figure

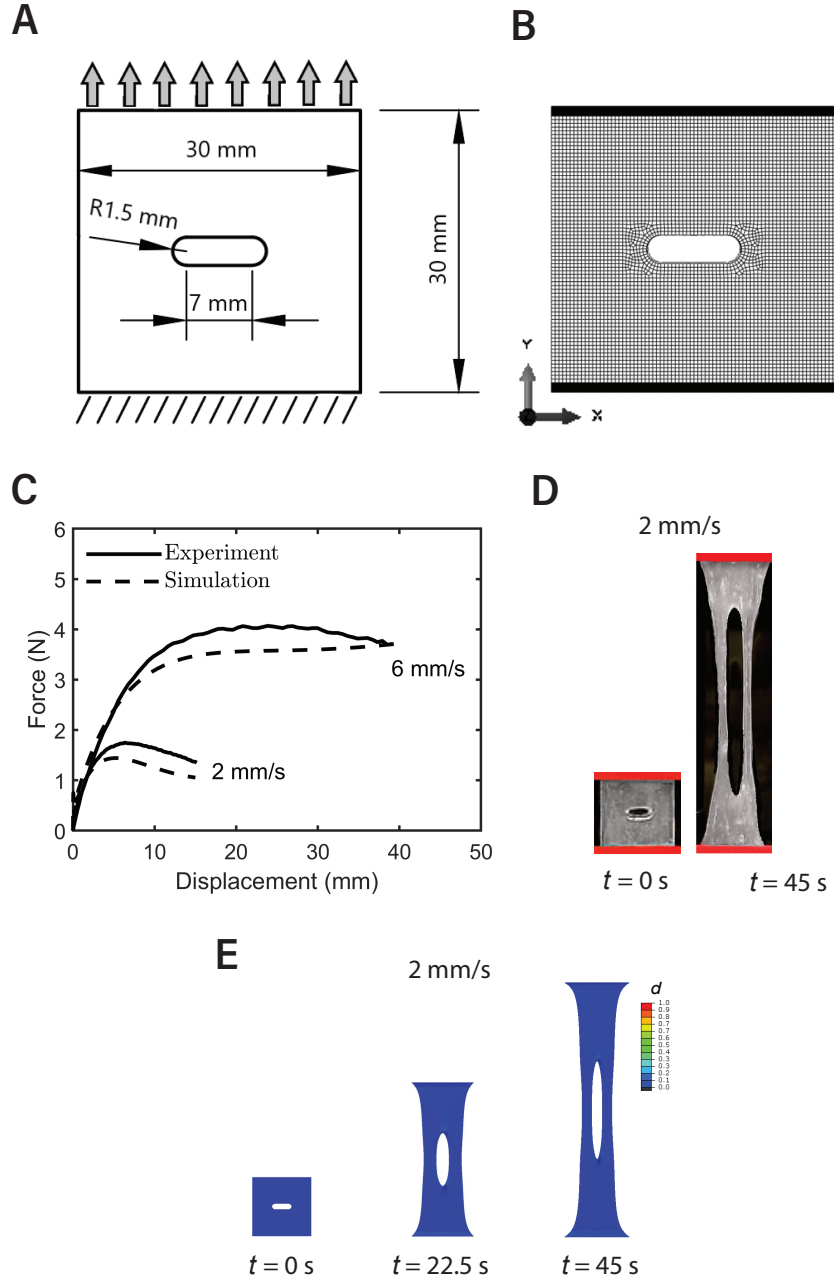


Figure 2.11: **Model predictions for center notch PBS specimen.** (A) The center notch PBS stretch experiment schematic. (B) The finite element mesh for the simulations. Quarter symmetry with appropriate symmetry boundary conditions was used in the simulations, but for clarity, the complete geometry with mesh is shown. (C) A good agreement between the model-predicted and experimentally measured forces can be seen for the two loading speeds of 2 mm/s and 6 mm/s. Necking instability seen in the experimental results is predicted by the model. Note that fracture does not occur at these slower loading rates. (D) The high extensibility is shown by comparing experimental specimen shapes at $t = 0$ s and $t = 45$ s. (E) The deformed shape predictions from the simulation for the 2 mm/s speed at different stretch levels.

2.12(C).

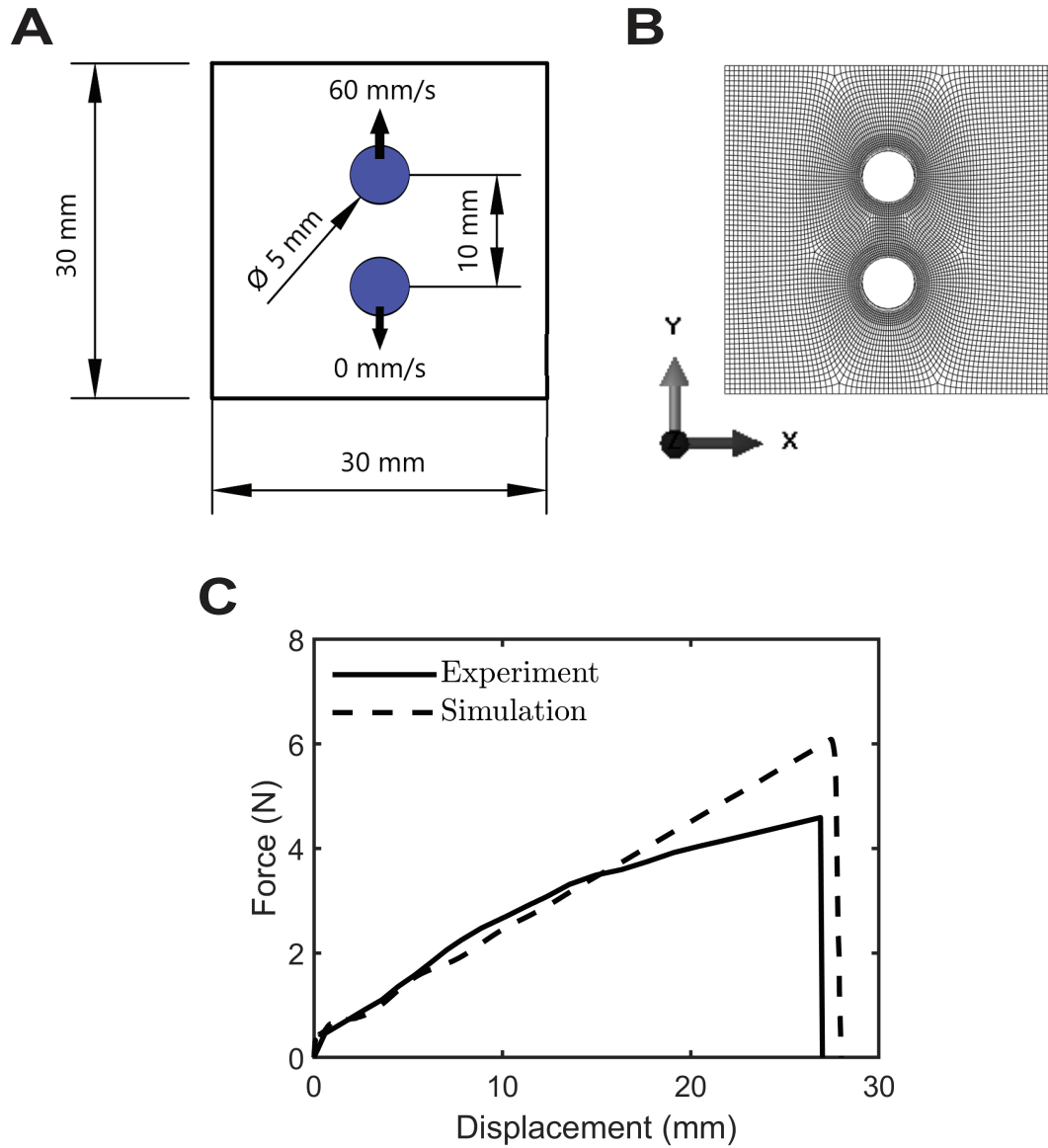


Figure 2.12: **Model prediction for stretching through internal holes in PBS specimen.** (A) The PBS specimen geometry and experiment schematic. The blue regions denote the inserts. The specimen has a thickness of 3.2 mm, and the top insert was displaced at a speed of 60 mm/s. (B) The finite element mesh used for the simulation. A quarter of the block is considered due to the symmetries of the problem with appropriate boundary conditions. For clarity, complete geometry with mesh is shown. (C) The model prediction for the extension force agrees well with the experimental result.

2.8.5 Fracture response considering viscous effects

We performed simulations where the viscous effects in the input material were removed (by suitably prescribing the material parameter values) to study the role of viscosity in the physical fracture phenomenon. These enabled qualitative model predictions for fracture profiles as the material varies from a viscous soft polymer to an elastomer. The geometry, loading schematic, and finite element mesh for the notched tension test shown in Figure 2.6(B) were considered with a stretching speed of 60 mm/s. The material parameters for PBS were used for the viscous soft polymer (except ζ) and elastomer (except G , k_{ns} , k_s , and ζ). We removed viscous effects by setting $G = 0$ (removing the elastic-viscoplastic A micro-mechanism) and $k_{ns} = k_s = 0$ (this provides a model representing permanently crosslinked material) to simulate a hyperelastic material. The parameter ζ , which determines the timescale for damage growth, was set equal to 3 kPa.s for both viscous soft polymer and elastomer for ease in elastomer numerical calculations. This numerical example shows differences in the general elastomer and viscous soft polymer fracture response. The computationally obtained fracture profiles in Figure 2.13 transition correctly from the experimentally observed trumpet profile in viscous soft polymers (Hui et al., 2022; Shen et al., 2021) to the single parabola-like profile in elastomers (Qi et al., 2019; Shrivastava et al., 2023).

The strains, and hence, the strain rates, are inhomogeneous around the notch tip. The simulations show that when there is local damage at the notch tip, the strains are further localized at the tip, and the strain rate increases significantly. In the case of rate-dependent viscous polymers, significantly higher strain rates around the notch tip increase the local stress, causing a small area near the notch tip to be damaged. This leads to the formation of a second, smaller notch and a trumpet-like profile that we observe in the experiments. Similar discussions about the trumpet fracture profile exist in the literature (de Gennes, 1996; Nagatakiya et al., 2023; Okumura, 2002a,b). On the other hand, the elastomers are rate-independent, and there is no additional rate-dependent

increase in stresses at the notch tip. As a result, the fracture progresses in a self-similar manner, i.e., the single parabola-like profile in elastomers. The model can predict the significantly varying important physical phenomena observed during the fracture of soft polymers.

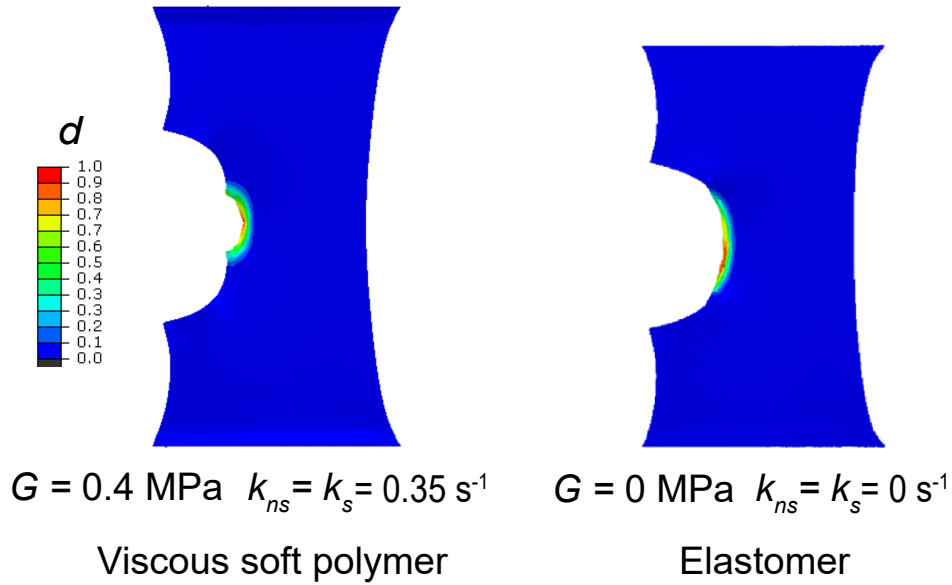


Figure 2.13: **Comparison of viscous soft polymer and elastomer fracture.** The notch profiles for the single edge notch specimen (Figure 2.6(B)) stretching simulations showing experimentally observed trumpet profile for a viscous soft polymer and a single parabola-like profile when viscous effects were removed in the model to simulate the elastomeric material response. The profiles are shown just before complete failure. Elements with $d > 0.95$ are hidden for visualization.

2.8.6 EPDM elastomer with V notch

Benvidi and co-workers (Benvidi et al., 2019) performed tension tests on V-notched EPDM rubber to demonstrate the efficacy of the average strain energy density criterion for fracture analysis of hyperelastic materials. We test our fracture model's ability to capture the experimental data reported in (Benvidi et al., 2019). The four notch configurations from (Benvidi et al., 2019) were applied in finite element simulations for our model's verification. These configurations encompass variations in the depth and radius of the notch. One case (notch diameter $a=2$ mm, notch depth $d=5$ mm) was used for model parameter calibration. Fixing the values of these parameters, the remaining three cases

were used for independent model validation. The finite element meshes for $a=2$ mm, $d=5$ mm, and $a=2$ mm, $d=10$ mm cases are shown in Figure 2.14(A). A total of 20265, 20019, 20241, and 19746 C3D8R elements were used in the finite element meshes for $a=2$ mm $d=5$ mm, $a=2$ mm $d=10$ mm, $a=3$ mm $d=5$ mm, and $a=3$ mm $d=10$ mm cases, respectively.

Due to the assumed hyperelastic nature of EPDM in (Benvidi et al., 2019), $W_{cr} = \psi_{cr}^+ = \psi_{cr}^{(B)}$. The Ogden free energy density function was used in (Benvidi et al., 2019) to model the large deformation response of the EPDM elastomer. Instead of the eight-chain free energy density function with the dynamic crosslink formulation for Mechanism *B*, the Ogden function is used with the same dynamic crosslink formulation for our fracture modeling framework. We set $G = 0$ MPa, $k_{ns} = k_s = 0$ s⁻¹ in our model and used the deformation-related material parameters from (Benvidi et al., 2019). As the force-displacement curves for the V notched tests are not presented in (Benvidi et al., 2019), it was assumed that rupture occurs instantly when $\psi_o^+ = \psi_o^{(B)} = \psi_{cr}^+$ at the notch tip. Independent predictions for the displacement at which rupture occurs for the $a=2$ mm $d=10$ mm, $a=3$ mm $d=5$ mm, and $a=3$ mm $d=10$ mm notch geometries (variations in notch depth and radius) agree well with the experimental results as shown in Figure 2.14(B).

2.8.7 EPS25 vitrimer: Notched pure shear

To test the model predictions for a different dynamically crosslinked soft polymer and a loading mode other than tension, we utilize the data reported by Shen and co-workers (Shen et al., 2021) for the fracture of EPS25 vitrimer. Pure shear tests over a range of loading speeds were performed on EPS25 vitrimer specimens with sharp single-edge notches in (Shen et al., 2021). The specimen geometry is shown in Figure 2.15(A). The finite element mesh used for our simulations is shown in Figure 2.15(B). The crack extension vs. stretch ratio (λ) experimental data for a fast and a slow loading speed reported in (Shen

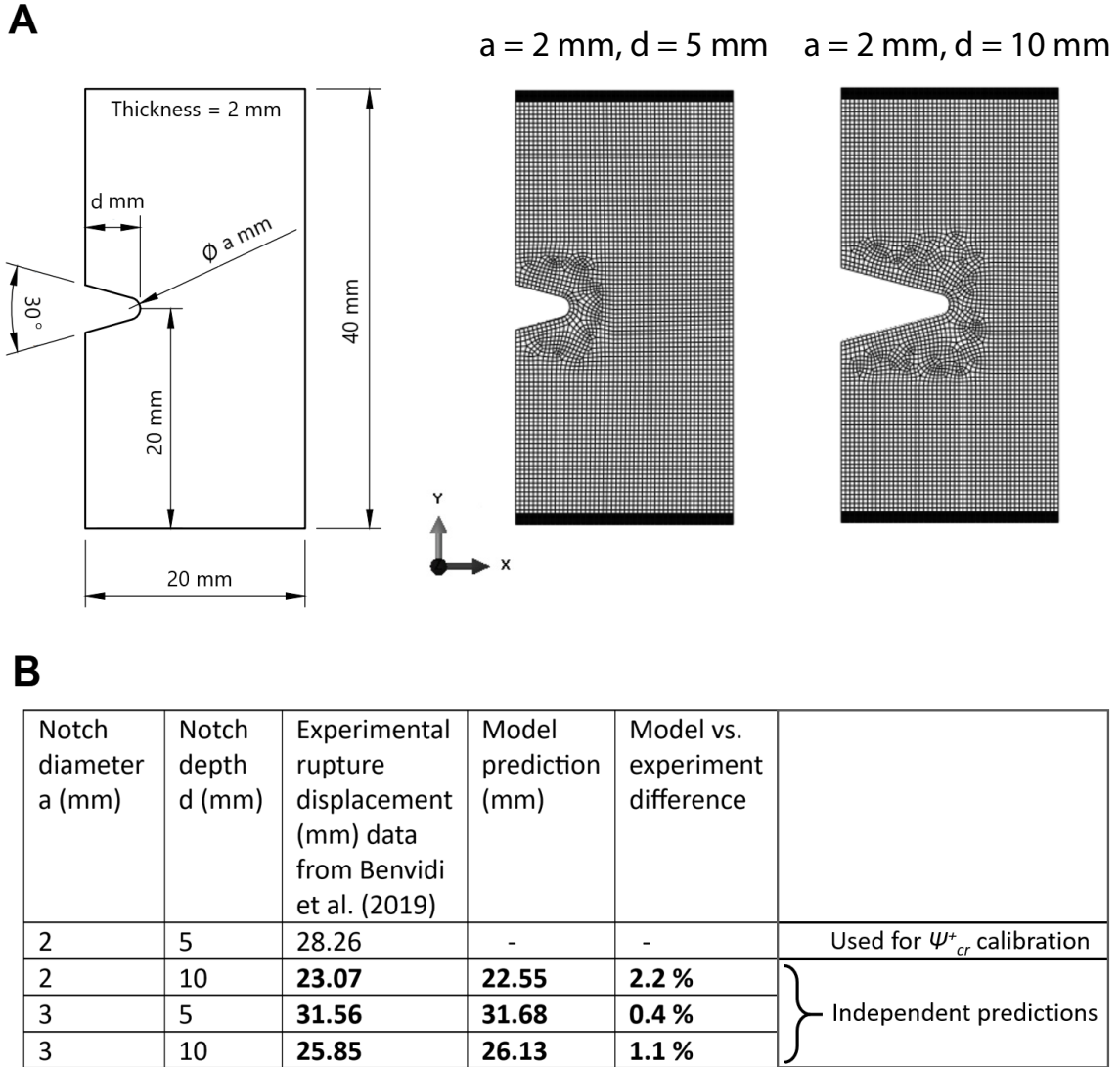


Figure 2.14: **Model prediction for fracture of EPDM elastomer.** (A) The V-notched specimen geometry for EPDM rubber used in (Benvidi et al., 2019). The finite element meshes used for the simulations of the $a=2 \text{ mm}$ $d=5 \text{ mm}$, $a=2 \text{ mm}$ $d=10 \text{ mm}$ notch geometries. (B) The experimental data for $a=2 \text{ mm}$, $d=5 \text{ mm}$ from (Benvidi et al., 2019) was used for $\psi_{cr}^+ = \psi_{cr}^{(B)} = W_{cr}$ calibration. Independent model predictions for the rupture displacements for $a=2 \text{ mm}$ $d=10 \text{ mm}$, $a=3 \text{ mm}$ $d=5 \text{ mm}$, and $a=3 \text{ mm}$ $d=10 \text{ mm}$ notch geometries show good agreement with the experimental results in (Benvidi et al., 2019).

et al., 2021) is shown in Figure 2.15(C). A transition from high extensibility, i.e., no crack propagation (0.0225 mm/s) to fracture (0.09 mm/s) with increasing loading rates was observed as seen in Figure 2.15(C). We used the neo-Hookean function for mechanism B with our same dynamic crosslink formulation. G is set equal to 0 MPa in our model, and the deformation-associated material parameters from (Shen et al., 2021) are used. In the

experimental work, the authors report creating a sharp notch in the specimens using a blade. As the exact radius of the notch is not specified in (Shen et al., 2021), we assumed a very sharp notch radius of 100 μm .

With the aim of modeling the initiation of crack propagation, only a VUMAT was used. A total of 28,716 C3D8 (three-dimensional, full integration) stress elements were used in the finite element mesh for the problem, as shown in Figure 2.15(B). $\psi_{cr}^+ = \psi_{cr}^{(B)}$, W_{cr} were evaluated as the values of $\psi_o^+ = \psi_o^{(B)}$, W at the notch tip at the time instant corresponding to the initiation of crack propagation for the 0.09 mm/s loading speed. Model predictions for ψ_o^+ at the notch tip vs. λ curves for the fast and slow loading speeds are shown in Figure 2.15(D). The model predicts no crack propagation for the relatively slow 0.0225 mm/s loading speed due to ψ_o^+ at the notch tip not being able to exceed ψ_{cr}^+ as seen in Figure 2.15(D). This agrees with the experimental observation from (Shen et al., 2021) in Figure 2.15(C). We also obtain the model prediction for the ψ_o^+ at the notch tip vs. λ curve at an intermediate loading speed (0.062 mm/s). For this intermediate speed, ψ_o^+ at the notch tip exceeds ψ_{cr}^+ after a certain stretch (point A in Figure 2.15(E)). However, due to subsequent significant network reconfiguration, the model predicts that ψ_o^+ drops below ψ_{cr}^+ after point B in Figure 2.15(E). The ψ_o^+ evolution at the notch tip represents crack nucleation (point A), propagation (point A to point B) when $\psi_o^+ > \psi_{cr}^+$, and subsequent end of propagation (point B) when $\psi_o^+ < \psi_{cr}^+$. The model prediction is in qualitative agreement with the experimental observation in (Shen et al., 2021), which showed that at an intermediate loading speed, the crack nucleated and propagated and later stopped propagating (arrested).

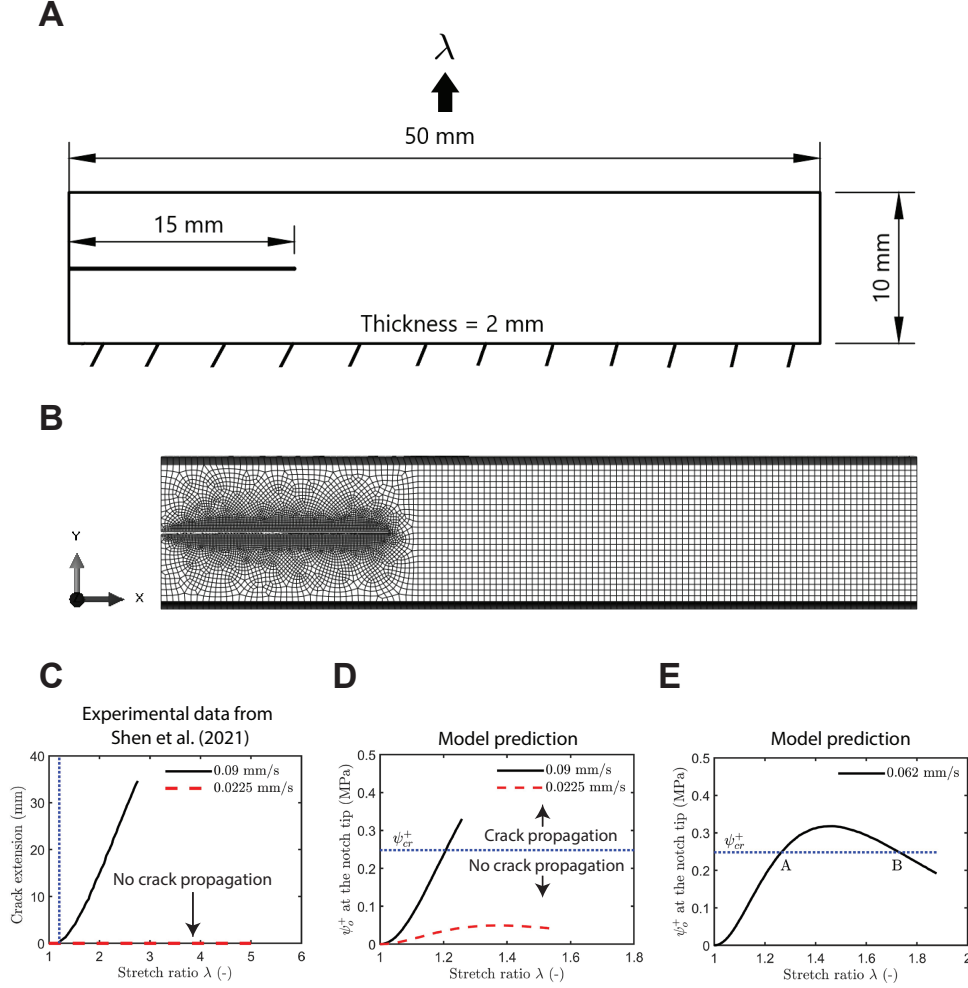


Figure 2.15: **Model prediction for fracture of EPS25 viscous soft polymer.** (A) The specimen geometry for EPS25 vitrimer pure shear tests with sharp single edge notches from (Shen et al., 2021). (B) The finite element mesh used for the simulations. (C) The crack extension vs. stretch ratio (λ) experimental data from (Shen et al., 2021) for the fast and slow loading speeds of 0.09 mm/s and 0.0225 mm/s. There is no crack propagation for the 0.0225 mm/s loading speed. $\psi_{cr}^+ = \psi_{cr}^{(B)}$ and W_{cr} were calibrated using the λ at initiation of crack propagation for 0.09 mm/s speed. (D) The model predictions for ψ_o^+ at the notch tip vs. λ curves for the fast and slow loading speeds. The model predicts no crack propagation for the 0.0225 mm/s speed due to ψ_o^+ at the notch tip not being able to exceed ψ_{cr}^+ . This agrees with the experimental observation in (Shen et al., 2021). (E) The model prediction for the ψ_o^+ at the notch tip vs. λ curve for an intermediate loading speed of 0.062 mm/s. The ψ_o^+ evolution at the notch tip represents crack nucleation (point A), propagation (point A to point B) when $\psi_o^+ > \psi_{cr}^+$, and subsequent end of propagation (point B) when $\psi_o^+ < \psi_{cr}^+$. This agrees with the experimental observation in (Shen et al., 2021), which showed that at an intermediate loading speed, the crack nucleated and propagated and later arrested.

2.8.8 SBR and NR: Failure surface predictions

Variational phase field models, which predict crack nucleation through minimization of a non-linear energy function without explicit criteria, are unable to capture the experimentally observed failure surfaces of various materials as discussed in the review article by Lopez and co-workers (Lopez-Pamies et al., 2025). We compare failure surface predictions from our model, which is based on the method of virtual power and considers an independent criterion for damage initiation, with the experimental measurements for SBR from (Hamdi et al., 2006) and NR¹⁰ from (Gent and Lindley, 1959). Specifically, our loading mode insensitive damage initiation criterion in the form of a unique critical free energy density ψ_{cr}^+ is used and validated. Due to the fast damage evolution of soft polymers, the failure occurs when $\psi_o^+ = \psi_{cr}^+$. The deformation responses of SBR and NR were modeled using the incompressible Ogden (N=2) hyperelastic function and Gent hyperelastic function, respectively, fit to the experimental data from (Hamdi et al., 2006). Unique ψ_{cr}^+ values were chosen based on the experimental failure data for both SBR and NR. Model predictions for the failure surfaces of SBR and NR using the proposed damage initiation criterion show good agreement with the experimental surfaces¹¹ as shown in Figure 2.16.

¹⁰A variational phase field model was shown in (Lopez-Pamies et al., 2025) to not be able to correctly predict the experimental data for SBR in (Hamdi et al., 2006) and NR in (Gent and Lindley, 1959). It showed unphysical predictions, specifically the multiaxial (biaxial for SBR, triaxial for NR) failure stress magnitudes to be larger than the uniaxial failure stress for both SBR and NR, with very significant deviations for NR.

¹¹It is prudent to note that the experimental fracture measurements in elastomers typically have a large variability and uncertainty, but the measures are useful to evaluate trends and obtain failure stress approximations within an order of magnitude.

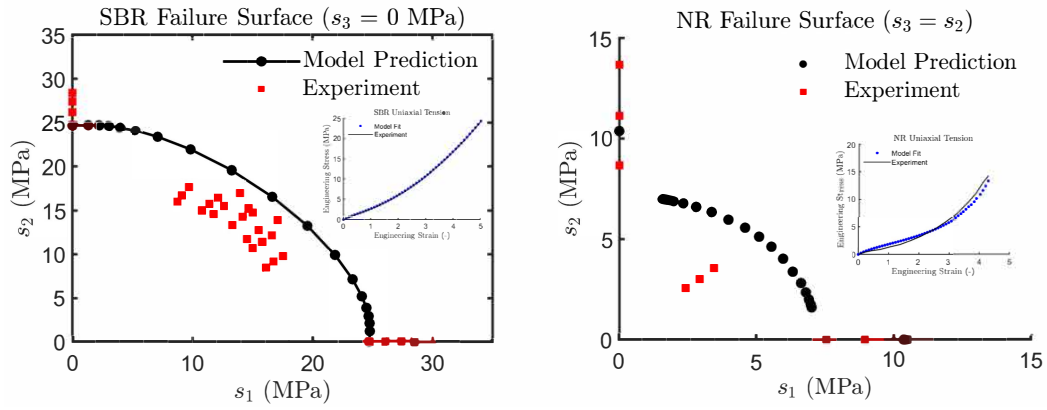


Figure 2.16: **Failure surface model predictions for SBR and NR.** The model predicts the failure surfaces for SBR ($s_3 = 0$ MPa) and NR ($s_3 = s_2$) reasonably well when its predictions are compared with the experimental surfaces from (Hamdi et al., 2006) and (Gent and Lindley, 1959). s_i are the nominal principal stresses. Insets (deformation response): Incompressible Ogden (N=2) hyperelastic model and Gent model fit to the experimental uniaxial stress-strain curves from (Hamdi et al., 2006) for SBR and NR, respectively. Unique values of ψ_{cr}^+ were chosen as model parameters for both SBR and NR.

2.9 Conclusion

Soft polymers are found in nature and as engineering materials, including elastomers, viscoelastic polymers, polymer gels, and biological tissues. Their properties vary from rate-independent to rate-dependent. Current fracture toughness measures are non-unique for rate-dependent soft polymers for varying loading profiles and specimen geometries. We propose and show that a critical value of stress work W_{cr} can quantify the fracture resistance of a certain class of soft polymers. W_{cr} can be interpreted as the total energy per unit referential volume associated with the complete failure of an arbitrary material point under a loading mode subjected to sufficiently fast loading rates for damage initiation.

We have developed a multi-mechanism, gradient-damage framework-based model to predict damage initiation, growth, and complete failure in soft polymers. The method of virtual power is used to formulate the gradient-damage framework. The energetic contribution to the critical stress work, ψ_{cr}^+ , is proposed as a damage initiation criterion that accounts for material strength. The proposed damage model shows successful predictions of PBS's fracture in several independent experiments involving different specimen geometries and loading conditions. It is also able to predict the fracture of three elastomers, EPDM, SBR, NR, and another viscous soft polymer, EPS25. A transition in fracture profile from trumpet to single parabola-like was observed in the model predictions as the polymer viscous effects were removed. This is consistent with experimental observations, demonstrating the model's ability to predict the significantly varying physical phenomena observed during the fracture of soft polymers. The continuum scale model is developed based on microscopic physical insights. We show that macroscopic model parameters can be used to obtain relevant physical quantities at the microstructural process scales. From the macroscopic parameter ψ_{cr}^+ , we estimated the value of subchain dissociation energy, which was verified by a set of independent experiments.

2.10 Further study: generally applicable damage initiation criteria for brittle materials

Since the evolution of damage in brittle materials (soft polymers are an example) is rapid (as observed experimentally through sharp load drops associated with fracture), fracture is primarily governed by damage initiation. Developing explicit, generally applicable criteria for damage initiation is an active research problem. The two current approaches were briefly discussed in Section 2.1. A Drucker-Prager type function that accounts for loading mode dependencies is used to construct a stress-based failure surface in the first approach. However, the stress-based approach lacks a physical interpretation and yields non-unique failure surfaces for rate-dependent materials. The free energy density (ψ) based approach is (i) physically interpretable - elastically stored energy supplied to a material point through mechanical forces and (ii) path-independent - can result in unique failure surfaces for rate-dependent materials. The experimental data from (Kamarei et al., 2026; Lopez-Pamies et al., 2025) is used to analyze the initiation of damage in a few materials using the critical free energy density criterion (ψ_{cr}^+).

2.10.1 How well can a unique ψ_{cr}^+ predict damage initiation under different loading modes?

A significant number of materials exhibit loading mode-based asymmetries. Hence, a ψ_{cr}^+ calibrated for one loading mode would be inaccurate to predict the onset of damage under different loading modes. This is tested using experimental data, material models, and parameters from (Kamarei et al., 2026) for two distinct materials.

Soda lime glass

- $\psi_{cr}^+ = 0.0115$ MPa (from uniaxial tension)
- Failure stretch prediction for biaxial loading = 1.0004, experimental value = 1.0003, % error = 0.0053 % (with respect to stretch), 17.5 % (with respect to strain)
- Failure twist prediction for torsion = 0.0015, experimental value = 0.0027, % error = 42.3%

Polyurethane (PU)

- $\psi_{cr}^+ = 0.0358$ MPa (from uniaxial tension)
- Failure stretch prediction for biaxial loading = 1.1198, experimental value = 1.12, % error = 0.02% (with respect to stretch), 0.19% (with respect to strain). Since PU is assumed to be compressible, the transverse stretch is obtained from the traction-free conditions.

The unique ψ_{cr}^+ approach may work for some cases, but a physics-motivated, general functional form for ψ_{cr}^+ with respect to the loading mode (I_1 , and/or J_2 of the stress tensor, alternatively Lode angle, J_3) will be impactful for broader applications.

2.10.2 Conversion of stress-based failure surfaces to ψ -based

Damage can be physically initiated only by ψ^+ . Under positive dilatation: $\psi^+ = \psi_o$ and under negative dilatation: $\psi^+ = \psi_o^{dis}$. Any valid ψ based criteria should account for this. The experimental stress-based failure surfaces for two materials (IG 11 Graphite and Titania) from (Lopez-Pamies et al., 2025) are used, along with the material parameter values for this conversion exercise; the results of which are shown in Figures 2.17 and 2.18.

Consistent observations for both materials are (i) the unique ψ_{cr}^+ approach fits well to the experimental data in the positive dilatation regime, (ii) when hydrostatic pressure

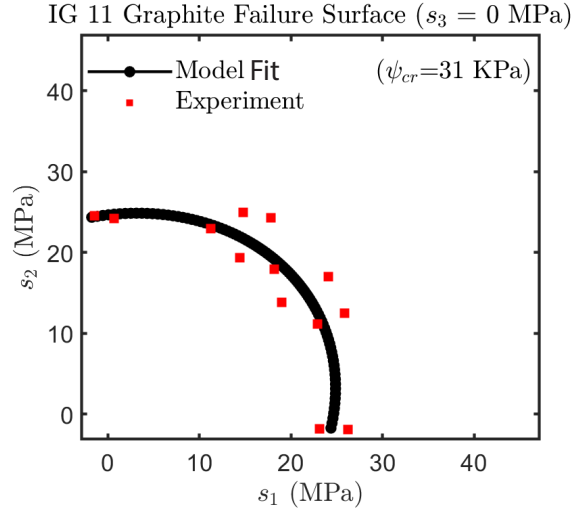
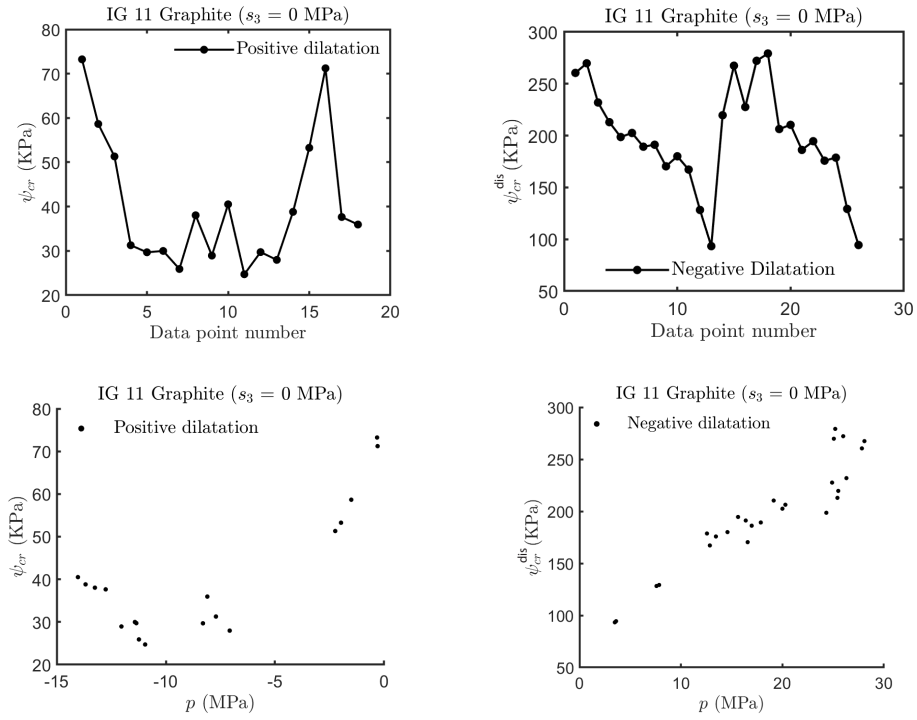
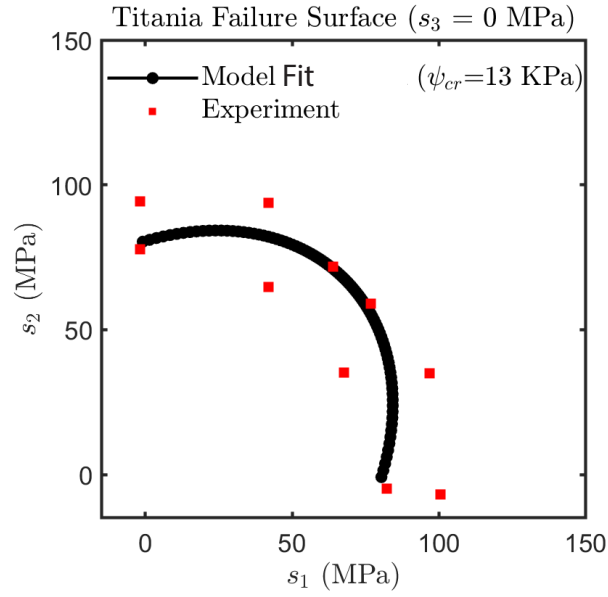
A**B**

Figure 2.17: **IG 11 Graphite: conversion of failure surface.** (A) Good fit of unique ψ_{cr} ($= 31$ kPa) model predictions to the positive dilatation experimental data. (B) Conversion of stress-based failure surface data to ψ_{cr} and ψ_{cr}^{dis} data (vs. data point number and hydrostatic pressure p). When $p > 0$ (hydrostatic compression), ψ_{cr}^{dis} increases with increasing p . When $p < 0$ (hydrostatic tension), ψ_{cr} decreases with increasing p magnitude.

A



B

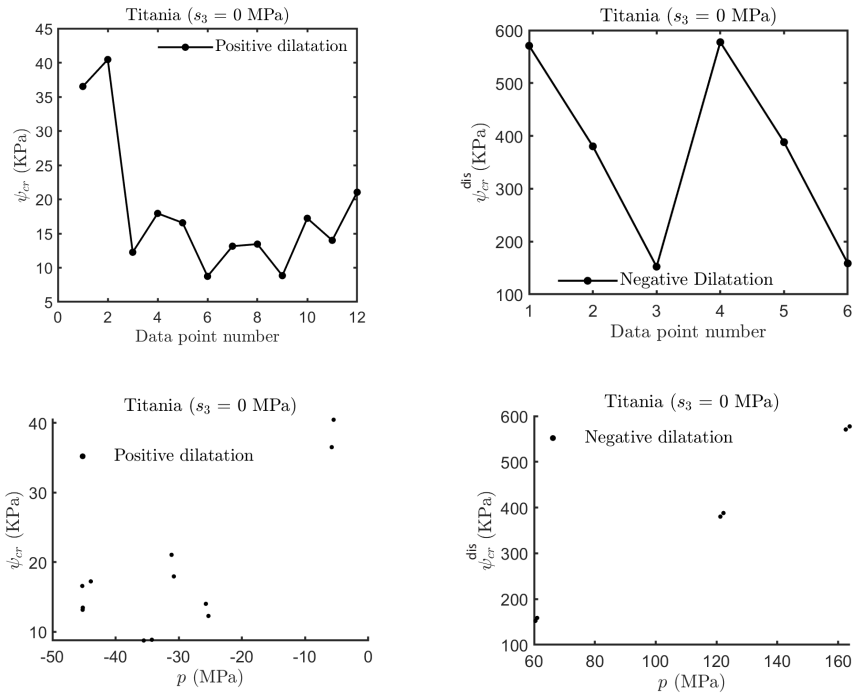


Figure 2.18: **Titania: conversion of failure surface.** (A) Good fit of unique ψ_{cr} ($= 13$ kPa) model predictions to the positive dilatation experimental data. (B) Conversion of stress-based failure surface data to ψ_{cr} and ψ_{cr}^{dis} data (vs. data point number and hydrostatic pressure p). When $p > 0$ (hydrostatic compression), ψ_{cr}^{dis} increases with increasing p . When $p < 0$ (hydrostatic tension), ψ_{cr} decreases with increasing p magnitude.

$p > 0$ (hydrostatic compression), ψ_{cr}^{dis} increases with increasing p and (iii) when $p < 0$ (hydrostatic tension), ψ_{cr} decreases with increasing p magnitude. The last two observations suggest that $\psi_{cr}, \psi_{cr}^{dis}$ have significant and intuitive dependence on p . Larger hydrostatic tension should make the damage initiation process easier (smaller ψ_{cr}). On the other hand, larger hydrostatic compression should make the damage initiation process more difficult (larger ψ_{cr}^{dis}). Microstructurally, this can be thought of as the bonds not being equally strong/susceptible to failure under all loading modes. Less strength under a loading mode implies less energy (ψ_{cr}^+) requirement and vice versa. The physical interpretability of the free energy-based approach is highlighted here. The unique ψ_{cr} model, as anticipated, does not extrapolate well to the negative dilatation regime as shown in Figure 2.19, emphasizing the need for better ψ -based damage initiation models.

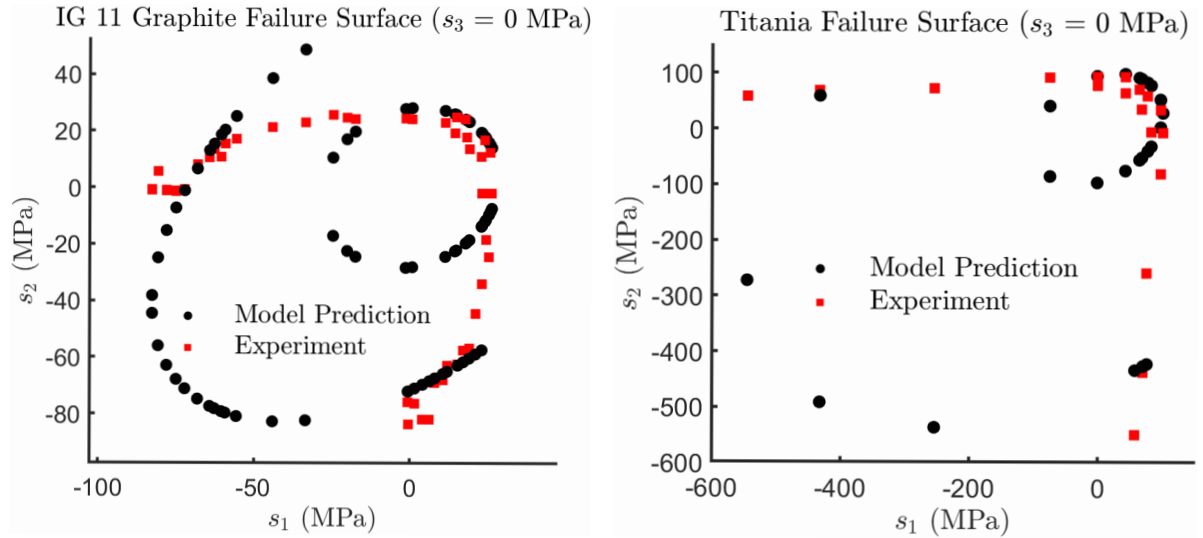


Figure 2.19: **IG 11 Graphite and Titania: extrapolation capabilities of the unique ψ_{cr}^+ model.** As anticipated, the unique ψ_{cr}^+ model does not extrapolate well to the negative dilatation regime for both materials. Both roots are plotted.

For linear elastic materials, the calibrated stress-based failure surface in (Kamarei et al., 2026; Lopez-Pamies et al., 2025) should predict $\psi_{cr}/\psi_{cr}^{dis}$ with reasonable accuracy. Figure 2.20 shows the good agreement for IG 11 Graphite, highlighting the consistency between the approaches.

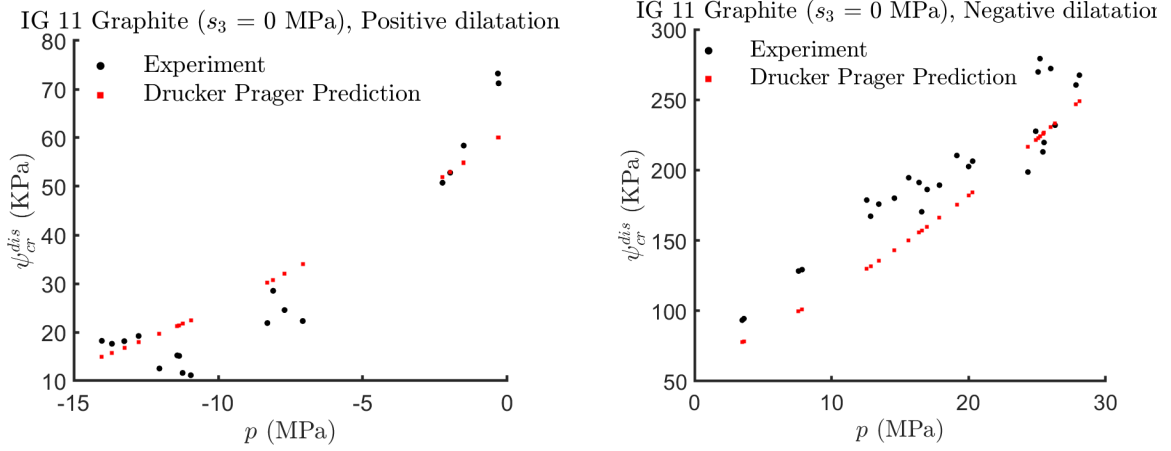


Figure 2.20: **IG 11 Graphite: stress-based failure surface predictions for ψ_{cr}^{dis} .** Since IG 11 Graphite is modeled as a linear elastic material, the calibrated stress-based failure should predict ψ_{cr}^{dis} with reasonable accuracy. Good agreement highlighting the consistency between the two approaches.

Finally, the stress-based failure surface for PU (modeled as hyperelastic; the two approaches should be consistent) from (Kamarei et al., 2026; Lopez-Pamies et al., 2025) was used to obtain ψ_{cr} vs. p predictions for positive dilatation. Figure 2.21 shows the results aligning with the observation of ψ_{cr} decreasing with increasing $|p|$ in Figures 2.17 and 2.18.

Future work should aim to develop a concise, physics-based, generally applicable ψ_{cr}^+ - p relation to accurately model damage initiation and fracture in brittle materials across a broad range of loading conditions.

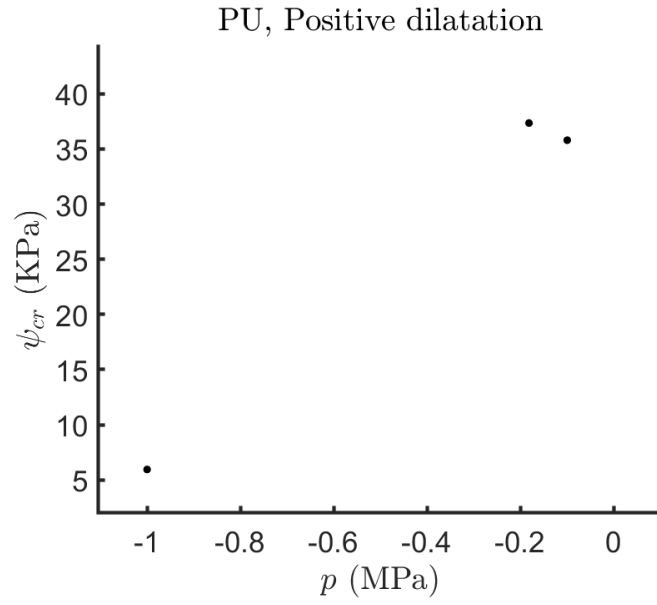


Figure 2.21: **PU: stress-based failure surface predictions for ψ_{cr} .** PU is modeled as hyperelastic, so the two approaches should be consistent. The predicted ψ_{cr} vs. p evolution for positive dilatation aligns with the trend in Figures 2.17 and 2.18, i.e., ψ_{cr} decreasing with increasing $|p|$.

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CHAPTER 3

A physics-informed neural network for modeling fracture without gradient-damage: formulation, application, and assessment

3.1 Introduction

The ability to computationally model and predict damage and fracture of materials and structures is essential for enhancing scientific understanding of failures and enabling robust real-world engineering solutions. The finite element method (FEM) is widely used for the numerical modeling of fracture in a variety of problems (Keyak et al., 2001; Wangen, 2011; Oh et al., 2011). Solutions from FEM without gradient-damage have been shown to exhibit significant mesh-dependency as discussed in Chapter 2 (Section 2.1). The predicted crack propagation path can be nonphysical and inaccurate if the finite element mesh is not aligned with the actual path, which can be difficult to guess a priori. The non-local gradient-damage term in mesh-based numerical modeling is needed to regularize

for mesh-independent solutions. FEM with gradient-damage can yield mesh-independent results, provided the typical element size is less than a factor of the regularization length scale. However, the mathematical and numerical implementation complexity of FEM with gradient-damage is significantly higher compared to that of the simple FEM without gradient-damage. An additional degree of freedom d has to be considered at each point in FEM with gradient-damage, and the corresponding partial differential equation for damage evolution has to be solved. This involves calculating spatial gradients and developing complex user-defined elements and user subroutines for implementation within FEM programs (Narayan and Anand, 2019; Konica and Sain, 2021).

Neural networks (NNs) have been increasingly explored as computational methods for solving engineering mechanics problems previously difficult to analyze (Niu and Srivastava, 2022b,a). Physics-informed neural networks (PINNs) have emerged in the recent literature as a numerical method for solving mechanics problems (Haghighat et al., 2021; Zhang et al., 2022; Bai et al., 2023; Jin et al., 2023; Niu et al., 2023; Hu et al., 2024; Fuhg and Bouklas, 2022; Abueidda et al., 2023; Wang and Yin, 2024; Zhang et al., 2020; Bai et al., 2025; Dong et al., 2025; Guo and Song, 2025). Although PINNs are relatively new compared to FEM (Grossmann et al., 2024), they have certain advantages, which include their open-source nature and relative ease of implementation (Rezaei et al., 2022). PINNs can solve partial differential equations (PDEs) by encoding the mathematical information explicitly into the loss functions without a need for training data. The application of PINNs for mechanics and their advantages and disadvantages with respect to the conventional numerical methods (e.g., FEM) are summarized in Table 3.1.

PINNs for Mechanics
• Spatial coordinates, time, PDE parameters, etc., as inputs to the NN. • Displacements, stress tensor components, field variables, PDE parameters, etc., as the NN outputs. • Loss function divided into two parts: data loss (IC/BCs, and any experimental data) and residual/governing equation loss (computed using the strong or weak formulation of PDEs). • Loss function minimization (e.g., MSE, MAE, Sobolev norm) for obtaining the optimal parameters of the NN. The loss is first computed point-wise (for the strong formulation) over identical and independently distributed residual (collocation) points as well as IC/BC points.

Advantages	Disadvantages
Relative ease of implementation from scratch and open-source nature compared to the conventional numerical methods.	Accuracy of the solution depends on various hyperparameters of the neural network. Lack of guidance for a general class of mechanics problems. Rigorous approaches such as grid search can be used for parameter tuning.
Mesh-free nature and automatic differentiation (AD) capabilities. AD computes exact gradients as the activation functions have analytical gradient forms.	Forward problems without training data need significantly larger computation time than conventional numerical methods.
Non-linear activation functions (e.g., tanh with guaranteed differentiability) are more expressive than conventional method basis functions.	Non-convex, non-linear, and multi-objective nature of loss functions often results in slow convergence.
Combination with data-driven learning is promising. Training can be achieved for sparse and high-dimensional data.	“Black box” nature lacks interpretability but can be used as a surrogate model for “Gray box” learning, such as SR/SINDy.
Handle ill-posed inverse problems better than conventional methods due to the incorporation of physics as a regularizer - mimetic to Tikhonov regularization.	Application to multi-scale and multi-physics problems is challenging.
Can recover missing physics or constitutive relations. Unlike conventional methods, IC/BCs can be partially described.	Detailed validations for a broad class of problems are still in their infancy.

Table 3.1: **A summary of PINNs for mechanics and their advantages and disadvantages with respect to the conventional numerical methods like Finite Element Method.** (IC - initial conditions, BC - boundary conditions, MSE - mean squared error, MAE - mean absolute error, SR - symbolic regression, SINDy - sparse identification of non-linear dynamics)

Machine Learning (ML)-based methods for solving mechanics problems are a rapidly evolving field. Although this chapter focuses on the more popular (and suitable for this study) PINNs, several other methodologies exist. It is prudent to summarize the key methods and discuss their advantages and disadvantages at this stage:

3.1.1 Deep Operator Networks (DeepONet)

NNs learn to map specific input values to corresponding outputs. The underlying ordinary/partial differential equations (ODEs/PDEs) for mechanics problems, however, take functions as inputs rather than single values. NNs (including PINNs) and numerical methods such as finite differences and FEM face a major relevant bottleneck. For each new input function, the solution procedure must be repeated. This can be computationally intensive and slow. DeepONet, first proposed by Lu et al. (Lu et al., 2021), learns the solution operator that maps entire functions to other functions. Specifically, it can learn to predict the outputs of ODEs/PDEs for any given input function without re-running the numerical solver each time.

The DeepONet architecture comprises two main components: a branch network and a trunk network (<https://towardsdatascience.com/operator-learning-via-physics-informed-deeponet-lets-implement-it-from-scratch-6659f3179887/> Shuai Guo, 2023). The branch network takes discrete function values as inputs and maps them to a feature vector. On the other hand, the trunk network takes the evaluation coordinates as inputs and also maps them to a feature vector of the same dimensionality. These two feature vectors are then combined via a dot product, yielding the final output. This separation enables the model to generalize across different input conditions without requiring retraining. In brief, DeepONet discovers the basis functions of the solution space and evaluates their corresponding weights using the data. To eliminate the purely data-driven nature of DeepONets, physics can be incorporated in a manner analogous to PINNs. These

physics-informed DeepONets combine the strengths of DeepONets and PINNs to produce physically consistent predictions, even with limited or noisy data. (Goswami et al., 2022a; He et al., 2023; Wang et al., 2021; Yin et al., 2022) are some recent works utilizing DeepONets for mechanics problems.

Advantages

- High-speed real-time inference and generalization over function spaces: DeepONets, once trained, can solve a new instance of a PDE (i.e., a new forcing function or initial condition) rapidly. Hence, DeepONets can be approximately 1,000 times faster than conventional numerical methods, enabling real-time digital twins and rapid design optimization.
- Zero-shot resolution: A model trained on a coarse grid can provide accurate predictions at a higher resolution without re-training.

Disadvantages

- Significantly large amounts of paired input-output data (typically from thousands of simulations using a conventional numerical method) are needed to train a standard DeepONet. This data generation can be computationally expensive.

3.1.2 PINO

Physics-informed neural operators (PINO) combine the learning capabilities of neural operators (typically the Fourier neural operator (FNO)) with the physics-informed design of PINNs. Similar to DeepONets, PINOs learn the solution operator but have a different architecture based on FNOs. Here, functions are processed on a grid using fast Fourier transforms (FFT) [Fourier basis functions are assumed, and the corresponding weights are evaluated] to capture global features. A few recent works utilizing PINOs for mechanics problems are (Kaewnuratchadasorn et al., 2024; Li et al., 2023; Ding et al., 2024).

Advantages

- High-speed real-time inference and generalization over function spaces.
- Zero-shot resolution.
- Superior accuracy and convergence: PINO often achieves significantly lower errors than DeepONet for complex fluid flows (e.g., Kolmogorov or Burgers' equations).

Disadvantages

- Grid dependency: PINO implementations are typically restricted to regular, rectangular grids.

A comparison of PINNs, DeepONets, and PINOs for mechanics summarizing the discussion above is presented in Table 3.2.

Feature	PINN	DeepONet	PINO
<i>Primary objective</i>	Solve a specific ODE/PDE	Learn mapping between functions	Learn mapping between functions (using Fourier basis functions)
<i>Generalization to new conditions</i>	None, must retrain	High	High
<i>Training data requirement</i>	Very low to none	High (if no physics incorporated), Moderate to low otherwise	Moderate to low
<i>Inference speed</i>	Very slow, must retrain	Very fast	Very fast
<i>Best for</i>	“One-off” complex problems or inverse problems	Real-time surrogates and design optimization	Large-scale, complex field mappings; high-resolution fluid dynamics/turbulence
<i>Key weakness</i>	Need to retrain for every case	Huge datasets needed without physics incorporation	Complex non-periodic geometries

Table 3.2: **Comparison of PINNs, DeepONets, and PINOs for mechanics**

A recent application of ML in mechanics is the use of symbolic regression (SR) to discover constitutive functional forms (Cranmer, 2023). Note that this ML application is significantly different from those described earlier, in which the goal was to solve a PDE given initial and boundary conditions. Unlike “black-box” ML models, SR can provide human-readable, explicit closed-form expressions (“gray-box”) which follow physical constraints (e.g., thermodynamic consistency, frame indifference). Using genetic programming, SR starts with a pool of basic operators (+, −, sin, exp, etc.) and evolves them through mutations and crossovers to find physically grounded expressions (sorted based on complexity and fit scores) that capture the provided data (Abdusalamov et al., 2022; Garbrecht et al., 2023; Li et al., 2025).

Some recent works have focused on the application of PINNs to damage and fracture problems in the small-strain regime (Goswami et al., 2022b, 2020; Zheng et al., 2022; Ghaffari Motlagh et al., 2023; Manav et al., 2024; Wang et al., 2024; Hu et al., 2024; Kiyani et al., 2024). These works directly adopt the relatively complex mathematical formulation of the gradient-damage method to develop the physics-based loss functions. Loss functions in the form of variational energy are more common than separate loss functions that individually enforce the underlying partial differential equations and boundary conditions. Variational energy-based loss functions require numerical integration over the domain to evaluate energy integrals. Variational energy-based PINNs have been developed in (Goswami et al., 2020; Zheng et al., 2022; Manav et al., 2024), which solve the problem incrementally using transfer learning and without training data. Goswami and co-workers (Goswami et al., 2022b) proposed a variational form of DeepONet (V-DeepONet), which is trained by imposing the governing equations in a variational form with labeled data. The V-DeepONet, once trained, can rapidly predict the global solution for any initial crack configuration and loading on that domain. Improvements in the V-DeepONet have been presented in (Kiyani et al., 2024). Yi and co-workers (Yi et al., 2025) proposed a mechanics-informed, model-free symbolic regression framework for solving small-strain

fracture problems.

The PINN and physics-informed ML-based damage and fracture modeling work in the literature is limited to small strains, and these PINNs directly incorporate the gradient-damage formulation. Further, due to the meshless nature of PINNs (Yang et al., 2024), the gradient-damage formulation utilized primarily for numerical reasons (to eliminate mesh dependency in FEM) is not necessary.

The main contributions of this chapter are:

1. We have proposed a PINN without gradient-damage for modeling fracture. Specifically, it does not require training data and does not have the gradient-damage method-associated mathematical and numerical implementation complexities.
2. The PINN is formulated to model large deformation fracture of elastomers. To the best of our knowledge, this has not been attempted in the literature.
3. We have validated the predictive capabilities of the PINN (PINN without gradient-damage) for various defect configurations using baseline solutions obtained from FEM with gradient-damage. The proposed modeling framework and neural network architecture should motivate PINNs without gradient-damage to model damage and fracture in a wide range of materials, as shown through successful application to a biological tissue.
4. The PINN as a fracture modeling method is evaluated through systematic variations in key parameters. The PINN crack path predictions are approximately insensitive to the distribution of collocation points. We also show that relatively computationally inexpensive one-time step PINN solutions provide reasonably accurate crack paths for rate-independent materials under monotonic loading.

Recently, large deformation fracture of elastomers was accurately modeled in Chapter 2 using the gradient-damage framework with FEM (Konale and Srivastava, 2025). We use the critical free energy density-based damage initiation criteria proposed in Chapter

2 for our PINN model. In the presence of multiple defects, crack path prediction is complex. Accurate crack path prediction is important for structural applications. We apply our PINN without gradient-damage to predict the crack paths for various defect configurations. The outline of the rest of the chapter is as follows. The constitutive equations for the modeling of large deformation fracture of elastomers without gradient-damage are presented in Section 3.2, and the corresponding PINN formulation is presented in Section 3.3. The results of the PINN predictions are validated in Section 3.4 with a variety of fracture problems using various defect configurations. In Section 3.5, we provide an assessment of the PINN's performance for various neural network parameter choices. Section 3.6 shows the successful application of the PINN to a biological tissue, highlighting its broader applicability, followed by a conclusion in Section 3.7.

3.2 Modeling framework and constitutive equations

The proposed PINN is applied to elastomers in this chapter to demonstrate its predictive capabilities. We first summarize the basic large-deformation modeling framework and then discuss the damage modeling approach used in this chapter. The deformation gradient $\mathbf{F}$, the right Cauchy-Green deformation tensor $\mathbf{C}$ and the left Cauchy-Green deformation tensor $\mathbf{B}$ are defined as

$$\mathbf{F}(\mathbf{X}, t) = \frac{\partial \mathbf{x}(\mathbf{X}, t)}{\partial \mathbf{X}}, \quad \mathbf{C} = \mathbf{F}^T \mathbf{F}, \quad \mathbf{B} = \mathbf{F} \mathbf{F}^T, \quad (3.1)$$

with $\mathbf{X}$ and $\mathbf{x}$ being the undeformed and deformed position vectors of a material point, respectively. $\mathbf{u} = \mathbf{x} - \mathbf{X}$ is the displacement of a material point at $\mathbf{X}$ and t is the time instant under consideration. The specific form for the referential undamaged free energy density function ψ considered here for the elastomer is compressible Neo-Hookean.

$$\psi = \frac{\mu}{2}(I_1 - 3) + \frac{K}{2}(J - 1)^2, \quad I_1 = \text{trace}(\mathbf{C}_{dis}), \mathbf{C}_{dis} = J^{-\frac{2}{3}}\mathbf{C} \quad \text{and} \quad J = \det \mathbf{F}, \quad (3.2)$$

where μ , K are the ground state shear and bulk modulus, respectively. $\mathbf{C}_{dis}$, I_1 , J are the distortional part of $\mathbf{C}$, the first invariant of $\mathbf{C}_{dis}$ and determinant of $\mathbf{F}$, respectively. The approach discussed in this section should apply to any functional form (hyperelastic model) of choice for ψ . A damage variable $d(\mathbf{X})$ is considered to model damage, which can take the values of either 0 (undamaged) or 1 (fully damaged). This is equivalent to instantaneous damage growth upon initiation at a material point experimentally observed in elastomers (Benvidi et al., 2019; Lee et al., 2024). A material point is considered fully damaged ($d = 1$) if ψ^+ at a material point at any deformation step exceeds a critical value ψ_{cr}^+ , i.e.,

$$d = \begin{cases} 0 & \text{if } \psi^+ \leq \psi_{cr}^+, \\ 1 & \text{if } \psi^+ > \psi_{cr}^+. \end{cases} \quad (3.3)$$

ψ^+ here is the part of ψ which is distortional and tensile dilatational, i.e.,

$$\psi^+ = \begin{cases} \frac{\mu}{2}(I_1 - 3) + \frac{K}{2}(J - 1)^2 & \text{if } J > 1, \\ \frac{\mu}{2}(I_1 - 3) & \text{if } J \leq 1. \end{cases} \quad (3.4)$$

This split accounts for damage growth being driven only by distortional and tensile dilatational deformation (Amor et al., 2009; Hesammokri et al., 2023). To ensure irreversibility of damage, it is required that

$$\text{if } d(\mathbf{X}, t) = 1, \text{ then } d(\mathbf{X}, t + dt) = 1, \quad (3.5)$$

with dt being an infinitesimal increment in t .

The first Piola-Kirchhoff stress $\mathbf{P}$ and Cauchy stress $\mathbf{T}$ tensors are then given as

$$\mathbf{P} = J\mathbf{T}\mathbf{F}^{-\mathrm{T}}, \quad \mathbf{T} = \begin{cases} (1-d) \left[\frac{\mu}{J} (\mathbf{B}_{dis})_0 + K(J-1)\mathbf{I} \right] & \text{if } J > 1, \\ (1-d) \left[\frac{\mu}{J} (\mathbf{B}_{dis})_0 \right] + K(J-1)\mathbf{I} & \text{if } J \leq 1, \end{cases} \quad (3.6)$$

with $(\mathbf{B}_{dis})_0$ denoting the deviatoric part of $\mathbf{B}_{dis}$.

3.3 PINN for large deformation fracture modeling

3.3.1 Neural network architecture

We now present the procedure for constructing the PINN to evaluate fracture solutions in arbitrary geometries subjected to large deformations. The PINN has an input layer, one or multiple hidden layers, and an output layer. Two-dimensional problems are solved using the plane strain assumption. The problems are cast in a Lagrangian framework with $\mathbf{X} = \{X_1, X_2\}$ denoting the coordinates of a material point in the reference configuration. A mixed formulation is used wherein the neural network approximates the first Piola-Kirchhoff stress field $\hat{\mathbf{P}}(\mathbf{X}, t)$ along with the displacement field $\hat{\mathbf{u}}(\mathbf{X}, t)$. The mixed formulation avoids the increased computational cost associated with higher-order partial derivative computations in the displacement-only formulation, along with accuracy and convergence issues (Niu et al., 2023). In addition, the undamaged free energy density $\hat{\psi}(\mathbf{X}, t)$, considered as a state variable, is also approximated by the PINN. All trainable PINN parameters, including the weights and biases in the neurons of the network, are denoted by the vector $\boldsymbol{\theta}$. The PINN formulation can then be written as

$$(\hat{\mathbf{u}}, \hat{\mathbf{P}}, \hat{\psi}) = \mathcal{NN}(\mathbf{X}, t, \boldsymbol{\theta}), \quad (3.7)$$

with $\hat{\cdot}$ denoting direct outputs of the neural network.

Specifically, $\hat{\mathbf{u}} = \{u_1, u_2\}$, $\hat{\mathbf{P}} = \{P_{11}, P_{12}, P_{21}, P_{22}\}$. Hence, there are a total of 7 outputs for a given $\mathbf{X}$ and t . Damage $d(\mathbf{X}, t)$ in this modeling framework can take values of either 0 or 1 as seen through (3.3). We evaluate the approximate field $\tilde{d}$ during training using $\tilde{\psi}(\hat{u})$ [using (3.4)] and (3.3), (3.5) as

$$\tilde{d}(\mathbf{X}, t) = \begin{cases} 0 & \text{if } \tilde{\psi}^+(\mathbf{X}, t) \leq \psi_{cr}^+, \\ 1 & \text{if } \tilde{\psi}^+(\mathbf{X}, t) > \psi_{cr}^+, \\ 1 & \text{if } \tilde{d}(\mathbf{X}, t - dt) = 1. \end{cases} \quad (3.8)$$

Loss function construction

A general boundary value problem involving a continuum solid body can be mathematically described as

$$\underbrace{\text{Div} \mathbf{P} + \mathbf{b}_R = \rho_R \ddot{\mathbf{x}}}_{\text{Balance of linear momentum}} \quad \text{and} \quad \underbrace{\mathbf{P} \mathbf{F}^T = \mathbf{F} \mathbf{P}^T}_{\text{Balance of angular momentum}} \quad \text{in } \Omega, \quad (3.9)$$

$$\mathbf{u} = \bar{\mathbf{u}} \text{ on } \Gamma_u,$$

$$\mathbf{P} \mathbf{n}_R = \bar{\mathbf{t}}_R \text{ on } \Gamma_t.$$

Ω denotes the problem's referential domain. $\mathbf{b}_R$, ρ_R are the referential body force per unit volume and referential density, respectively. $\mathbf{u}$ is the unknown displacement field and $\bar{\mathbf{u}}$ is the prescribed displacement field on part of the boundary Γ_u . $\mathbf{P}$ is the unknown first Piola-Kirchhoff stress field, $\mathbf{n}_R$ is the outward unit normal vector to the referential configuration surface and $\bar{\mathbf{t}}_R$ is the prescribed traction field on the part of the boundary Γ_t . The PINN consists of a physics-informed loss function encoding the discrete version of the underlying boundary value problem, in this case, (3.9). Ω is discretized using N_d points located at $\mathbf{X}_d^i, i = 1, 2, \dots, N_d$. Similarly, Γ_u and Γ_t are discretized using N_u points ($\mathbf{X}_u^i, i = 1, 2, \dots, N_u$) and N_t points ($\mathbf{X}_t^i, i = 1, 2, \dots, N_t$), respectively.

The time domain is discretized into N_t time steps ($t^j, j = 1, 2, \dots, N_t + 1$). Neglecting body forces, the balances of linear and angular momentum in (3.9) can be collocated at a given time instant t^j as

$$\begin{aligned} \text{Div} \hat{\mathbf{P}}(\mathbf{X}_d^i, t^j) &= \rho_R \ddot{\mathbf{u}}(\mathbf{X}_d^i, t^j), \\ \hat{\mathbf{P}}(\mathbf{X}_d^i, t^j) \left(\mathbf{I} + \frac{\partial \hat{\mathbf{u}}(\mathbf{X}_d^i, t^j)}{\partial \mathbf{X}} \right)^T &= \left(\mathbf{I} + \frac{\partial \hat{\mathbf{u}}(\mathbf{X}_d^i, t^j)}{\partial \mathbf{X}} \right) \hat{\mathbf{P}}^T(\mathbf{X}_d^i, t^j). \end{aligned} \quad (3.10)$$

The displacement and traction boundary conditions at a time instant t^j can be applied similarly by the following collocations

$$\hat{\mathbf{u}}(\mathbf{X}_u^i, t^j) = \bar{\mathbf{u}}(\mathbf{X}_u^i, t^j), \quad (3.11)$$

$$\hat{\mathbf{P}}(\mathbf{X}_t^i, t^j) \mathbf{n}_R(\mathbf{X}_t^i) = \bar{\mathbf{t}}_R(\mathbf{X}_t^i, t^j). \quad (3.12)$$

The first Piola-Kirchhoff stress and undamaged free energy density fields are related to the displacement field through constitutive relations in (3.2), (3.3), (3.4), (3.5), and (3.6). Hence, the constitutive first Piola-Kirchhoff stress and undamaged free energy density fields are defined as

$$\begin{aligned} \tilde{\mathbf{P}}^{\text{const}}(\mathbf{X}_d^i, t^j, t^{j-1}) &= \tilde{\mathbf{P}}^{\text{const}}\left(\hat{\mathbf{u}}(\mathbf{X}_d^i, t^j), \tilde{d}^{\text{const}}(\mathbf{X}_d^i, t^{j-1})\right), \\ \tilde{\psi}^{\text{const}}(\mathbf{X}_d^i, t^j) &= \tilde{\psi}^{\text{const}}\left(\hat{\mathbf{u}}(\mathbf{X}_d^i, t^j)\right). \end{aligned} \quad (3.13)$$

Note that $\tilde{d}^{\text{const}}$ is evaluated using $\tilde{\psi}^{\text{const}}$ and (3.3), (3.5).

The constitutive relations can be applied through the following collocations

$$\hat{\mathbf{P}}(\mathbf{X}_d^i, t^j) = \tilde{\mathbf{P}}^{\text{const}}(\mathbf{X}_d^i, t^j, t^{j-1}), \quad \hat{\psi}(\mathbf{X}_d^i, t^j) = \tilde{\psi}^{\text{const}}(\mathbf{X}_d^i, t^j). \quad (3.14)$$

The total loss function $\mathcal{L}_{Total}(\boldsymbol{\theta}^j)$ at an arbitrary time instant $t = t^j$ can now be constructed as the weighted mean squared error with contributions from the balances of linear and angular momentum, boundary conditions, and constitutive relations as

$$\mathcal{L}_{Total} = \alpha_{BLM}\mathcal{L}_{BLM} + \alpha_{BAM}\mathcal{L}_{BAM} + \alpha_{BC}\mathcal{L}_{BC} + \alpha_C\mathcal{L}_C. \quad (3.15)$$

$\mathcal{L}_{BLM}$, $\mathcal{L}_{BAM}$, $\mathcal{L}_{BC}$, $\mathcal{L}_C$ denote the individual loss functions for the balance of linear momentum, the balance of angular momentum, boundary conditions and constitutive relations, respectively. α 's are the corresponding weights. During PINN training for a time instant t^j , the network parameters $\boldsymbol{\theta}^j$ are optimized to minimize the total loss function ($\mathcal{L}_{Total}$),

$$\check{\boldsymbol{\theta}}^j = \underbrace{\text{argmin}}_{\boldsymbol{\theta}^j} \mathcal{L}_{Total}(\boldsymbol{\theta}^j). \quad (3.16)$$

The expanded forms of the individual loss functions that penalize errors in the collocation equations in the least-squares sense can be written as

$$\mathcal{L}_{BLM} = \frac{\alpha_{BLM}}{N_d} \sum_{i=1}^{N_d} |\text{Div} \hat{\mathbf{P}}(\mathbf{X}_d^i, t^j, \boldsymbol{\theta}^j) - \rho_R \ddot{\mathbf{u}}(\mathbf{X}_d^i, t^j, \boldsymbol{\theta}^j)|^2, \quad (3.17)$$

$$\begin{aligned} \mathcal{L}_{BAM} = & \frac{\alpha_{BAM}}{N_d} \sum_{i=1}^{N_d} \left| \hat{\mathbf{P}}(\mathbf{X}_d^i, t^j, \boldsymbol{\theta}^j) \left(\mathbf{I} + \frac{\partial \hat{\mathbf{u}}(\mathbf{X}_d^i, t^j, \boldsymbol{\theta}^j)}{\partial \mathbf{X}} \right)^T \right. \\ & \left. - \left(\mathbf{I} + \frac{\partial \hat{\mathbf{u}}(\mathbf{X}_d^i, t^j, \boldsymbol{\theta}^j)}{\partial \mathbf{X}} \right) \hat{\mathbf{P}}^T(\mathbf{X}_d^i, t^j, \boldsymbol{\theta}^j) \right|^2, \end{aligned} \quad (3.18)$$

$$\begin{aligned}
\mathcal{L}_{BC} &= \alpha_{BCu} \mathcal{L}_{BCu} + \alpha_{BCt} \mathcal{L}_{BCt} \\
&= \frac{\alpha_u}{N_u} \sum_{i=1}^{N_u} |\hat{\mathbf{u}}(\mathbf{X}_u^i, t^j, \boldsymbol{\theta}^j) - \bar{\mathbf{u}}(\mathbf{X}_u^i, t^j)|^2 \\
&\quad + \frac{\alpha_t}{N_t} \sum_{i=1}^{N_t} |\hat{\mathbf{P}}(\mathbf{X}_t^i, t^j, \boldsymbol{\theta}^j) \mathbf{n}_R(\mathbf{X}_t^i) - \bar{\mathbf{t}}_R(\mathbf{X}_t^i, t^j)|^2,
\end{aligned} \tag{3.19}$$

$$\begin{aligned}
\mathcal{L}_C &= \alpha_S \mathcal{L}_S + \alpha_{\text{UFE}} \mathcal{L}_{\text{UFE}} \\
&= \frac{\alpha_S}{N_d} \sum_{i=1}^{N_d} |\hat{\mathbf{P}}(\mathbf{X}_d^i, t^j, \boldsymbol{\theta}^j) - \tilde{\mathbf{P}}^{\text{const}}(\mathbf{X}_d^i, t^j, t^{j-1}, \boldsymbol{\theta}^j, \check{\boldsymbol{\theta}}^{j-1})|^2 \\
&\quad + \frac{\alpha_{\text{UFE}}}{N_d} \sum_{i=1}^{N_d} |\hat{\psi}(\mathbf{X}_d^i, t^j, \boldsymbol{\theta}^j) - \tilde{\psi}^{\text{const}}(\mathbf{X}_d^i, t^j, \boldsymbol{\theta}^j)|^2,
\end{aligned} \tag{3.20}$$

with subscripts S , UFE , BCu , BCt denoting terms associated with stress, undamaged free energy density, displacement boundary condition, and traction boundary conditions, respectively. $\tilde{d}$ from (3.8) after training for every time step is stored in a global variable for use in the next time step's training calculations. The trained parameters $\check{\boldsymbol{\theta}}$ are stored at desired intervals for visualizing the solution's temporal evolution. The approximate PINN solution for the damage problem at a referential point $\mathbf{X}$ and time instant t^j can then be evaluated for post-processing as

$$(\hat{\mathbf{u}}, \hat{\mathbf{P}}, \hat{\psi}) = \mathcal{NN}(\mathbf{X}, t^j, \check{\boldsymbol{\theta}}^j), \tag{3.21}$$

$$\tilde{d} = \begin{cases} 0 & \text{if } \hat{\psi}^+ \leq \psi_{cr}^+, \\ 1 & \text{if } \hat{\psi}^+ > \psi_{cr}^+, \end{cases}$$

$$\tilde{\mathbf{T}} = \tilde{J}^{-1} \tilde{\mathbf{P}} \tilde{\mathbf{F}}^T, \quad \tilde{\mathbf{F}} = \mathbf{I} + \frac{\partial \hat{\mathbf{u}}}{\partial \mathbf{X}}, \quad \tilde{J} = \det \tilde{\mathbf{F}}.$$

The architecture of the PINN for large deformation fracture modeling is summarized in Figure 3.1.

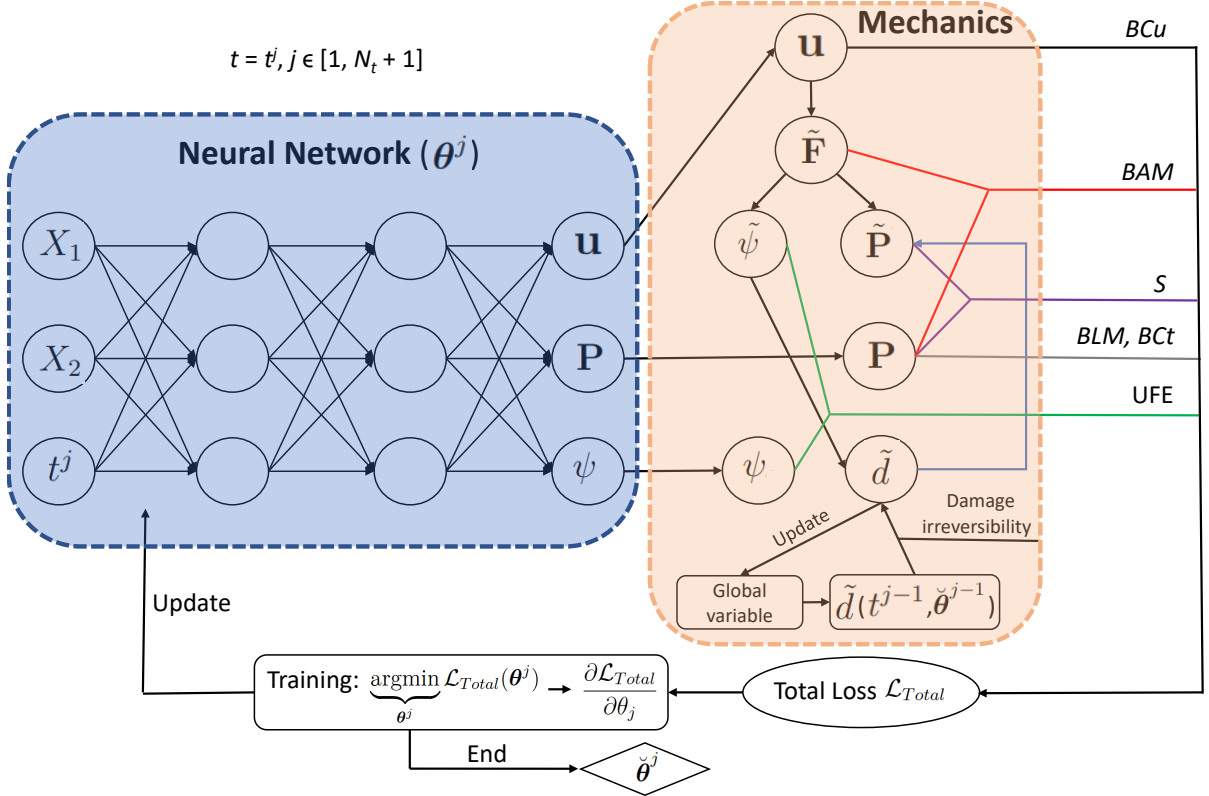


Figure 3.1: **The architecture of the PINN for large deformation fracture modeling.** The formulation is based on temporal discretization into N_t time steps. The loss function formulation and training process for a time instant t^j , $j \in [1, N_t + 1]$ is summarized. Mechanics-related information of boundary conditions (BCu , BCt), governing equations (BLM , BAM) and constitutive relations (S , UFE) is encoded in the total loss function $\mathcal{L}_{Total}$. BLM and BAM denote the individual loss functions corresponding to the balance of linear momentum and angular momentum, respectively. BCu and BCt are the individual loss functions corresponding to displacement and traction boundary conditions, respectively. S and UFE are the individual loss functions for stress and undamaged free energy density constitutive relations, respectively.

For the fracture problems considered in this chapter, the inertial terms have a negligible effect, and they are neglected in (3.17) used for the PINN loss function formulation. In the baseline FEM solutions, we have incorporated the inertial terms and show that this simplification has negligible effects. The same neural network is trained for each time step

without re-initialization, i.e., the trained network parameters for a time step are used as the initial values during training for the next time step.

3.4 Fracture predictions for various defect configurations

We apply the PINN to model fracture in three plane-strain geometries that capture a variety of defect (notches and holes) configurations. The chosen geometries probe different phenomena and are used in the literature to validate new numerical methods for modeling fracture. The units and material parameters are specified in a dimensionless manner, as is standard in PINN applications (Cai et al., 2021; Cuomo et al., 2022). The geometries are stretched uniaxially at a speed of 2 and have dimensions of 1×1 . The values for μ , K , and ψ_{cr}^+ are taken as 0.77, 1.66, and 3, respectively. Note that $\tilde{\mathbf{P}}^{\text{const}}$ in (3.20) is obtained using $\tilde{\mathbf{F}}$, (3.6) and the standard relation between $\mathbf{P}$ and $\mathbf{T}$ which encodes the balance of angular momentum as

$$\mathbf{P} = J\mathbf{T}\mathbf{F}^{-\text{T}} \implies \tilde{\mathbf{P}}^{\text{const}} = \tilde{J} \left[\frac{\mu}{\tilde{J}} (\tilde{\mathbf{B}}_{dis})_0 + K(\tilde{J} - 1)\mathbf{I} \right] \tilde{\mathbf{F}}^{-\text{T}}, \quad d = 0. \quad (3.22)$$

Hence, $\mathcal{L}_{BAM}$ in (3.15) is not considered in the total loss function for training. The default grids of collocation points for all geometries are obtained by creating a uniform 80×80 grid on the 1×1 referential domain and deleting the points lying inside the defects. In addition, the circular regions of the defect boundaries are seeded with 60 residual points.

The PINN is implemented in the PyTorch framework and has three hidden layers, each with 30 neurons. The loss weights α_{BLM} , α_C , α_{BC} are set equal to 1.¹ The optimization algorithm used is ‘Adam’, with an initial learning rate of 0.0001. Using an exponential decay scheduler, the learning rate is gradually decreased during training.² The activation

¹A discussion on this choice is presented in Section 3.5.10.

²Section 3.5.11 presents a discussion on the effects of initial learning and decay rate.

function is the ‘Tanh’ function, except for the last layer, where the ‘Softplus’ activation function is used. This configuration is shown to be optimal for capturing sharp interfaces in the computational domain (Niu et al., 2023), here between the undamaged ($d = 0$) and completely damaged ($d = 1$) regions. Fourier random feature mapping is performed on the neural network input $\mathbf{X} = \{X_1, X_2\}$ as

$$\gamma(\mathbf{X}) = \left(\cos(\mathbf{B}\mathbf{X}), \sin(\mathbf{B}\mathbf{X}) \right), \quad (3.23)$$

with $\mathbf{B}$ being the Fourier random feature matrix with dimensions $m \times 2$. Entries in $\mathbf{B}$ are sampled from a Gaussian distribution. $\gamma(\mathbf{X})$ is then used as an input to the neural network instead of $\mathbf{X}$. This input mapping has been shown to help capture the high-frequency features in PINN solutions (Niu et al., 2023), which are likely to exist here due to the sharp interfaces between undamaged and completely damaged regions. The training is performed with GPU acceleration on the computation cluster provided by the Center for Computation and Visualization (CCV). The corresponding codes can be found at <https://github.com/SrivastavaResearchLab/Fracture-PINN-2025.git>. It is important to note that, due to hyperelasticity, training can be started at any time before damage initiation. This helps reduce the computation time.

The finite element software ABAQUS/Explicit is used to implement FEM with gradient-damage to obtain the baseline solutions for the PINN validation. ABAQUS has been used extensively to successfully model a broad variety of complex mechanics problems (Vaishakh et al., 2024; Konale et al., 2023; Bai et al., 2021; Zhong and Srivastava, 2021; Srivastava et al., 2011). The FEM with gradient-damage implementation procedure using a combination of a user-defined material subroutine (VUMAT) for the deformation problem and a user-defined element (VUEL) for the damage problem described in Chapter 2 (Section 2.6) is followed here.

The gradient-damage method's mathematical formulation is summarized as

$$\mathbf{T} = g(d)\mathbf{T}_o^+, \quad \mathbf{T}_o = \frac{\mu}{J}(\mathbf{B}_{dis})_0 + K(J-1)\mathbf{I}, \quad (3.24)$$

with $g(d) = (1-d)^2$ being the degradation function and $\mathbf{T}_o$, $\mathbf{T}_o^+$ being the undamaged Cauchy stress and its distortional and tensile dilatational part, respectively. The evolution equation for d is given as

$$\zeta \dot{d} = 2(1-d)\mathcal{H} - 2\psi_*(d - l^2 \Delta d), \quad (3.25)$$

where $\zeta > 0$ is a constant kinetic modulus governing the timescale of damage growth. ψ_* is a coefficient with units of energy per unit volume. It represents part of the energy dissipated during damage growth. Δ denotes the Laplacian of a scalar field. $\mathcal{H}$ is a monotonically increasing history function expressed as

$$\begin{aligned} \mathcal{H}(t) &\stackrel{\text{def}}{=} \max_{s \in [0, t]} [\langle \psi^+(s) - \psi_{cr}^+ \rangle], \quad \psi = \frac{\mu}{2}(I_1 - 3) + \frac{K}{2}(J-1)^2, \\ \langle \psi^+(s) - \psi_{cr}^+ \rangle &= \begin{cases} 0 & \text{if } \psi^+(s) - \psi_{cr}^+ < 0, \\ \psi^+(s) - \psi_{cr}^+ & \text{if } \psi^+(s) - \psi_{cr}^+ \geq 0. \end{cases} \end{aligned} \quad (3.26)$$

The number of elements used in the finite element meshes for each plate geometry is sufficiently large ($\sim 10,000$) to ensure good accuracy of the baseline FEM with gradient-damage solutions. Specifically, the finite element meshes used give results reasonably close to the converged solutions while not drastically increasing the computation time. The damage governing equations in the PINN [(3.3), (3.5)], and the gradient-damage method [(3.25), (3.26)] are fundamentally different. Hence, the PINN fracture solutions are validated through comparison of the crack path with the baseline solutions. Point-wise and global L_2 errors are evaluated in the deformation stage before fracture.

The normalized errors for displacement, Cauchy stress, and undamaged free energy density are defined as

$$\text{Displacement : } L_2^u = \frac{\|\hat{\mathbf{u}} - \mathbf{u}^{ref}\|_2}{\|\mathbf{u}^{ref}\|_2}, \quad \mathbf{u} = u_1, u_2, \quad (3.27)$$

$$\text{Cauchy stress : } L_2^S = \frac{\|\tilde{\mathbf{T}} - \mathbf{T}^{ref}\|_2}{\|\mathbf{T}^{ref}\|_2}, \quad \tilde{\mathbf{T}} = \{\tilde{T}_{11}, \tilde{T}_{22}, \tilde{T}_{12}\}, \quad (3.28)$$

$$\text{Undamaged free energy density : } L_2^{\text{UFE}} = \frac{\|\hat{\psi} - \psi^{ref}\|_2}{\|\psi^{ref}\|_2}, \quad (3.29)$$

where the superscript *ref* denotes quantities obtained from the baseline FEM with gradient-damage solutions. FEM without gradient-damage solutions for different mesh structures were also obtained using ABAQUS/Explicit to reemphasize their mesh-dependency. For this, the deformation-only model implemented using a VUMAT was used along with the in-built element deletion in ABAQUS. The solutions of the PINN after training on the 80×80 grid were evaluated on a testing grid consisting of 1000×1000 points to test its extrapolation capabilities. For L_2 error evaluation and crack path comparison, nodal data exported from ABAQUS were interpolated on the testing grid.

3.4.1 Asymmetric double-edge notched plate

A specimen geometry often used to test the predictive capabilities of damage and fracture models in terms of crack path is a plate with asymmetric double-edge notches subjected to tension under plane-strain conditions (Ambati et al., 2016; Simoes and Martínez-Pañeda, 2021; Han et al., 2022). The loading schematic and specimen geometry considered are shown in Figure 3.2(A). The crack path predictions obtained using FEM with and without gradient-damage for a mesh not aligned with the anticipated crack path are shown in Figure 3.2(B). As expected, the results from FEM without a gradient-damage

solution are nonphysical. The significant mesh-dependency of FEM without gradient-damage is shown through comparison of the predicted crack paths for two different meshes in Figure 3.2(C). The nonphysical crack paths are dissimilar for the two meshes. In FEM without gradient-damage, upon failure of an element, the subsequent damage growth depends on the local mesh structure around that element, resulting in the solution's strong mesh dependency.

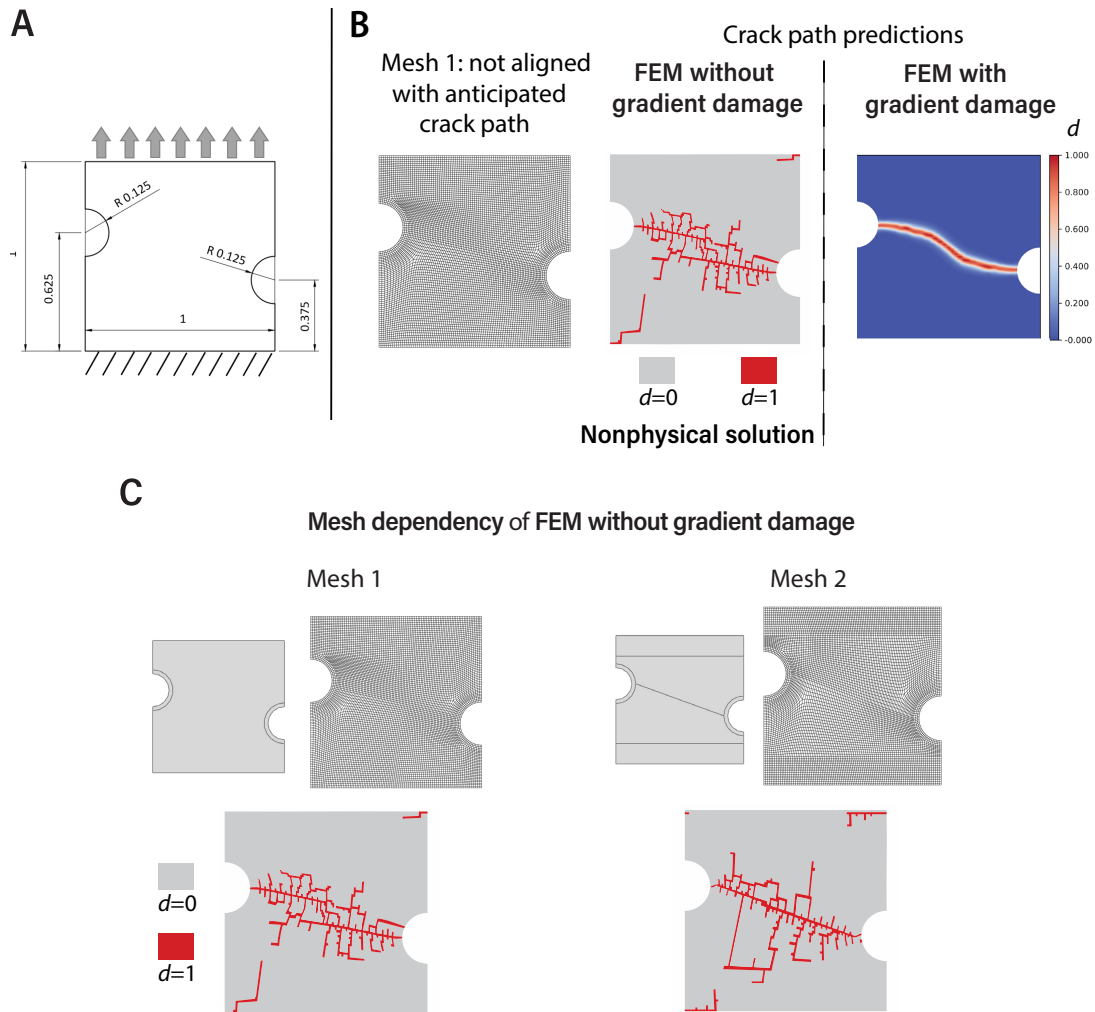


Figure 3.2: **Asymmetric double-edge notched plate: schematic, FEM with gradient-damage and FEM without gradient-damage crack path predictions.** (A) Schematic for an asymmetrically double-edge notched plate specimen subjected to tension under plane strain conditions. The geometry is stretched at a speed of 2. (B) Mesh not aligned with the anticipated crack path. The FEM without gradient-damage solution, as expected, is nonphysical, while the FEM with gradient-damage predicts the correct crack path. (C) The crack paths from FEM without gradient-damage for two different meshes. The solutions differ significantly, highlighting FEM without gradient-damage's mesh dependency.

For the collocation grid in Figure 3.3, the PINN was applied incrementally [multiple time increments, referred to as PINN (incremental) hereon]. Specifically, the time step size was chosen to be 0.0025 s with the first increment at $t = 0.42$ s. Training for each time increment was performed for 25,000 epochs. The time interval from the initiation of damage to the complete rupture ($t = 0.08$ s) is small compared to the time to the initiation of damage (FEM with gradient-damage: 0.477 s, PINN (incremental): 0.457 s), highlighting the rapid damage growth upon initiation built into the PINN formulation. The crack path predictions from the PINN (incremental) and the baseline FEM with gradient-damage solution show good agreement, as shown in Figure 3.3.

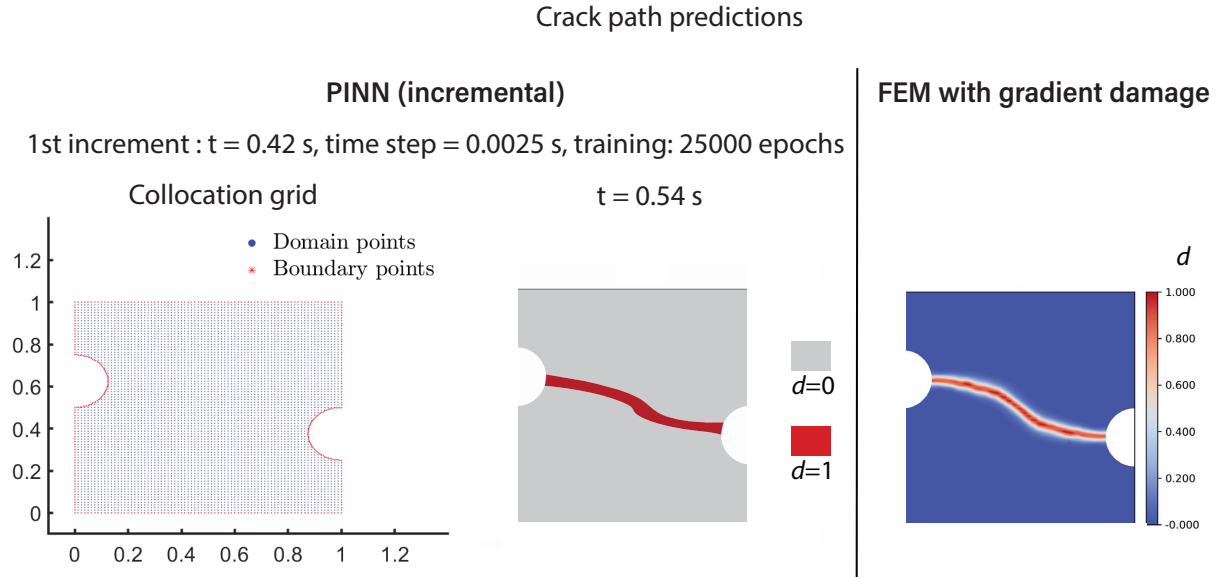


Figure 3.3: **Asymmetric double-edge notched plate: PINN (incremental) crack path prediction.** The collocation grid is shown. For the PINN (incremental) application, the time step size used is 0.0025 s, with the first increment being $t = 0.42$ s. Training for each time increment is performed for 25,000 epochs. Damage solution from PINN (incremental) at $t = 0.54$ s. The crack paths from PINN (incremental) and the baseline FEM with gradient-damage show good agreement.

3.4.2 Single-edge notched plate with a hole

A single-edge notched plate with a hole under plane-strain conditions is a specimen geometry utilized to validate damage and fracture models (Ambati et al., 2015; Sargado et al., 2021; Muixí et al., 2020). The loading schematic and specimen geometry considered

are shown in Figure 3.4(A). The crack path predictions obtained using FEM with and without gradient-damage for a mesh not aligned with the anticipated crack path are shown in Figure 3.4(B). The FEM without gradient-damage solution is nonphysical. Figure 3.4(C) shows the physical solution using FEM without gradient-damage for a mesh aligned with the anticipated crack path, highlighting the method's mesh dependency.

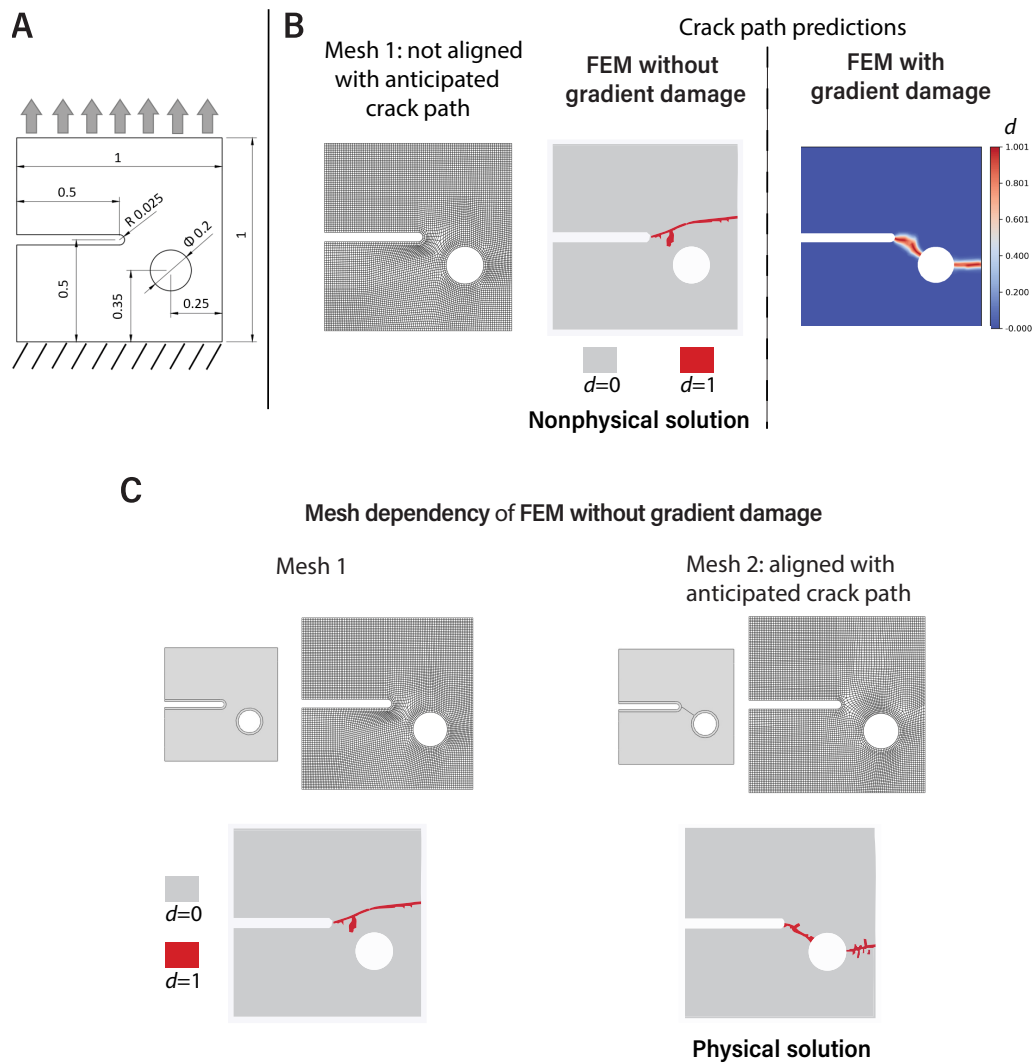


Figure 3.4: **Single-edge notched plate with a hole: schematic, FEM with and without gradient-damage crack path predictions.** (A) Schematic for a single-edge notched plate with a hole subjected to tension under plane strain conditions. The geometry is stretched at a speed of 2. (B) The crack paths from FEM with and without gradient-damage for a mesh not aligned with the anticipated crack path. The FEM without gradient-damage solution is nonphysical. (C) Mesh dependency of FEM without gradient-damage is highlighted, where a mesh aligned with the anticipated crack path shows the anticipated answer (shown on the right) while a general mesh without the knowledge of the crack path makes a very incorrect prediction (shown on the left).

PINN (incremental) is applied to the collocation grid in Figure 3.5(B) with time step size = 0.01 s, the first time increment at $t = 0.23$ s, and 50,000 epochs training for each time increment. The progression of damage at four different stages and the crack path in the PINN (incremental) solution agree with the FEM with gradient-damage solution as seen in Figure 3.5.

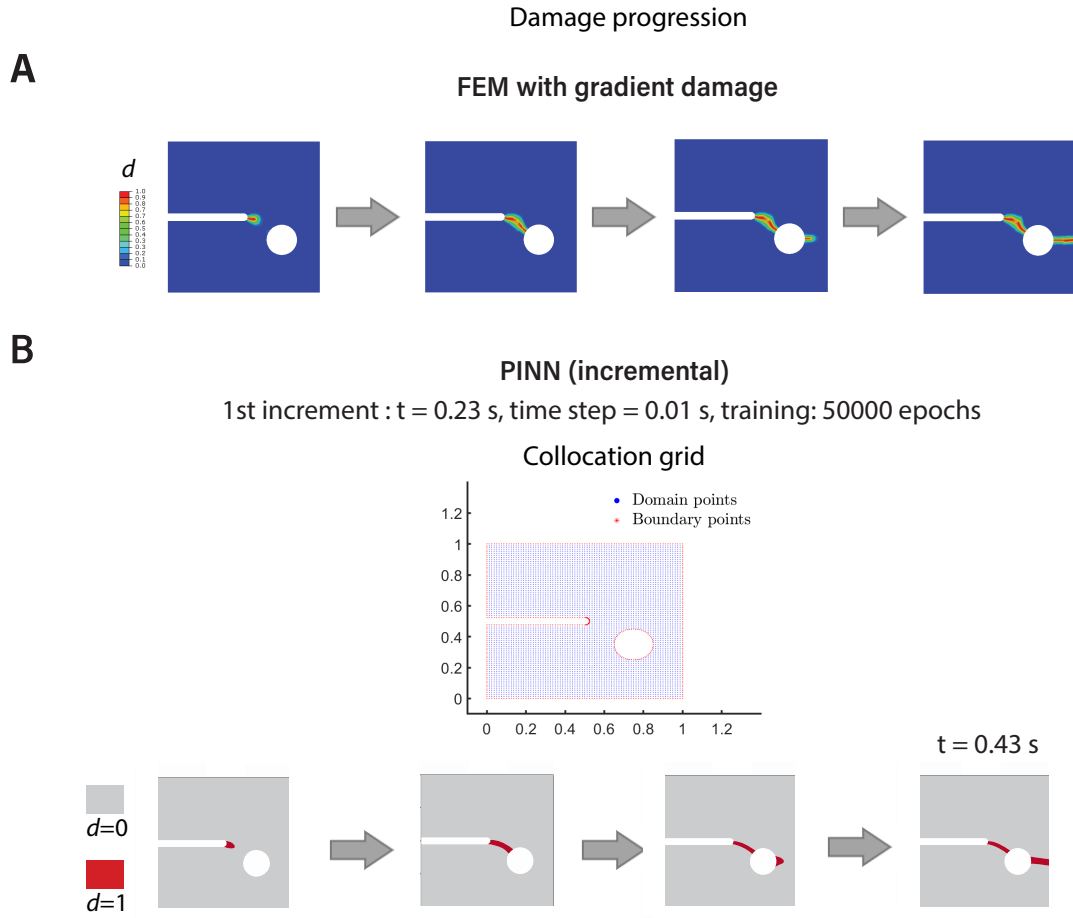


Figure 3.5: Single-edge notched plate with a hole: PINN (incremental) damage progression and crack path predictions. Damage progression at four different stages and crack paths from **(A)** FEM with gradient-damage and **(B)** PINN (incremental) [along with collocation grid considered]. For the PINN (incremental) application, the time step size used is 0.01 s, with the first increment being $t = 0.23$ s. Training for each time increment is performed for 50,000 epochs. Damage progression and crack paths from the PINN (incremental) and the FEM with gradient-damage solutions show good agreement.

We also consider a variation in this geometry where the hole is moved further away from the notch. Specifically, only the separation of the hole center from the notch centerline in Figure 3.4(A) is increased by 0.2. The crack path in the FEM with gradient-damage

solution changes significantly as seen in Figure 3.6. The PINN [$t = 0.55$ s, time step size = 0.55 s, 200,000 epochs training for the time increment] captures the crack path variation with the defect positioning accurately, as shown in Figure 3.6.

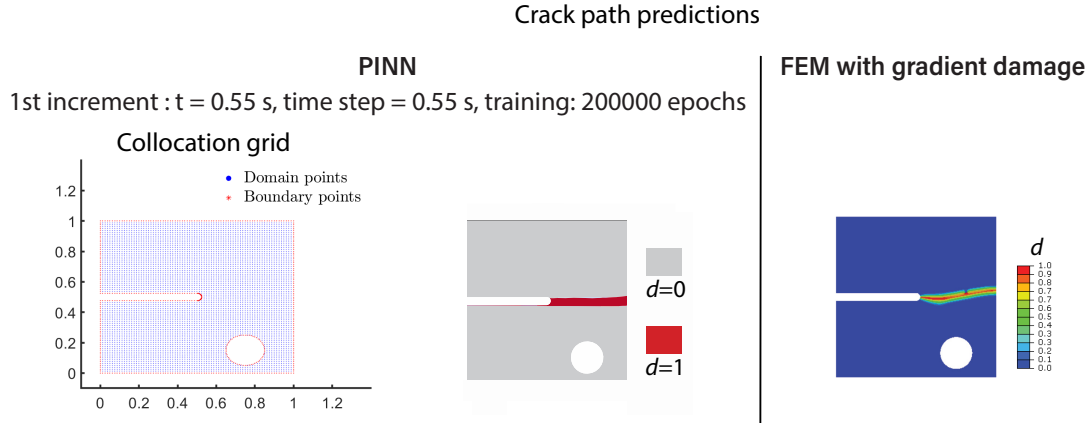


Figure 3.6: **Single-edge notched plate with a hole (increased notch and hole separation): PINN crack path prediction.** A variation of the single-edge notched plate with a hole geometry is considered. The separation of the hole center from the notch centerline in Figure 3.4(A) is increased by 0.2. This results in a significant change in the crack path, as seen through the FEM with the gradient-damage solution. The PINN solution was evaluated at $t = 0.55$ s. The time step size used was 0.55 s, and training was performed for 200,000 epochs for the time increment. Good agreement of the crack path from the PINN with that from the FEM with baseline gradient-damage solution. The PINN can accurately capture variations in the crack path with defect positioning.

3.4.3 Plate with four randomly distributed holes

We now consider a relatively complex geometry consisting of four holes with different diameters randomly distributed on the 1x1 plate domain. The loading schematic, specimen geometry, and finite element mesh considered are shown in Figure 3.7(A). The crack path prediction obtained using FEM with gradient-damage is shown in Figure 3.7(B).

The PINN (incremental) is applied to the collocation grid in Figure 3.8(B) with time step size = 0.0025 s, the first time increment at $t = 0.3$ s, and 25,000 epochs training for each time increment. Good agreement of the damage progression at four different stages and the crack path from the PINN (incremental) solution with the baseline FEM with gradient-damage solution can be seen in Figure 3.8.

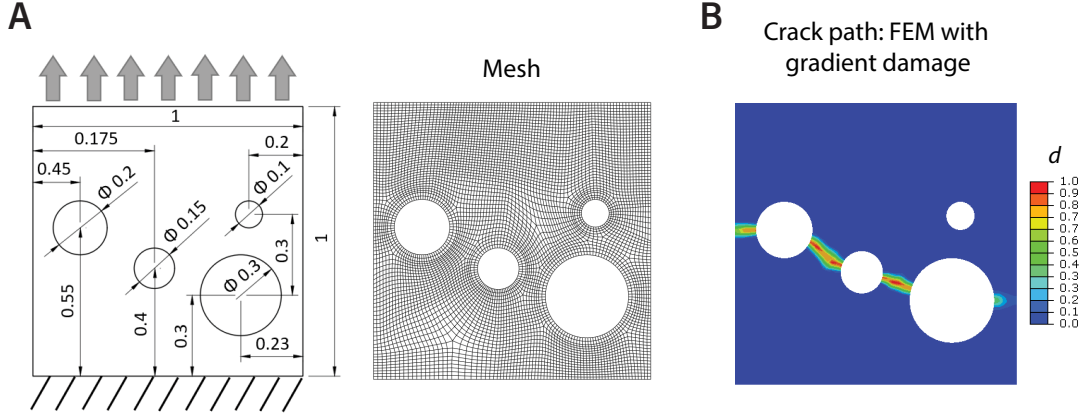


Figure 3.7: **Plate with four randomly distributed holes: schematic and FEM with gradient-damage crack path prediction.** (A) Schematic for a plate with four randomly distributed holes subjected to tension under plane strain conditions. The geometry is stretched at a speed of 2. The mesh used for FEM with gradient-damage application. (B) The crack path solution using FEM with gradient-damage.

3.5 Assessment of the PINN performance for fracture

Previously, we demonstrated the success of PINN without gradient-damage formulation in predicting the fracture path for a variety of problems. The performance of any new and emerging computational method has to be evaluated to guide the choice of underlying parameters for future applications. Towards this, we investigate the effect of a few key neural network parameters on the PINN’s prediction performance. The PINN solutions in this section for the asymmetric double-edge notched plate geometry are obtained using a relatively computationally inexpensive single time increment approach [referred to as PINN (one-time step) hereon]. In this chapter, we have considered (i) hyperelastic (rate-independent) material response, (ii) instantaneous damage growth, which is approximately observed in experiments for elastomers and several soft polymers, where damage, when it initiates, grows very rapidly, and (iii) monotonic loading.³ For the general class of fracture problems, PINN (incremental) [multiple time increments] solutions will have better accuracy. However, PINN (one-time step) can enable computationally cheaper neural network parameter and architecture optimization before PINN (incremental) is

³Under these conditions, the PINN (one-time step) solutions provide crack paths with *reasonable* accuracy, as shown in Section 3.5.1.

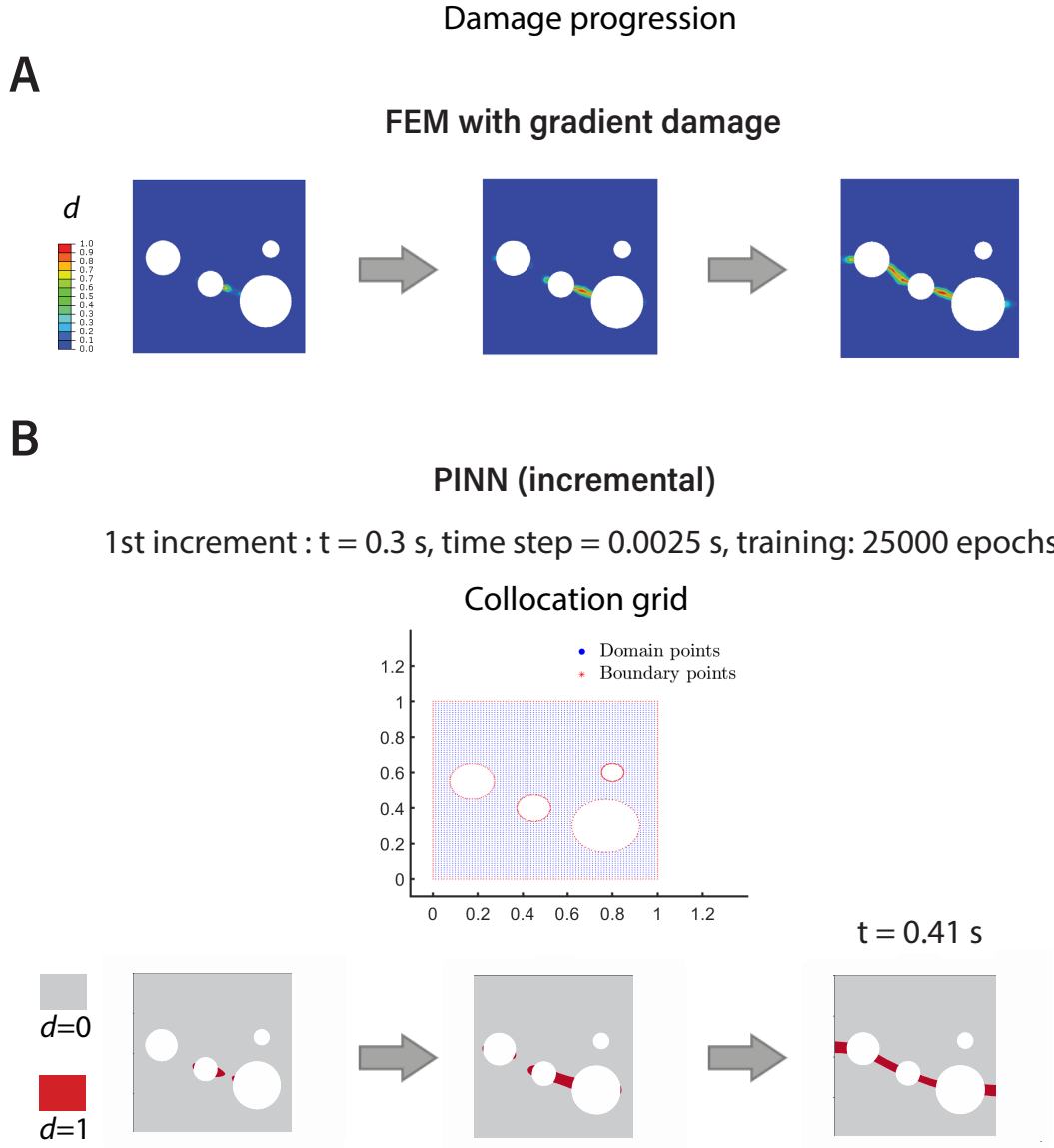


Figure 3.8: **Plate with four randomly distributed holes: PINN (incremental) damage progression and crack path predictions.** Damage progression at four different stages and crack paths from **(A)** FEM with gradient-damage and **(B)** PINN (incremental) [along with collocation grid considered]. For the PINN (incremental) application, the time step size used is 0.0025 s, with the first increment being $t = 0.3$ s. Training for each time increment is performed for 25,000 epochs. Good agreement of the damage progression and crack paths from the PINN (incremental) and the FEM with gradient-damage solutions.

applied for accurate final results.

The crack paths are obtained through PINN (one-time step) solutions at $t = 0.65$ s with 100,000 epochs of training. L_2 relative errors and losses are evaluated through PINN (one-time step) application at $t = 0.45$ s (before damage initiation) with 100,000 epochs

training. First, variations in the collocation point distribution (approximately constant grid density) are considered. Further, the number of layers, the number of neurons in each layer, and the grid density (in a fixed grid structure) are systematically varied. The base number of layers, number of neurons in each layer, and grid density are the values used in Section 3.4, i.e., 5, 30, and 80×80 , respectively. We also compare the computation time for the PINN using the base network and base grid density with those for FEM without and with gradient-damage. The effects of simple pre-training and output normalization strategies on the solutions are also explored.

3.5.1 Comparison of PINN (incremental) and PINN (one-time step)

The crack path predictions using PINN (incremental) [1st increment at $t=0.45$ s, time step= 0.0025 s, 50,000 epochs training] and PINN (one-time step) [$t=0.65$ s, 100,000 training epochs] are shown in Figure 3.9. The PINN (incremental) crack path solution exhibits better agreement with the baseline FEM with gradient solution. However, the computationally inexpensive PINN (one-time step) predicts the crack path with reasonable accuracy. Hence, neural network parameter and architecture optimization can be performed in a computationally efficient manner using PINN (one-time step) before PINN (incremental) applications for accurate final results.

3.5.2 L_2 relative error and loss evolution during training

L_2 relative error (u - displacement, S - stress, UFE - undamaged free energy density) and loss (total, BLM - balance of linear momentum, BC - boundary conditions, C - constitutive equations) evolution with number of epochs during training [PINN (one-time step), $t=0.45$ s, 100,000 epochs, grid from Figure 3.3] for the asymmetric double-edge notched plate geometry is shown in Figure 3.10. The errors and losses decrease monotonically with the number of epochs in this range. The low errors with respect to

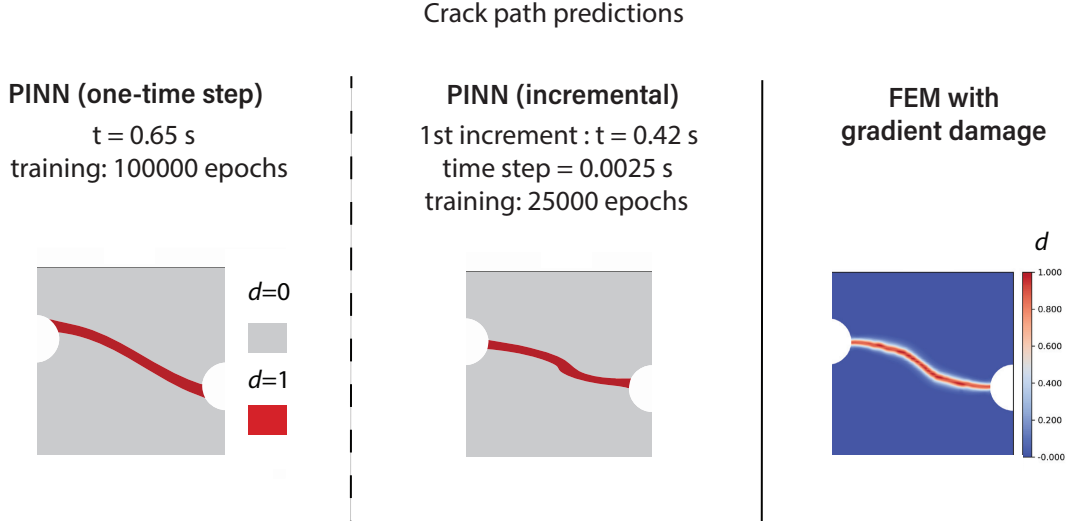


Figure 3.9: **Asymmetric double-edge notched plate: comparison of PINN (incremental) and PINN (one-time step) crack path predictions.** Comparison of PINN (incremental) [1st increment at $t=0.45$ s, time step= 0.0025 s, 25,000 epochs training] and PINN (one-time step) [$t=0.65$ s, 100,000 training epochs] crack predictions. The PINN (incremental) crack path solution shows better agreement with the baseline FEM with gradient-damage solution. The computationally inexpensive PINN (one-time step), however, predicts the crack path with reasonable accuracy. PINN (one-time step) can thus enable computationally cheaper neural network parameter and architecture optimization before PINN (incremental) applications for accurate final results.

the baseline FEM solutions highlight the validity of the simplification discussed in Section 3.3.1.

Corresponding results for the single-edge notched plate with a hole and the plate with four randomly distributed holes geometries are shown in Figure 3.11. For the plate with four randomly distributed holes, only L_2^u is presented due to numerical issues in the interpolation of the relatively complex stress and undamaged free energy density fields on the testing grid. The errors and losses decrease monotonically with the number of epochs in this range for both geometries.

3.5.3 PINN solution convergence

The convergence of the PINN solutions for the asymmetric double-edge notched plate geometry is verified as a function of the number of training epochs. The L_2 error, loss ($t = 0.45$ s), and final crack path ($t = 0.65$ s) evolution with training epochs (1,000,000

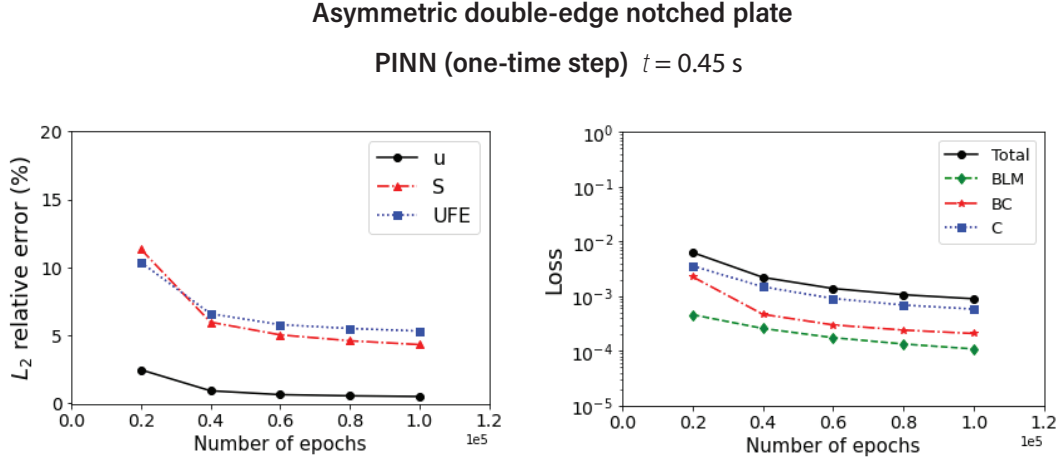


Figure 3.10: **Asymmetric double-edge notched plate: L_2 relative error and loss evolution during training.** PINN (one-time step) is applied at $t = 0.45$ s (before damage initiation) with 100,000 epochs training. The following notations are used: u - displacement, S - stress, UFE - undamaged free energy, BLM - balance of linear momentum, BC - boundary conditions, C - constitutive equations. A monotonic decrease in errors and losses is observed during training.

epochs - 10 times the number of epochs used) is shown in Figure 3.12. The errors and losses decrease monotonically and approximately converge. The overall width of the crack path decreases with epochs, and the path converges. The PINN formulation does not have an inherent length scale parameter to control the damage zone width. Consequently, the predictions are a function of the neural network parameters. There is no physical correlation. Even in the baseline FEM with gradient-damage approach, where a diffusive crack is considered, the “length scale” parameter is present purely for numerical reasons.

3.5.4 Effect of collocation point distribution

The four different collocation point distributions considered and the corresponding PINN (one-time step) solutions for the asymmetric double-edge notched plate geometry are shown in Figure 3.13. The PINN crack path solutions are approximately insensitive to the distribution and show good agreement with the baseline FEM with gradient-damage solution.

A

Single-edge notched plate with a hole

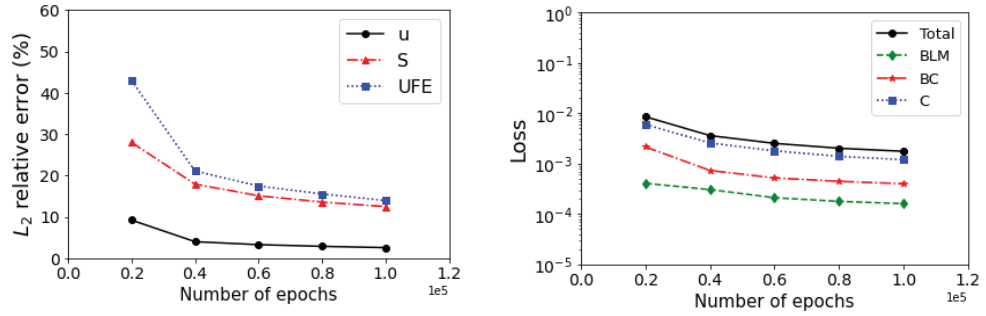
PINN (one-time step) $t = 0.18$ s**B**

Plate with four randomly distributed holes

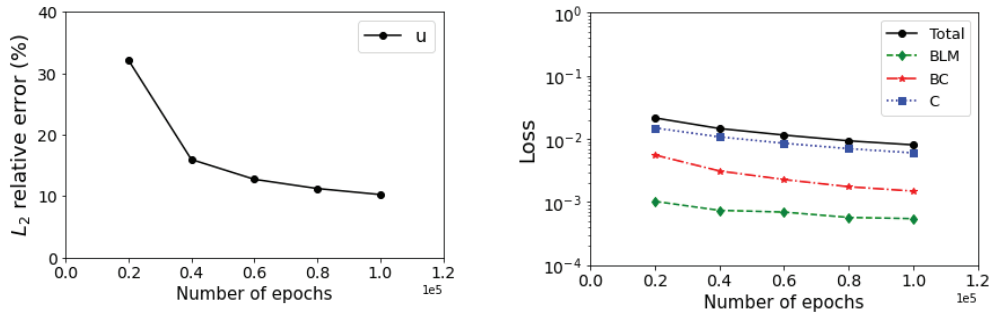
PINN (one-time step) $t = 0.3$ s

Figure 3.11: **Single-edge notched plate with a hole and plate with four randomly distributed holes: L_2 relative error and loss evolution during training.** PINN (one-time step) is applied at $t = 0.18$ s and $t = 0.3$ s (both before damage initiation) with 100,000 epochs training, respectively, for **(A)** the single-edge notched plate with a hole and **(B)** the plate with four randomly distributed holes geometries. Only L_2^u is presented for the plate with four randomly distributed holes due to numerical issues in the interpolation of the relatively complex stress and undamaged free energy density fields on the testing grid. The errors and losses decrease monotonically with the number of epochs in this range for both geometries.

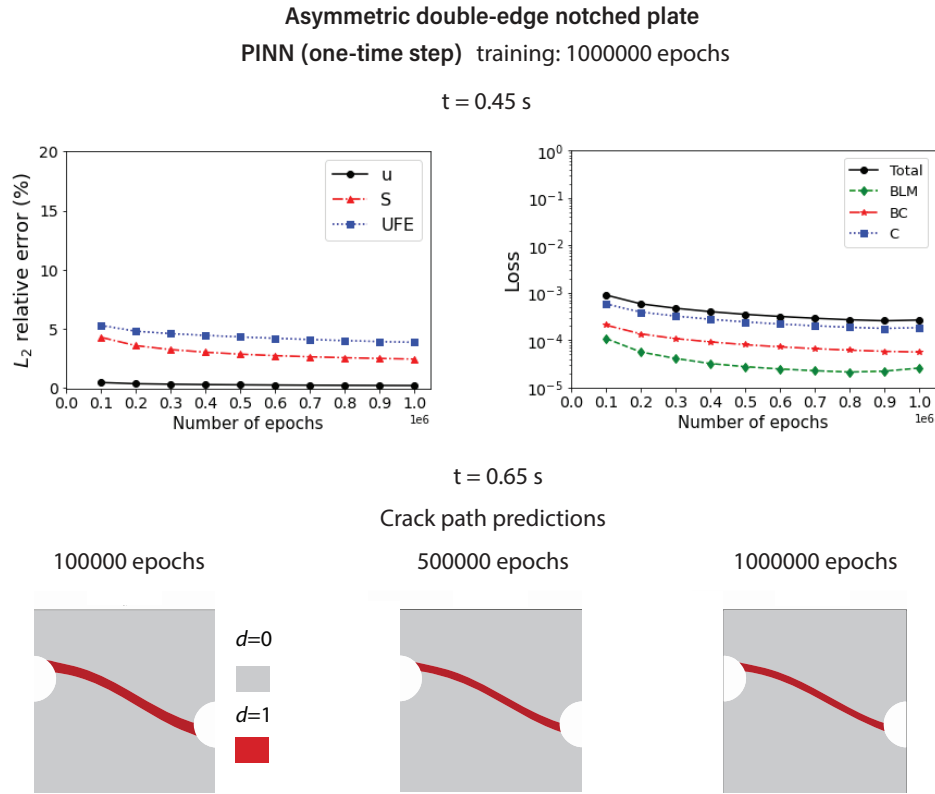


Figure 3.12: **Asymmetric double-edge notched plate: PINN solution convergence.** For PINN (one-time step) application, the solution is evaluated at $t = 0.45 \text{ s}$ and $t = 0.65 \text{ s}$ with 1,000,000 training epochs (10 times the number of epochs used). The L_2 errors and losses decrease monotonically and approximately converge with respect to the number of training epochs. The overall width of the crack path decreases with epochs, and the path converges.

The length scale parameter in FEM with gradient-damage creates a diffuse damage zone around a completely damaged element. The restriction of $h_e \lesssim 0.2l$ (typical element size) prescribed for FEM with gradient-damage in the literature ensures that the diffuse zone is sufficiently large for the local mesh structure not to affect the following damage growth and the crack path. The meshless form of the PINN, i.e., physics-informed loss function minimization at collocation points not connected by a mesh, results in the crack path solutions being approximately insensitive to the collocation point distribution.

Figure 3.14 shows the different collocation point distributions considered and the corresponding PINN (one-time step) crack path solutions for the single-edge notched plate with a hole and the plate with four randomly distributed holes geometries. The PINN solutions are approximately insensitive to the collocation point distribution and agree well with the baseline FEM with gradient-damage solutions for both geometries.

3.5.5 Effect of the number of layers, number of neurons in each layer, and grid density

The three different numbers of layers, number of neurons in each layer, and grid density considered were 3, 5, 8; 10, 30, 50; and 20×20 , 40×40 , 80×80 , respectively. A numerical $3 \times 3 \times 3$ Factorial Design of Experiments (DOE) was performed to understand the main, simple, and interacting effects of the three hyperparameters. Figures 3.15(A), (B), (C) and (D) show the L_2^{UFE} % errors for a few cases, highlighting the effect of (i) number of layers (80×80 grid density), (ii) number of neurons in each layer (80×80 grid density), (iii) grid density (5 layers), and (iv) grid density (30 neurons in each layer), respectively. All three factors have main effects. Significant interaction effects are observed between the number of layers and the number of neurons, as well as between the grid density and the number of layers. Underfitting or overfitting occurs when the neural network parameters are not optimal.

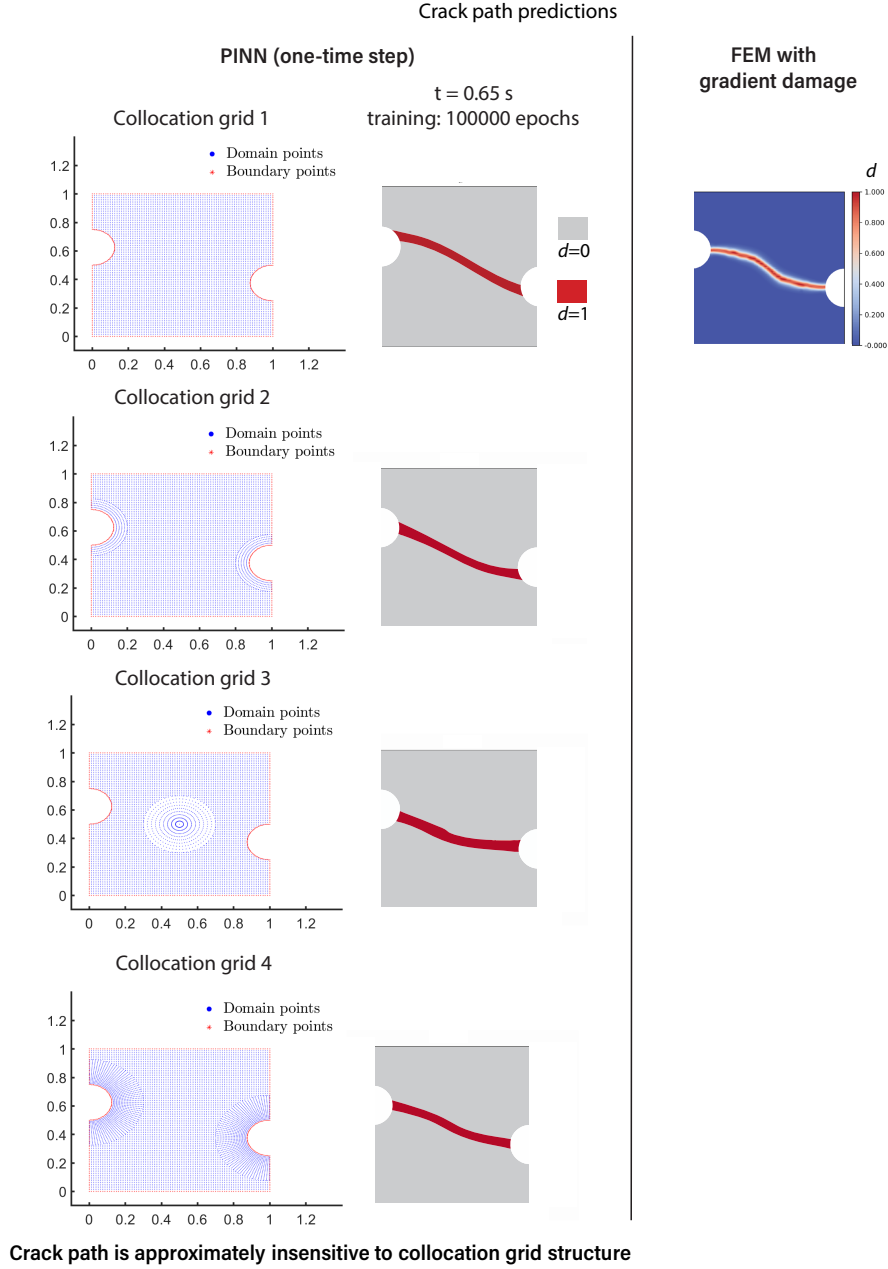


Figure 3.13: **Asymmetric double-edge notched plate: effect of collocation point distribution on crack path.** The four different collocation point distributions considered are shown. PINN (one-time step) is applied at $t = 0.65 \text{ s}$ with training for 100,000 epochs. The crack paths for the four distributions using PINN (one-time step). The crack paths are approximately insensitive to the collocation point distribution, contrary to the significant mesh-dependency of FEM without gradient-damage solutions (see Figure 3.2). Good agreement of the PINN (one-time step) crack path predictions for the four distributions with the baseline FEM with gradient-damage solution.

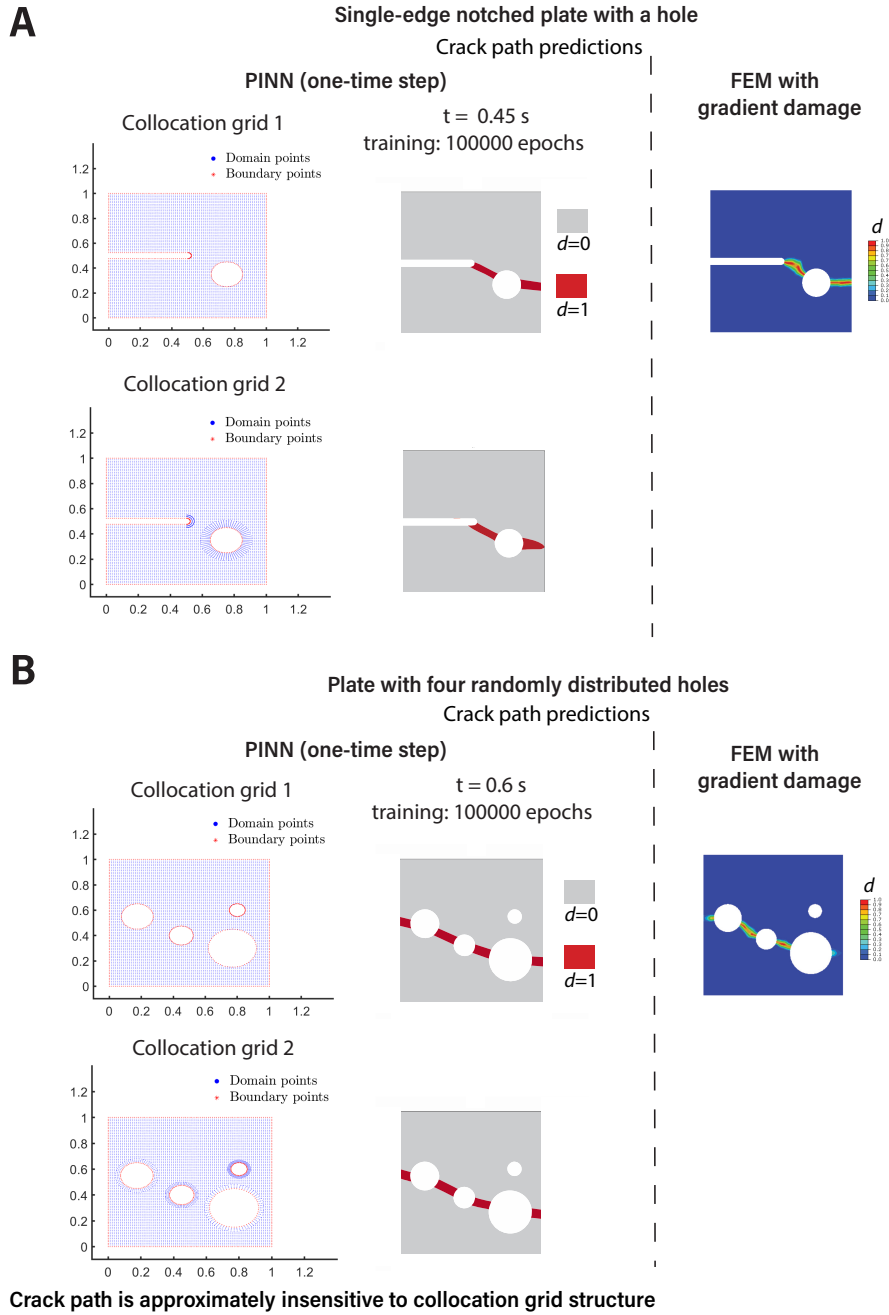


Figure 3.14: **Single-edge notched plate with a hole and plate with four randomly distributed holes: effect of collocation point distribution on crack path.** PINN (one-time step) is applied at $t = 0.45$ s and $t = 0.6$ s with 100,000 epochs training, respectively, for (A) the single-edge notched plate with a hole and (B) the plate with four randomly distributed holes geometries. The two collocation point distributions for the two geometries are shown. Good agreement of the PINN (one-time step) predictions with the baseline FEM with gradient-damage solutions for both geometries. The PINN solution's insensitivity to the collocation point distribution is reemphasized.

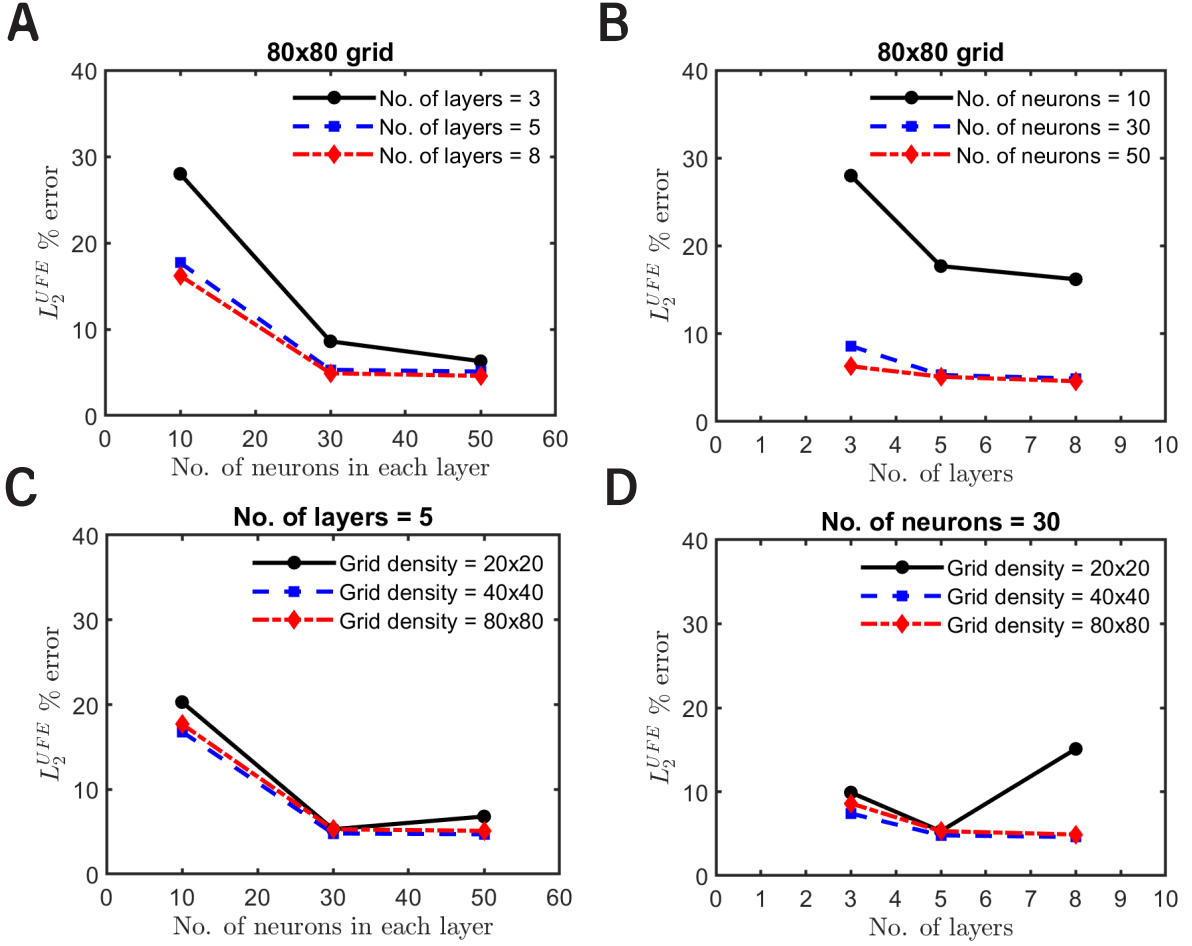


Figure 3.15: **Asymmetric double-edge notched plate: numerical factorial experiment** (L_2^{UFE} % errors, $t = 0.45$ s). For L_2 error comparison, PINN (one-time step) is applied at $t = 0.45$ s with 100,000 epochs training. The effects of (A) the number of layers (80×80 grid density), (B) the number of neurons in each layer (80×80 grid density), (C) grid density (5 layers) and (D) grid density (30 neurons). The three factors each exhibit a main effect. Significant interaction effects can be seen between the number of layers and the number of neurons, and between grid density and the number of layers. Non-optimal hyperparameter values lead to underfitting or overfitting.

The complete results for the L_2 relative errors and the crack paths for the DOE study are shown in Figures 3.16 and 3.17, respectively.

Asymmetric double-edge notched plate
PINN (one-time step) $t=0.45$ s training: 100000 epochs

L_2^u					L_2^S					L_2^{UFE}				
20x20 grid		Layers			20x20 grid		Layers			20x20 grid		Layers		
L_2^u relative error %		3	5	8	L_2^S relative error %		3	5	8	L_2^{UFE} relative error %		3	5	8
Neurons	10	6.7	4.3	9.3	Neurons	10	26.6	55.2	190.9	Neurons	10	30.2	20.3	42.8
	30	0.8	0.5	3.3		30	7.4	5.2	174.4		30	9.9	5.3	15.1
	50	0.6	1.0	6.1		50	6.1	5.5	24.2		50	8.0	6.8	23.9
40x40 grid					40x40 grid					40x40 grid				
L_2^u relative error %		3	5	8	L_2^S relative error %		3	5	8	L_2^{UFE} relative error %		3	5	8
Neurons	10	6.7	5.3	5.1	Neurons	10	24.6	22.9	22.1	Neurons	10	28.5	16.8	16.1
	30	0.8	0.4	0.4		30	6.0	4.1	3.6		30	7.4	4.8	4.6
	50	0.4	0.3	0.3		50	4.3	3.6	2.6		50	5.7	4.7	4.2
80x80 grid					80x80 grid					80x80 grid				
L_2^u relative error %		3	5	8	L_2^S relative error %		3	5	8	L_2^{UFE} relative error %		3	5	8
Neurons	10	6.7	5.4	5.3	Neurons	10	24.9	22.7	22.5	Neurons	10	28.0	17.7	16.2
	30	1.0	0.5	0.4		30	6.7	4.3	3.7		30	8.6	5.3	4.9
	50	0.6	0.4	0.3		50	5.1	3.8	3.0		50	6.3	5.1	4.6

Figure 3.16: **Asymmetric double-edge notched plate: numerical factorial experiment (L_2 errors, $t=0.45$ s).** For L_2 error comparison, PINN (one-time step) is applied at $t = 0.45$ with 100,000 epochs training. L_2^u , L_2^S , and L_2^{UFE} relative errors (%) with variations in the number of layers, number of neurons in each layer, and grid density are shown. The trends of the L_2 errors with respect to each hyperparameter considered are non-monotonic. Non-optimal hyperparameter values lead to underfitting or overfitting.

3.5.6 Comparison of computation time: PINN, FEM without and with gradient-damage

The computation time using the base network (5 layers each with 30 neurons) and the base grid density (80×80) for 1 epoch of PINN training remained approximately constant for all numerical problems considered and was equal to 0.06 s (GPU accelerated, QuadroRTX, CCV@Brown), 0.18 s (GPU accelerated, NVIDIA T600 & Intel Core i7-10700 Processor, Dell Precision 3650 Tower), and 0.24 s (not GPU accelerated, CCV@Brown).

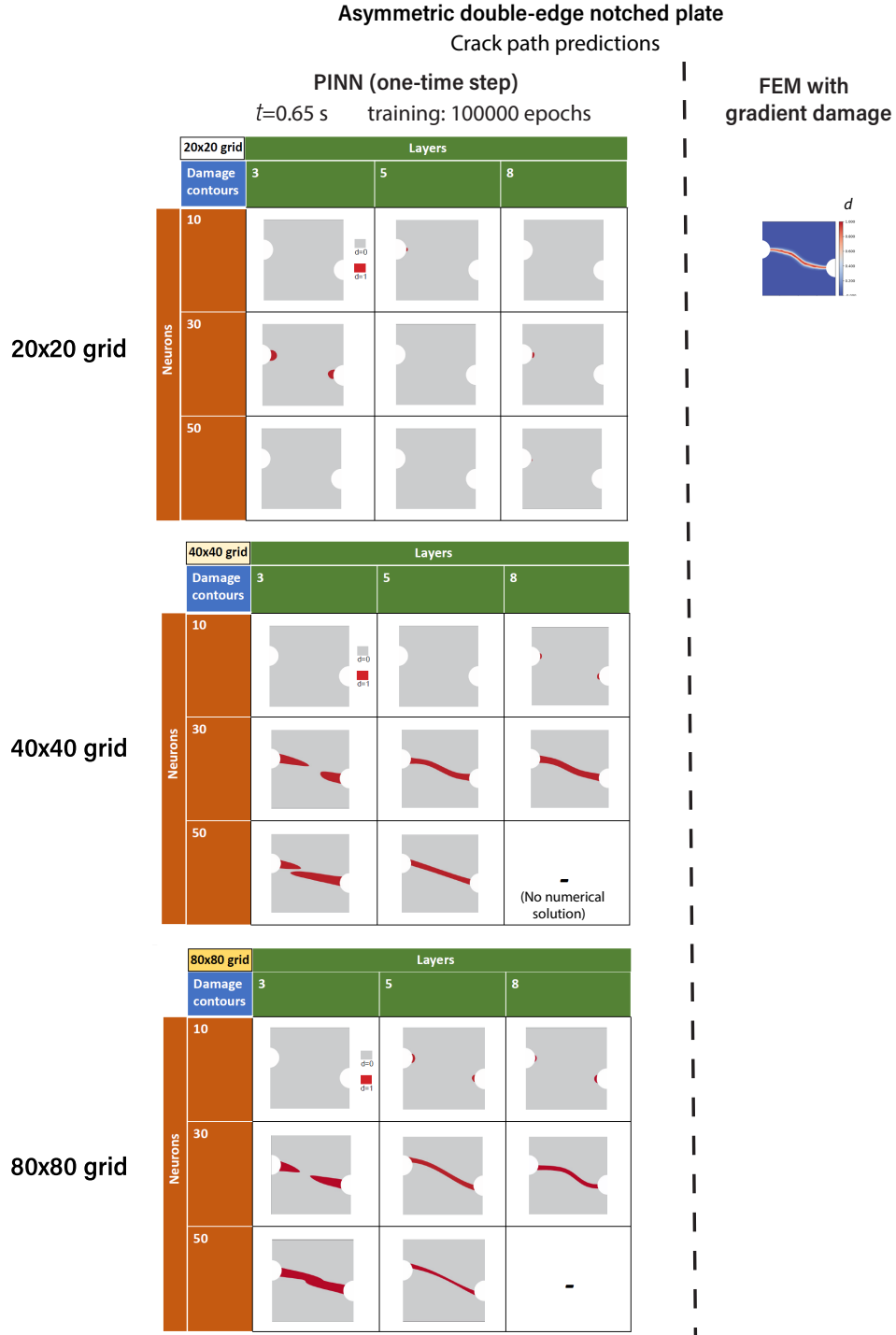


Figure 3.17: **Asymmetric double-edge notched plate: numerical factorial experiment (Crack path, $t=0.65$ s).** PINN (one-time step) is applied at $t = 0.65$ with 100,000 epochs training for the crack path comparison. The crack paths for grid densities of 20×20 , 40×40 , and 80×80 are shown as a function of the number of layers and the number of neurons in each layer. The evolution of the crack path with respect to each hyperparameter considered is non-monotonic. Non-optimal hyperparameter values lead to underfitting or overfitting.

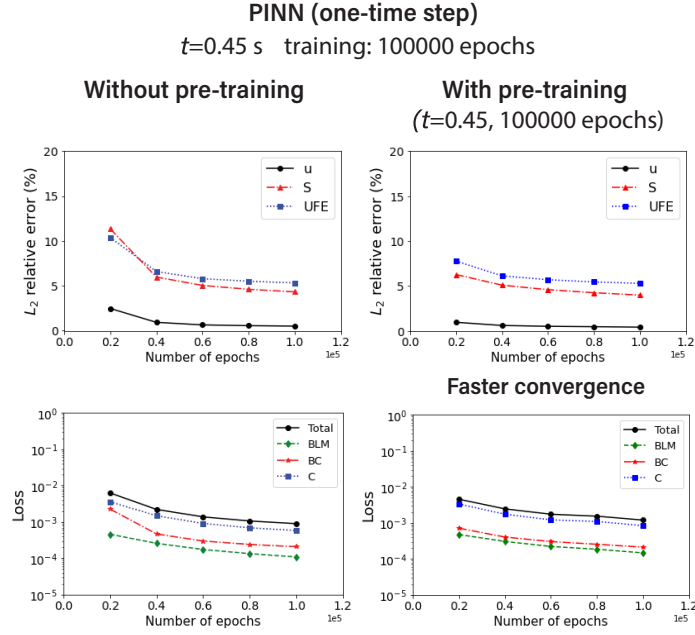
FEM without and with gradient-damage (both implemented in ABAQUS/Explicit (2018)) were parallelized using 32 processors on a system with an Intel Xeon Gold 6326 CPU. For the finite element meshes considered in this chapter ($\sim 10,000$ elements), the approximate computation time for an increment with a time step size of approximately 3×10^{-7} s was 0.003 s for both FEM approaches.

PINN (incremental) in general requires more computation time than FEM. The complexity of computationally implementing gradient-damage models in FEM and the associated implementation time costs are important considerations. Some reduction in computation time for PINN (incremental) can be achieved when the material response is rate-independent. As done in this chapter for an elastomer with hyperelastic response, the solution evaluation in PINN (incremental) for a rate-independent material can be started at any time instant before damage initiation. The computation time for PINN (one-time step), however, can be comparable to FEM.

3.5.7 Effect of pre-training

Pre-training has been shown in the literature to improve the accuracy and convergence of PINNs (Guo et al., 2023; Wang et al., 2025). We investigate the effect of one pre-training strategy. Specifically, the 1×1 domain seeded with a uniform 80×80 collocation point grid is assigned the same material properties and boundary conditions discussed in Section 3.4. The trained network parameters from stretching this unit square ($t = 0.45$ s in one-time step, 100,000 epochs) are used to initialize the PINN for application to the asymmetric double-edge notch plate geometry. Comparison between the PINN (one-time step) results (at $t = 0.45$ s [before damage initiation] and 0.65 s [after damage initiation], 100,000 epochs) with and without pre-training in Figure 3.18 shows the simple pre-training associated convergence and accuracy enhancement.

A



B

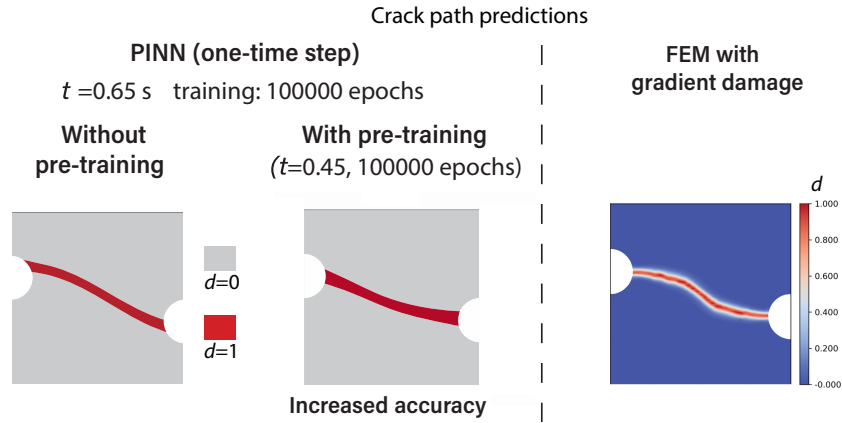


Figure 3.18: **Asymmetric double-edge notched plate: effect of pre-training.** The same material properties and boundary conditions discussed in Section 3.4 were used for pre-training on the 1×1 domain with a uniform 80×80 collocation point grid. **(A)** Comparison of L_2 errors and losses from PINN (one-time step) application at $t = 0.45$ s, 100,000 training epochs with and without pre-training ($t = 0.45$ s, 100,000 epochs). Faster convergence is achieved with pre-training. **(B)** Comparison of crack paths from PINN (one-time step) application at $t = 0.65$ s, 100,000 epochs with and without pre-training ($t = 0.45$ s, 100,000 epochs) and from the baseline FEM with gradient-damage solution. The simple pre-training increases the accuracy of the solution.

3.5.8 Effect of PINN output normalization

Normalization of PINN outputs is another technique that has been shown in the literature to enhance accuracy and convergence (Xu et al., 2024; Rasht-Behesht et al., 2022). We consider a simple normalization scheme in the form of fixed normalization numbers that ensures physical consistency. Specifically, $\{\hat{\mathbf{u}}, \bar{\mathbf{u}}\}$, $\{\hat{\mathbf{P}}, \tilde{\mathbf{P}}^{const}, \bar{\mathbf{t}}_R\}$, $\{\hat{\psi}, \tilde{\psi}^{const}\}$ are normalized by u^{norm} , P^{norm} , ψ^{norm} , respectively before evaluating the loss functions in (3.17), (3.19), (3.20). Figure 3.19 shows the comparison between the PINN (one-time step) results (at $t = 0.45$ s [before damage initiation] - $u^{norm} = 1$, $P^{norm} = 5$, $\psi^{norm} = 5$, 100,000 epochs and 0.65 s [after damage initiation] - $u^{norm} = 2$, $P^{norm} = 10$, $\psi^{norm} = 200$, 100,000 epochs) for the asymmetric double-edge notched plate geometry with and without output normalization. Faster convergence and increased accuracy are achieved with this simple output normalization scheme.

3.5.9 Effect of combining pre-training and output normalization

The two strategies of pre-training and output normalization, and individually shown to improve accuracy and convergence, are now combined. Specifically, for the asymmetric double-edge notched plate ($t = 0.45$ s, 100000 epochs), the pre-training was performed with $u^{norm} = 1$, $P^{norm} = 2$, and $\psi_o^{norm} = 1$. The normalization numbers for training were kept the same as in Section 3.5.8. Figure 3.20 compares the L_2 error evolution with training epochs for only pre-training, only output normalization, and both pre-training and output normalization. A slight decrease in errors is observed when the two strategies are combined using this simple approach.

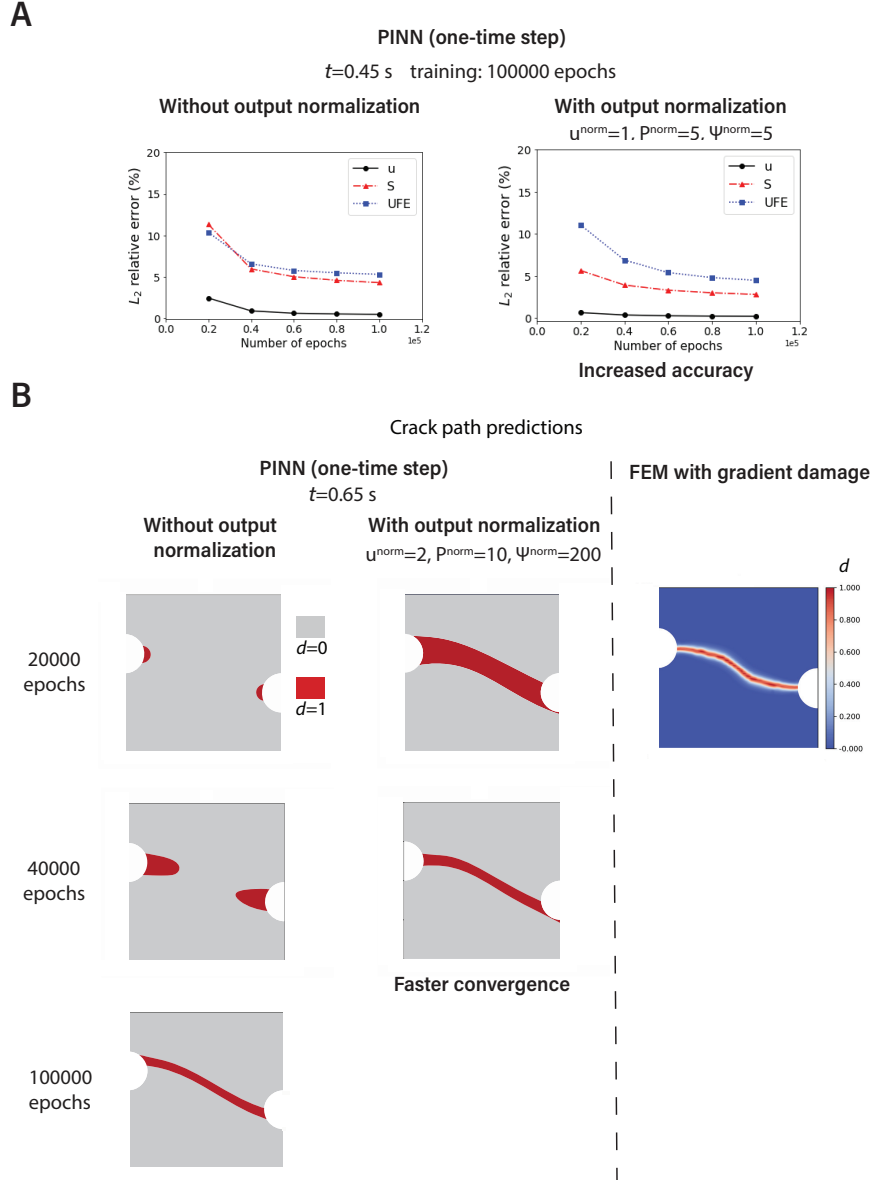


Figure 3.19: **Asymmetric double-edge notched plate: effect of output normalization.** (A) Comparison of L_2 errors from PINN (one-time step) application at $t = 0.45$ s, 100,000 training epochs with and without output normalization ($u^{norm} = 1$, $p^{norm} = 5$, $\psi^{norm} = 5$). Faster convergence and increased accuracy are achieved with output normalization. (B) Comparison of crack paths from PINN (one-time step) application at $t = 0.65$ s, 100,000 epochs with and without output normalization ($u^{norm} = 2$, $p^{norm} = 10$, $\psi^{norm} = 200$) and from the baseline FEM with gradient-damage solution. The simple output normalization led to increased accuracy of the solution and faster convergence.

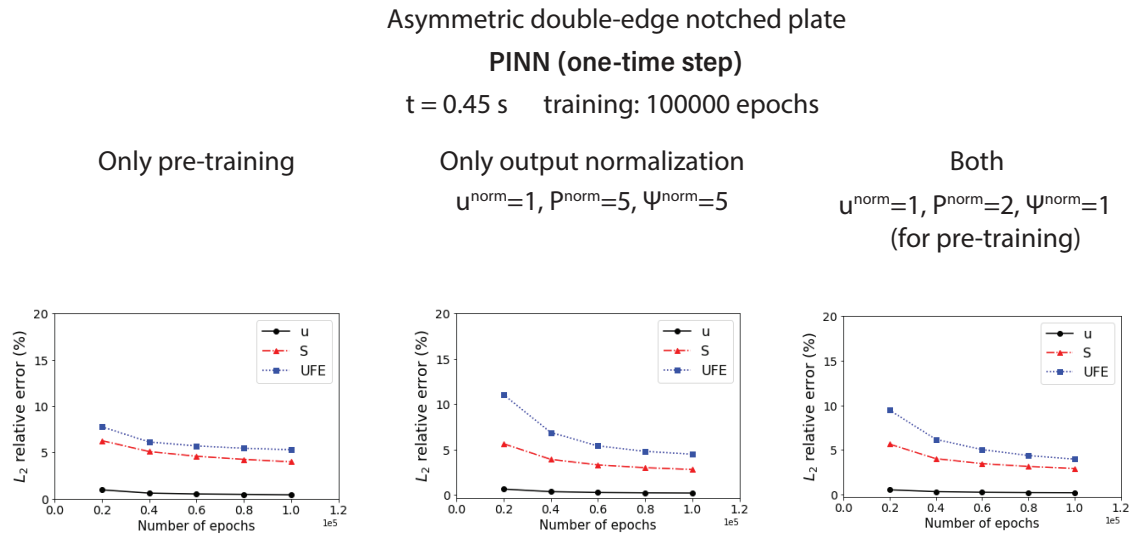


Figure 3.20: **Asymmetric double-edge notched plate: effect of combining pre-training and output normalization.** For PINN (one-time step) application, the solution is evaluated directly at $t = 0.45$ s with 100000 epochs. The pre-training and output normalization techniques, which individually enhanced the PINN performance, are combined through a simple approach. The same normalization parameters used for training in Section 3.5.8 are employed. For pre-training, $u^{\text{norm}} = 1$, $P^{\text{norm}} = 2$, $\psi_o^{\text{norm}} = 1$ were used. Comparison of L_2 error evolution with training epochs using only pre-training, only output normalization, and both. The errors decrease slightly when the two approaches are combined.

3.5.10 Effect of loss weights

The individual loss terms (using all weights = 1) for the asymmetric double-edge notched plate ($t = 0.45$ s, 100,000 training epochs) are not in the same order of magnitude as seen in Figure 3.10. Particularly, the constitutive loss $\mathcal{L}_C$ is significantly higher than the balance of linear momentum (BLM) and the boundary condition (BC) losses. The loss weights α can be adjusted to minimize the total loss further. First, only α_C was increased ($= 10, 100$) to reduce $\mathcal{L}_C$. Figure 3.21(A) shows the corresponding loss and L_2 error evolution with training epochs. $\mathcal{L}_C$ decreased with increasing α_C as anticipated. However, both $\mathcal{L}_{BC}$ and $\mathcal{L}_{BLM}$ increased, with a significant increase in $\mathcal{L}_{BC}$. The total loss also increased along with the L_2 errors. Since $\mathcal{L}_{BC}$ was the most sensitive to an increase in α_C , we also simultaneously increased α_{BC} ($\alpha_C = \alpha_{BC} = 10$ and 100). A similar observation was made with $\mathcal{L}_{BLM}$ increasing with the weights, and hence the total loss increasing as seen in Figure 3.21(B). The L_2 errors changed insignificantly compared to the baseline (all weights = 1) in this case. Finally, the three weights were simultaneously increased ($\alpha_C = \alpha_{BC} = \alpha_{BLM} = 10$ and 100). The corresponding losses and L_2 error evolution were similar to the baseline, as shown in Figure 3.21(C). Hence, it was concluded that the baseline weight values provide a good balance between loss and L_2 error minimization. It can be noted that the optimal loss weight values can vary for different sets of material parameters (e.g., elastic modulus orders of magnitude different from that used in this study). Suitable weights need to be obtained for specific boundary value and material type problems.

3.5.11 Effects of initial learning and decay rates

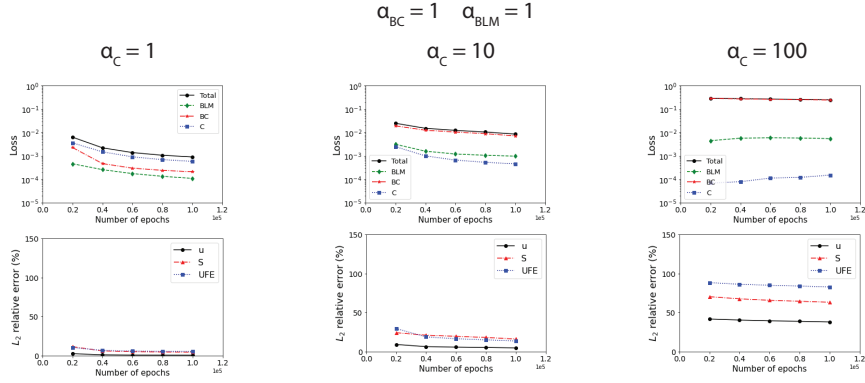
The perturbation of the network parameters after an epoch is determined by the learning rate. High and low learning rates can lead to solution fluctuations (or blowup) and the solution becoming stuck at a local optimum (or slower convergence), respectively. The learning rate is usually set high initially and then decreased with epochs through a

Asymmetric double-edge notched plate

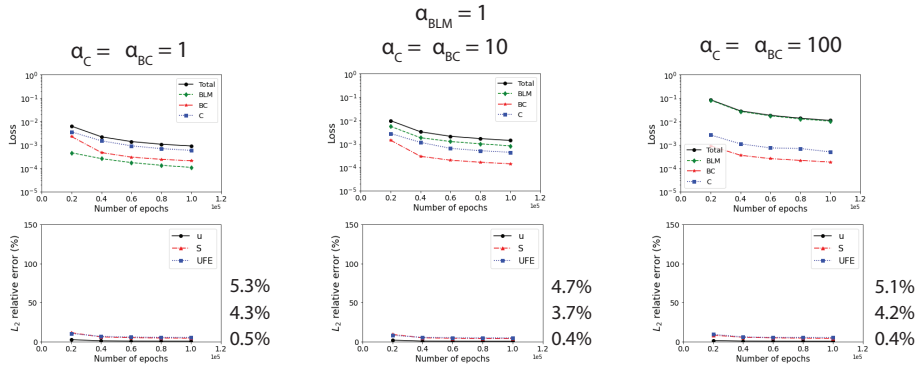
PINN (one-time step)

$t = 0.45$ s training: 100000 epochs

A



B



C

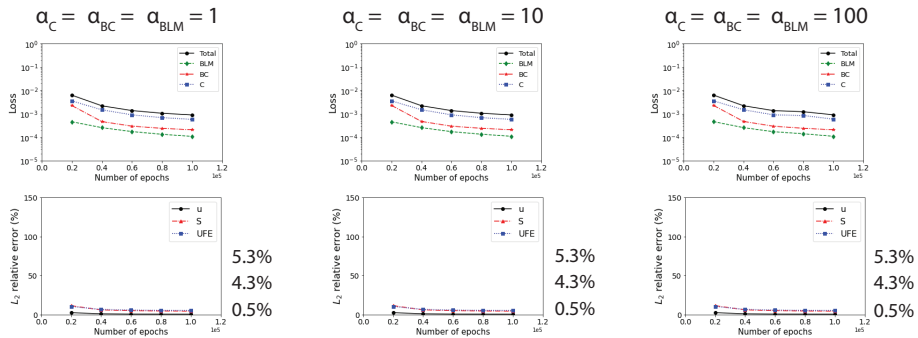


Figure 3.21: **Asymmetric double-edge notched plate: effect of loss weights.** For PINN (one-time step) application, the solution is evaluated at $t = 0.45$ s with 100,000 epochs training. For the baseline weight values $\alpha_C = \alpha_{BC} = \alpha_{BLM} = 1$, $\mathcal{L}_C$ is significantly higher than $\mathcal{L}_{BC}$ and $\mathcal{L}_{BLM}$. The loss and L_2 error evolution with training epochs for **(A)** increase in only α_C , **(B)** simultaneous increase in α_C and α_{BC} , and **(C)** simultaneous increase in all three weights. A common observation is that the loss corresponding to the unchanged weight and the total loss increase upon an increase in the weights. The baseline weight values provide a good balance between loss and L_2 error minimization for the material parameters used in this chapter.

decay rate. The initial learning and decay rate values used were 0.0001 and 0.00000005, respectively. First, the decay rate was fixed at the base value, and positive (0.001) and negative (0.0001) deviations from the base initial learning rate were considered. Figure 3.22(A) shows the corresponding L_2 errors ($t = 0.45$ s) and crack paths ($t = 0.65$ s) for the asymmetric double-edge notched plate geometry. The increased initial learning rate at $t = 0.65$ s led to a solution that blew up, i.e., no numerical solution. On the other hand, the reduced initial learning rate at $t = 0.45$ s resulted in slower convergence. Then, with the initial learning rate fixed at the base value, the results from the positive and negative deviations for the decay rate are shown in Figure 3.22(B). The effects of the decay rate are not easily discernible.

3.5.12 Effect of random collocation grid structure

A random collocation grid structure can enhance the accuracy of PINNs in some cases (Florido et al., 2024) by ensuring that the entire domain is well sampled. Figure 3.23 compares the results ($t = 0.45$ s, 100000 training epochs: L_2 error and loss evolution, and $t = 0.65$ s, 100000 training epochs: final crack path) for the asymmetric double-edge notched plate geometry with a structured and random grid. No significant differences are observed in the results from the two grid structures.

3.5.13 Effect of pre-training with randomly sampled material properties

Pre-training with randomly sampled material parameter values can enhance the robustness of PINNs by exploring the parameter space (Yin et al., 2021; Wu et al., 2023a). Pre-training was performed on a unit square ($t=0.45$ s, 50000 epochs) with the grid shown in Figure 3.24. G and ν were randomly sampled into five sets as $\{G=3.81, \nu=0.22\}$, $\{G=7.65, \nu=0.32\}$, $\{G=7.95, \nu=0.35\}$, $\{G=1.87, \nu=0.37\}$, $\{G=4.89, \nu=0.14\}$. Pre-training was performed sequentially, with the trained network parameters from the first

Asymmetric double-edge notched plate

PINN (one-time step)

training: 100000 epochs

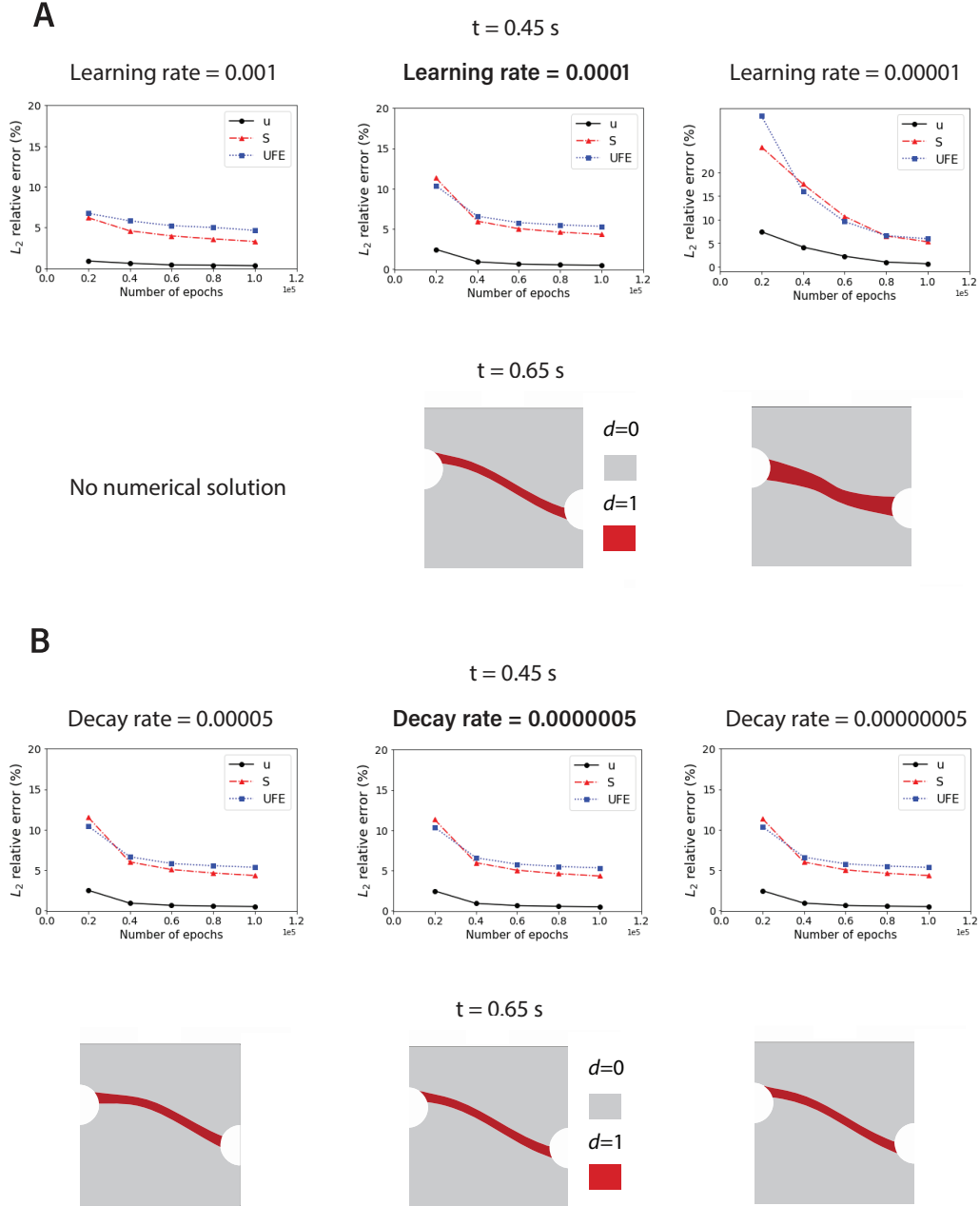


Figure 3.22: **Asymmetric double-edge notched plate: effects of initial learning and decay rate.** For PINN (one-time step) application, the solution is evaluated directly at $t = 0.45$ s and $t = 0.65$ s with 100000 epochs. The L_2 errors and final crack path solutions for variations in (A) the initial learning rate and (B) the decay rate from the base values used. A higher initial learning rate led to the solution blowing up at $t = 0.65$ s. A lower initial learning rate resulted in slower convergence at $t = 0.45$ s. The effects of the decay rate are not significant.

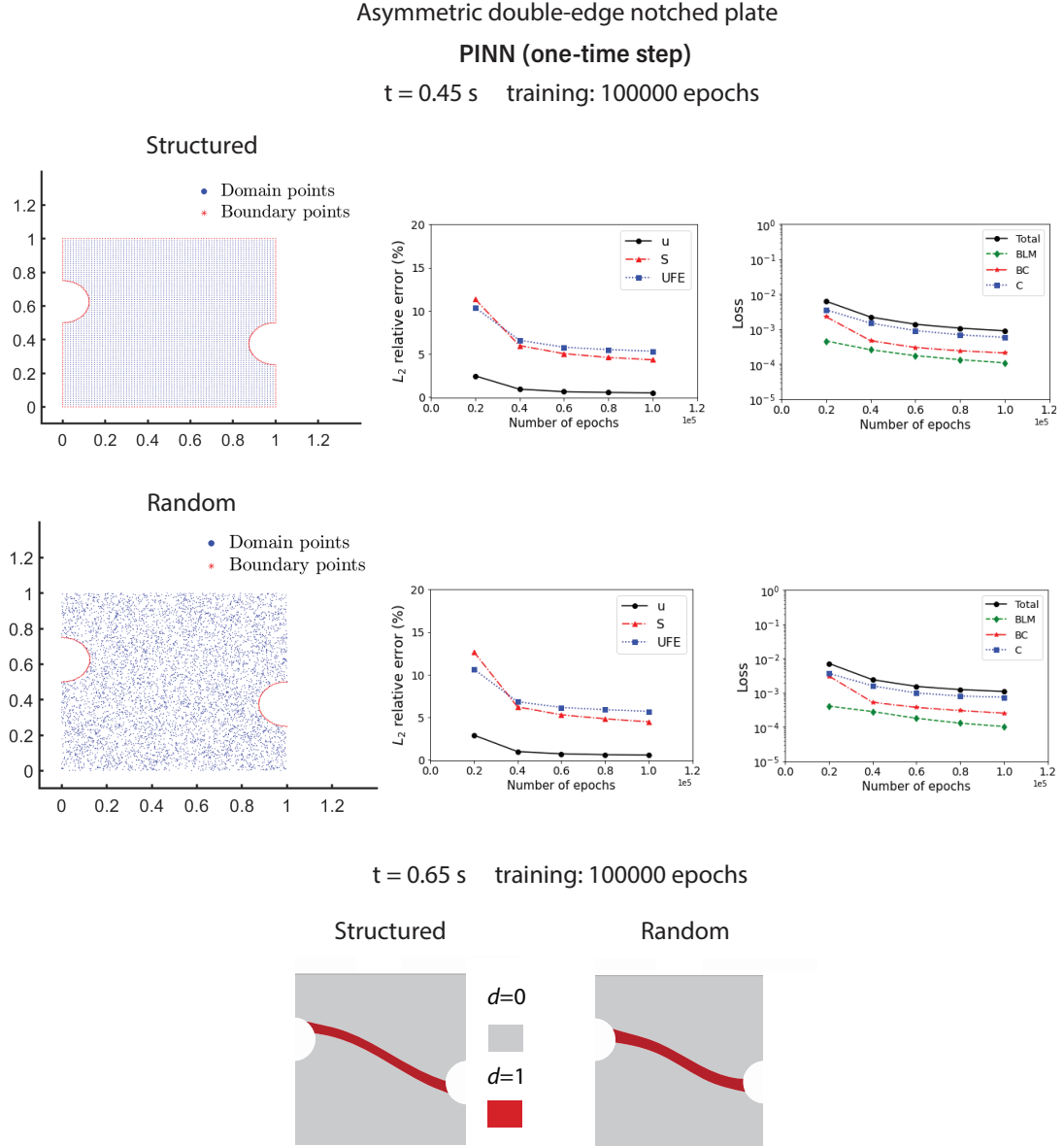


Figure 3.23: **Asymmetric double-edge notched plate: effect of random grid structure.** For PINN (one-time step) application, the solution is evaluated directly at $t = 0.45$ s and $t = 0.65$ s with 100000 epochs training. L_2 error and loss evolution with training epochs ($t = 0.45$ s) and final crack path ($t = 0.65$ s) for (A) structured (Grid 1 in Figure) and (B) random grid structures. The results from the two grid structures do not differ significantly.

material parameter set used to initialize the second set, and so on. The final pre-training network parameters were used to initialize training for the asymmetric double-edge notched plate ($t=0.45$ s, 100000 epochs, uniform grid). Comparison of L_2 error and loss evolution with and without pre-training is shown in Figure 3.24. The L_2 errors and losses increased slightly with this pre-training approach.

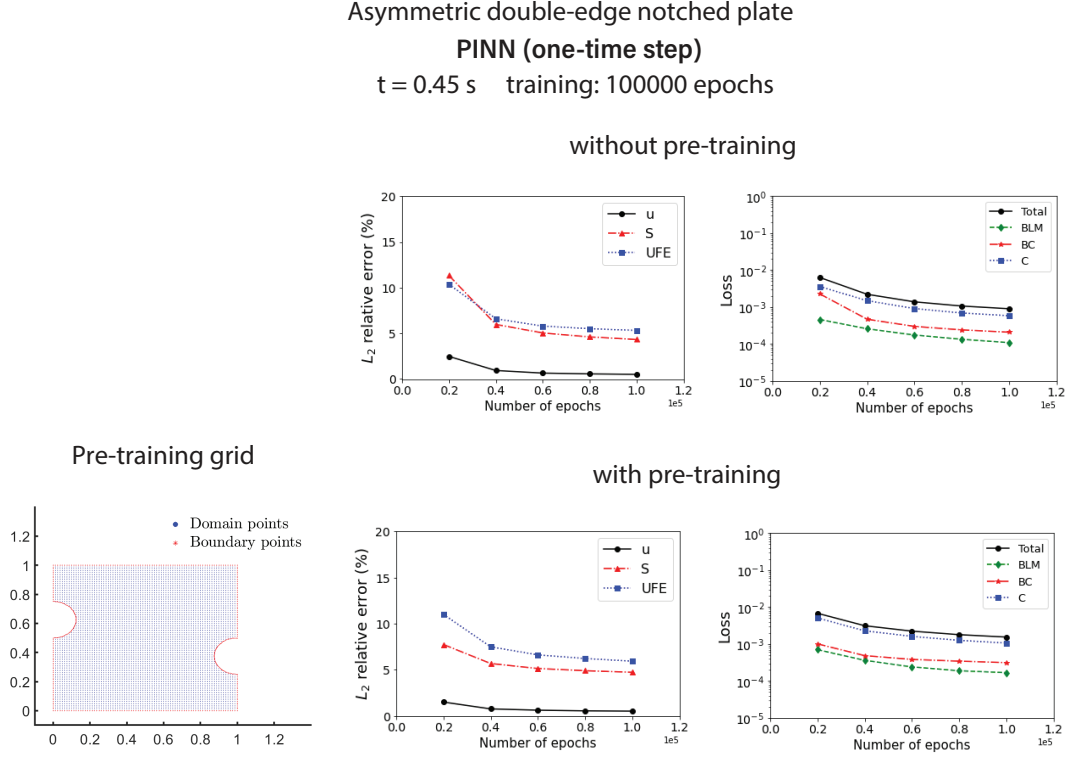


Figure 3.24: **Asymmetric double-edge notched plate: effect of pre-training with randomly sampled material parameters.** For PINN (one-time step) application, the solution is evaluated directly at $t = 0.45$ s with 100000 epochs training. Grid used for pre-training on a unit square ($t=0.45$ s, 50000 epochs). G and ν were randomly sampled into five sets, and pre-training was performed sequentially on them. L_2 error and loss evolution with training epochs with and without pre-training. Solution quality declined slightly with this training approach.

3.5.14 Residuals and grid point redistribution

Redistributing grid points to regions with high residuals can enhance the accuracy of PINNs (Wu et al., 2023b). S ($P_{xx} + P_{yy} + P_{xy} + P_{yx}$) and UFE residuals are defined as the absolute value of the difference between the PINN and the constitutive predictions. For the asymmetric double-edge notched plate ($t=0.65$ s, 50000 epochs, uniform grid), the S and UFE residuals are shown in Figure 3.25. The residuals are not highly localized and well distributed, highlighting good solution quality. The non-local residual distribution indicates that simple grid redistribution is infeasible, and more sophisticated techniques are required to improve solution quality further.

Asymmetric double-edge notched plate

PINN (one-time step)

$t = 0.65$ s 50000 epochs

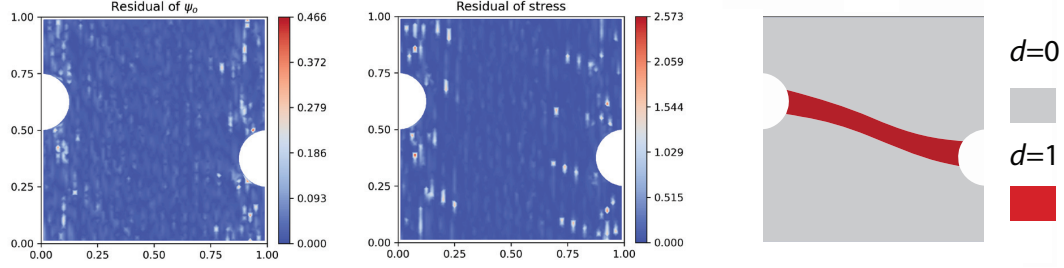


Figure 3.25: **Asymmetric double-edge notched plate: residuals and grid point re-distribution.** For PINN (one-time step) application, the solution is evaluated directly at $t = 0.65$ s with 50000 epochs training. Well-distributed residuals indicate good solution quality. Sophisticated grid redistribution techniques can further enhance the solution quality.

3.6 PINN application to a biological tissue

To demonstrate the broader applicability of the PINN, we apply it to predict the fracture of a biological material. Specifically, the peel test of a circumferentially oriented human abdominal aorta strip is modeled (see Figure 3.26(A)). The dissection of aortic media is a challenging clinical problem, and the response was characterized to understand the underlying mechanics and modeled in (Sommer et al., 2008) and (Raina and Miehe, 2016), respectively. The base neural network structure and parameters discussed in Section 3.4 are used. The transversely isotropic, plane strain specimen has dimensions of 30 mm x 8 mm x 1.2 mm (Sommer et al., 2008) and an initial cut size of 0.4 mm (Raina and Miehe, 2016). A notch radius of 0.1 mm is assumed for computational tractability. Only the left half of the strip is considered with appropriate boundary conditions, exploiting the symmetry of the problem. The collocation grid used and problem schematic are shown in Figure 3.26(B). With the constitutive model and deformation-related parameters from (Raina and Miehe, 2016), ψ_{cr}^+ is calibrated to the ψ^+ (at the notch center) at the start (see Figure 3.26(C)) of the approximately constant peeling force/width region. Figures

3.26(C) and (D) show the good agreement of PINN predicted peeling force/width vs. displacement curve with the experimental result and a zoomed-in view of the qualitatively consistent (with experiments) crack path prediction near the notch, respectively. The broader applicability of the PINN is hence highlighted.

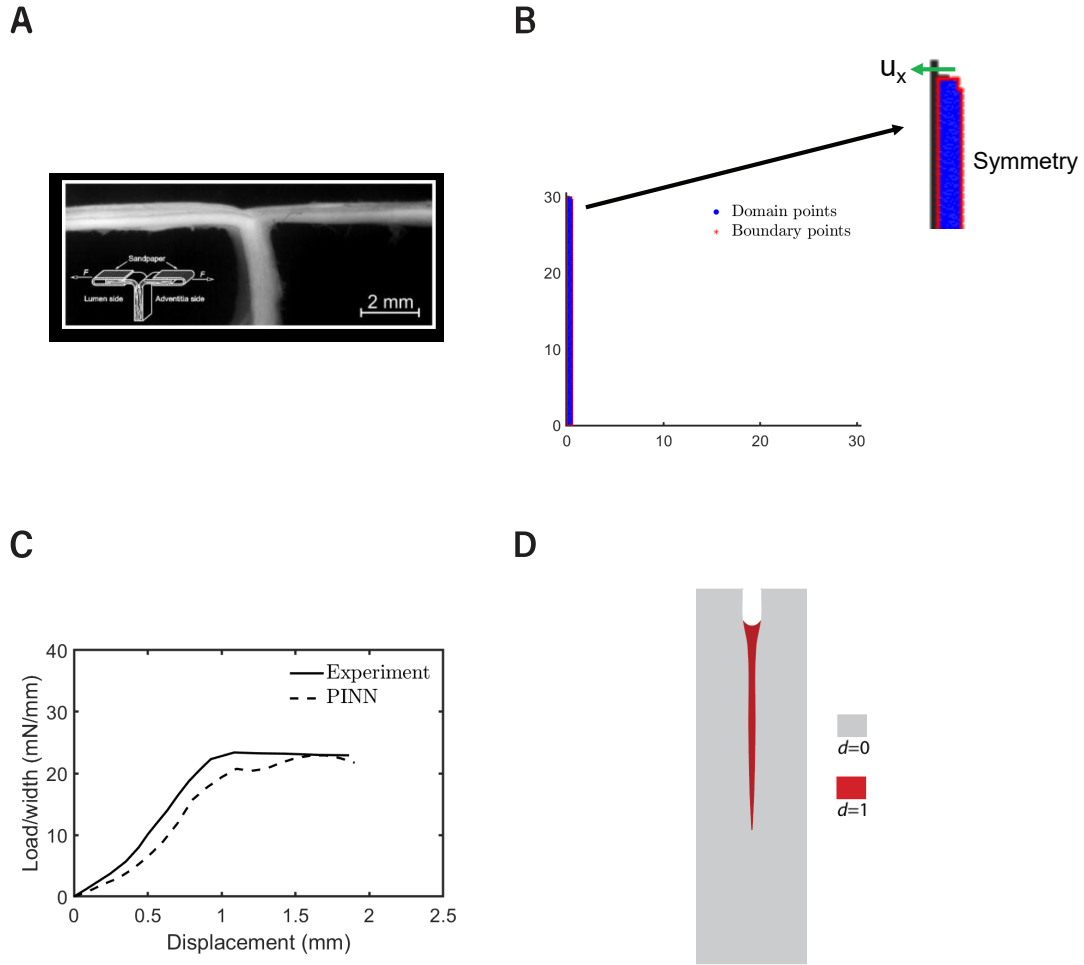


Figure 3.26: **PINN application to a biological material.** (A) Peel test to understand the underlying mechanics of aortic media dissection (Sommer et al., 2008). (B) The collocation grid used and problem schematic. Only the left half is considered due to the problem's symmetry. The circumferentially oriented human abdominal aorta strip (transversely isotropic, plane strain) dimensions are 30 mm x 8 mm x 1.2 mm (Sommer et al., 2008) with an initial cut size of 0.4 mm (Raina and Miehe, 2016). A notch radius of 0.1 mm is assumed for computational tractability. (C) The constitutive model and deformation-related parameters from (Raina and Miehe, 2016) are used. ψ_{cr}^+ is calibrated to the ψ^+ (at the notch center) at the start of the approximately constant peeling force/width region. The PINN predicted peeling force/width vs. displacement curve agrees well with the experimental result, highlighting the PINN's broader applicability. (D) Zoomed-in view of the crack path prediction near the notch, which is qualitatively consistent with the experiments.

3.7 Conclusion

The important and evolving field of fracture modeling commonly uses the finite element method (FEM). FEM without gradient-damage is relatively easy to implement. However, the corresponding results are highly mesh-dependent and often inaccurate. FEM with gradient-damage enables mesh-independent predictions at the cost of increased mathematical and numerical implementation complexities. Specifically, additional spatial gradient calculations and complex user-defined element and user subroutine development are necessary. Physics-informed neural networks (PINNs) have recently emerged as a computational method for solving solid mechanics problems.⁴ The limited studies on PINNs for damage and fracture modeling in the literature are confined to the small-strain regime and use the gradient-damage formulation by default. Due to the meshless nature of PINNs, we posed the question of whether we can correctly solve fracture problems without needing the gradient-damage formulation. We propose a PINN without gradient-damage to model large deformation fracture in elastomers. The PINN does not require training data and utilizes the collocation method. The capability of the PINN to accurately predict the crack path has been validated by studying a variety of benchmark defect configuration problems, where we used accurate solutions from FEM with gradient-damage and a highly refined mesh. We also conducted an assessment of the PINN as a fracture modeling method by systematically varying a few key parameters to guide future design of PINNs for fracture problems. The PINN crack path predictions are largely insensitive to the collocation point distribution. The proposed modeling framework and neural network architecture could motivate the application of PINNs without gradient-damage for modeling fracture in a broad class of materials. This is demonstrated through the PINN’s successful application to a biological tissue.

⁴Chapter 4 presents the results from an attempt to develop a PINN for modeling large deformation contact in elastomers.

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CHAPTER 4

A physics-informed neural network for modeling large deformation contact in elastomers

4.1 Abstract

Elastomers are important engineering materials with high elasticity and applications ranging from sealing to wearable technology, where they frequently undergo large-deformation contact with rigid surfaces. Also, feasible material characterization experiments for very soft materials involve rigid surface indentation and, hence, contact. While the finite element method (FEM) is the traditional modeling standard for these interactions, physics-informed neural networks (PINNs) have emerged as an alternative numerical method for mechanics problems. The application of PINNs for contact problems is relatively new and limited, with a primary focus on small deformations. Recently, a surface potential-based PINN has been proposed for modeling large deformation contact with a relatively complex contact formulation. The mixed PINN formulation, automatic differentiation (AD), and Python geometric packages remain underutilized, which can

enable simple and efficient PINNs for modeling large deformation contact. We present a PINN with a mixed formulation for modeling large deformation frictionless contact between elastomeric and rigid bodies. Since geometric packages that can directly output intersection measures currently do not support Autograd, we utilized the gap function approach from FEM. Several approaches using AD are proposed and assessed to evaluate the tangential vector on the deformed body’s surface necessary for enforcing the frictionless conditions. The PINN’s predictive capabilities have been validated for various contact surface configurations using baseline solutions from FEM. The approach using the deformation gradient for evaluating the deformed tangential vector produces the best results. A preliminary extension of the formulation to elastomeric deformable-deformable body frictionless contact is also discussed. The proposed simplistic modeling framework and neural network architecture should motivate PINNs to model large deformation contact in a variety of materials.

4.2 Introduction

Elastomers are a class of soft polymers with important applications, which include automotive tires (Lei et al., 2016), seals and gaskets (Al-Hiddabi et al., 2015), footwear (Ames, 2004), and wearable consumer electronics (Chakraborti et al., 2025). They are characterized by high elasticity and are often subjected to large deformation contact with rigid bodies during their applications, e.g., sealing. For very soft materials, feasible experiments for material response characterization usually involve indentation of a rigid surface and hence contact (Duncan et al., 2020; Montanari et al., 2023). The conventional finite element method (FEM) has been extensively used to model elastomeric-rigid body contact (Wriggers and Reinelt, 2009; Mansouri et al., 2017).

Physics-informed neural networks (PINNs) have emerged in the recent literature as a numerical method for solving mechanics problems (Niu et al., 2023; Jin et al., 2023). PINNs can solve partial differential equations (PDEs) by encoding the mathematical information

explicitly into the loss functions without a need for training data. We have summarized the application of PINNs for mechanics and their advantages and disadvantages with respect to the conventional numerical methods (e.g., FEM) in our recent work (Konale and Srivastava, 2026) and Chapter 3.

The application of PINNs for contact problems in mechanics is gaining traction (Sahin et al., 2024; Bai et al., 2025; Zhou and Song, 2025; Zhao et al., 2025). Although the examples are limited, they primarily focus on the small deformation regime (Sahin et al., 2024; Xia and Meng, 2024; Zhou and Song, 2025). Bai and co-workers (Bai et al., 2025) developed a PINN for modeling frictionless contact problems under large deformation. Specifically, they employed an energy-based formulation with a relatively complex surface contact potential to prevent body penetration. The PINN is shown to perform well on a Hertzian benchmark problem and on more complex problems involving material and geometric nonlinearities. However, the capabilities of the mixed PINN formulation (stresses as outputs along with displacements), automatic differentiation (AD), and Python geometric packages have not been fully utilized for contact problems. These should yield relatively simple and efficient PINNs for modeling large deformation contact.

A simple PINN-based approach to contact problems is to formulate a loss function that also minimizes the intersection area/volume of geometries during the solution procedure. The geometric packages in Python can play a crucial role here. An example is the Shapely package in Python, which returns the intersection area of geometries input as arrays of coordinates. However, Shapely, along with similar packages, does not currently support Autograd (which is necessary for backpropagation to optimize the neural network parameters). Although it is possible to define custom Autograd functions, it may not be straightforward for a user who is not well-versed in the relevant mathematical background. Hence, the gap function approach (i.e., minimizing point-wise penetration into a master surface) used in FEM was deemed tractable for this study. The following contributions are presented here:

(i) A large deformation PINN with mixed formulation for modeling rigid-elastomeric deformable body frictionless contact utilizing our recent work ([PINN for fracture (Konale and Srivastava, 2026)], Chapter 3). Various AD-based approaches are presented and assessed for evaluating the tangential vector on the deformed body’s surface required to enforce the frictionless condition. Although friction is an important consideration for accurate contact modeling in practical scenarios, we have considered frictionless conditions to demonstrate the PINN’s capabilities.

(ii) Validation of the PINN’s predictive capabilities for various contact surface configurations using baseline solutions obtained from FEM. The proposed simplistic modeling framework and neural network architecture should motivate PINNs to model large deformation contact in a variety of materials.

(iii) A preliminary extension of the formulation to elastomeric deformable-deformable body frictionless contact.

4.3 Rigid body - deformable body frictionless contact under large deformation

4.3.1 Constitutive equations

The proposed PINN is applied to elastomeric deformable bodies here to demonstrate its predictive capabilities. The deformation-related constitutive equations from Chapter 3 (Section 3.2) are used directly.

4.3.2 Contact modeling

As discussed in the Introduction, the gap function approach is used to prevent body penetration. If an analytical function for the rigid body’s outer surface (Γ_{rigid}) is known (or approximated), the penetrating points on the deformable body’s outer surface (Γ_{df}) during

the solution procedure can be identified (Γ_{df}^{pen}). It can be noted here that $\Gamma_{df} - \Gamma_{df}^{pen}$ ($-\Gamma_{df}^u$ if some part of Γ_{df} is prescribed displacement boundary conditions) has to be prescribed traction-free boundary conditions. Treating Γ_{rigid} as the master surface, a gap function (g) can be defined to quantify the penetration of Γ_{df}^{pen} (see Figure 4.1) as

$$g = |\mathbf{x} - \mathbf{x}_c|, \mathbf{x} \in \Gamma_{df}^{pen}, \quad (4.1)$$

where $\mathbf{x}_c$ is the closest projection of $\mathbf{x}$ on the rigid surface. To impose the frictionless contact condition, the unit tangential vectors $\mathbf{e}_t$ in the deformed configuration have to be evaluated on Γ_{df} . The contact conditions enforcement can then be summarized as:

$$\begin{aligned} \text{No penetration} - g(\mathbf{x}) &= 0, \mathbf{x} \in \Gamma_{df}^{pen}, \\ \text{Frictionless contact} - \mathbf{t}_R(\mathbf{x}) \cdot \mathbf{e}_t(\mathbf{x}) &= 0, \mathbf{x} \in \Gamma_{df}^{pen}. \end{aligned} \quad (4.2)$$

In the above equation, for frictionless contact, the tangential component (with respect to $\mathbf{e}_t$) of $\mathbf{t}_R$ (referential traction vector) is set equal to zero. It can be noted here that $\mathbf{t}_R$ has the same direction as its spatial counterpart ($\mathbf{t}$); they differ by the area used for the elemental force normalization. Since the tangential component is set to zero, the choice of $\mathbf{t}_R$ vs. $\mathbf{t}$ is irrelevant. We choose $\mathbf{t}_R$ here since the problems are formulated referentially.

4.3.3 Approaches for evaluating $\mathbf{e}_t$

Two generic approaches exploiting AD are considered for evaluating $\mathbf{e}_t$ in planar geometries. The third approach is a simplification for specific cases.

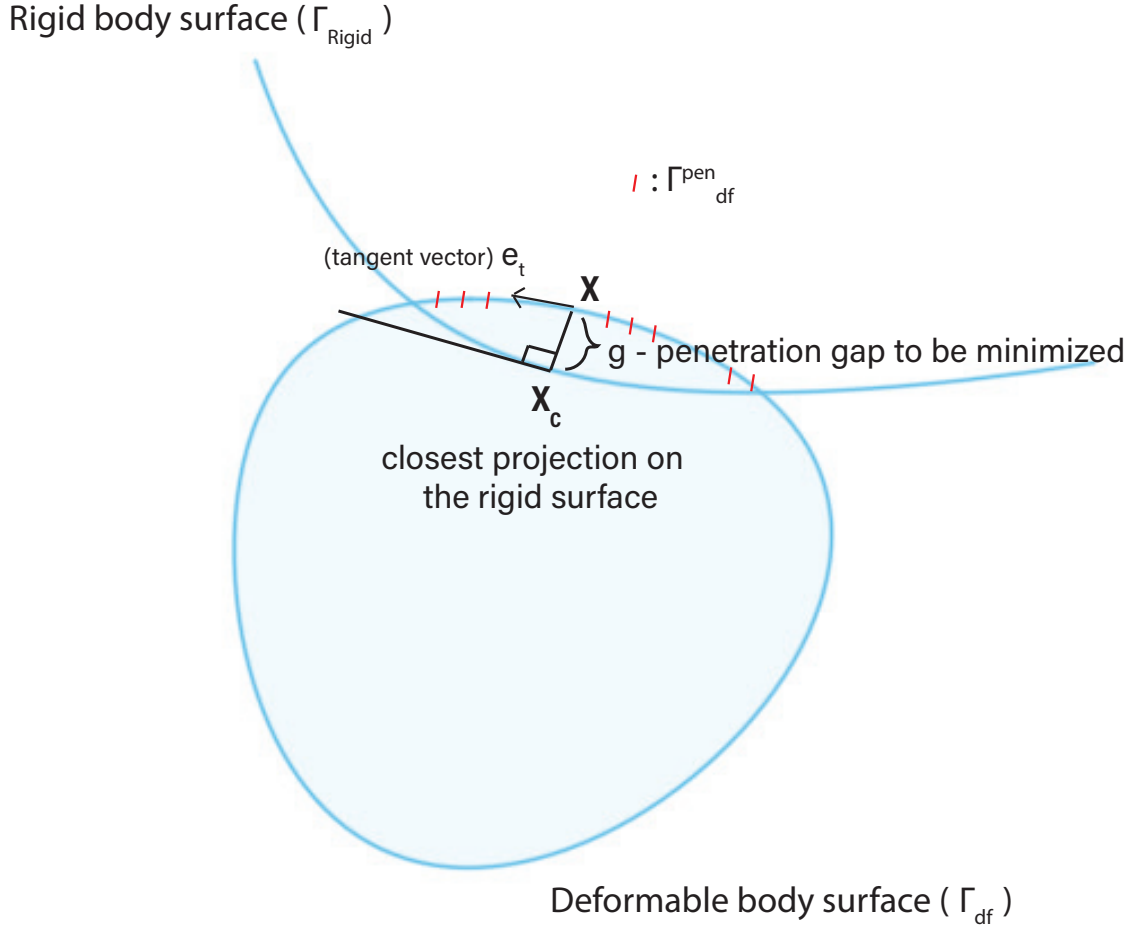


Figure 4.1: **Rigid body - deformable body contact schematic.** A possible instant during the solution procedure is shown where the deformable body's outer surface (Γ_{df}) penetrates the rigid body's outer surface (Γ_{rigid}). For points on the penetrating part of Γ_{df} (Γ_{df}^{pen}), a gap function (g) is defined as the distance from the closest projection on Γ_{rigid} , which will be minimized. Further, to enforce the frictionless contact condition, the spatial tangent vectors ($\mathbf{e}_t$) for the points on Γ_{df}^{pen} are necessary. Subsequently, the tangential component of $\mathbf{t}_R$ i.e. $\mathbf{t}_R \cdot \mathbf{e}_t$ will be set equal to zero on Γ_{df}^{pen} .

dy/dx approach

The slope of the tangent at any point $\mathbf{x} = \{x_1, x_2\} \in \Gamma_{df}^{pen}$ can be evaluated as $\frac{dx_2}{dx_1} \cdot \mathbf{e}_t$ can be then expressed as

$$\mathbf{e}_t = \cos\theta \mathbf{e}_1 + \sin\theta \mathbf{e}_2, \quad \theta = \tan^{-1}\left(\frac{dx_2}{dx_1}\right). \quad (4.3)$$

It can be noted here that since $x_1 = x_1(X_1, X_2)$, $x_2 = x_2(X_1, X_2)$,

$$\frac{dx_2}{dx_1} = \frac{\frac{\partial x_2}{\partial X_1}}{\frac{\partial x_1}{\partial X_1}} + \frac{\frac{\partial x_2}{\partial X_2}}{\frac{\partial x_1}{\partial X_2}}. \quad (4.4)$$

Each of the four partial differentiation terms in the above equation can be evaluated easily using AD and $x_1 = X_1 + u_1$, $x_2 = X_2 + u_2$.

Ftang approach

From the definition of $\mathbf{F}$ we get,

$$\mathbf{e}_t(\mathbf{x}) = \mathbf{F}(\mathbf{X})\mathbf{e}_t^R(\mathbf{X}), \quad \mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}}, \quad (4.5)$$

where $\mathbf{e}_t^R$ is the referential unit tangent vector, i.e. $\mathbf{e}_t^R$ is mapped to $\mathbf{e}_t$ through the deformation $\mathbf{F}$. $\mathbf{e}_t^R$ is known at each $\mathbf{X}$ from the reference deformable body geometry. $\mathbf{F}$ can be easily evaluated using AD and $\mathbf{F} = \mathbf{1} + \frac{\partial \mathbf{u}}{\partial \mathbf{X}}$.

Ideal tangent approach

Consider a scenario where the rigid body surface Γ_{Rigid} has a globally unique tangent vector ($\mathbf{e}_t^{Rigid}$). A simplified approach to evaluate $\mathbf{e}_t$ is to set it equal to the ideal (zero penetration, $g = 0$) tangent vector $\mathbf{e}_t^{rigid}$ (which is known when the rigid body's motion is

prescribed). An example is shown in Figure 4.2. This approach should be suitable when the penetration is small during the solution procedure so that $|\mathbf{e}_t - \mathbf{e}_t^R| \approx 0$.

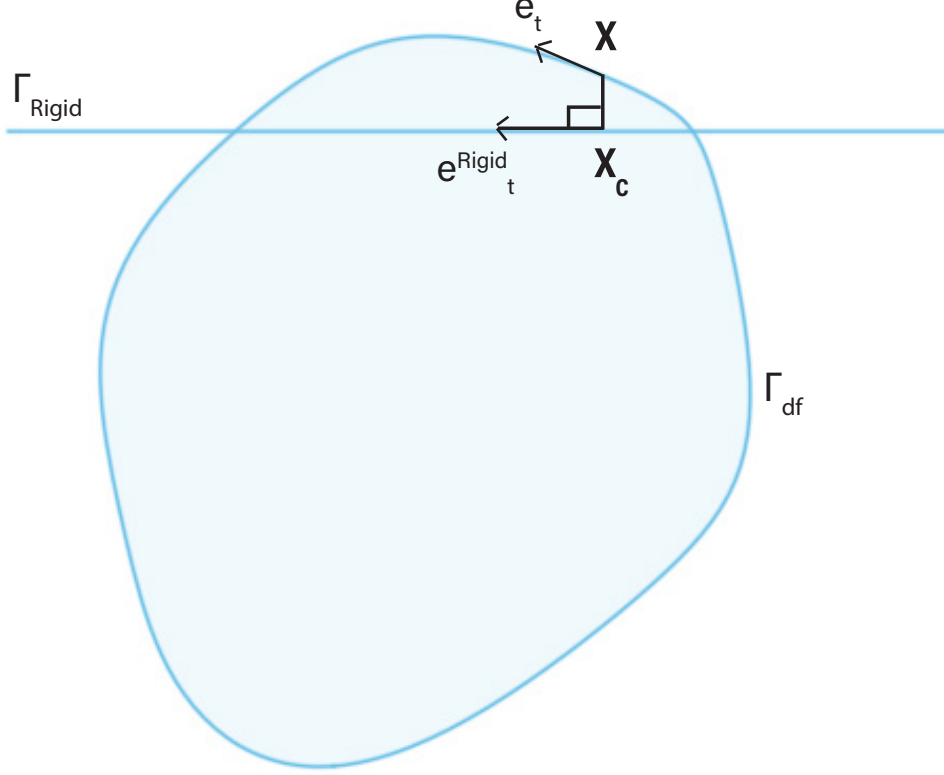


Figure 4.2: **Ideal tangent approach.** A possible instant during the solution procedure is shown during a scenario where Γ_{Rigid} has a globally unique tangent ($\mathbf{e}_t^{Rigid}$) during deformation. When g is small, i.e., $|\mathbf{e}_t - \mathbf{e}_t^R| \approx 0$, $\mathbf{e}_t = \mathbf{e}_t^{Rigid}$ (the ideal tangent vector for zero penetration/ $g=0$, known when the rigid body's motion is prescribed) can be a good simplifying approximation.

4.3.4 Neural network architecture

The PINN formulation is similar to that used for fracture modeling in Chapter 3 (Section 3.3).¹ Two-dimensional problems are solved using the plane strain assumption. The problems are cast in a Lagrangian framework with $\mathbf{X} = \{X_1, X_2\}$ denoting the coordinates of a material point in the reference configuration. A mixed formulation is used wherein the neural network approximates the first Piola-Kirchhoff stress field $\hat{\mathbf{P}}(\mathbf{X}, t)$ along with the displacement field $\hat{\mathbf{u}}(\mathbf{X}, t)$. All trainable PINN parameters, including the

¹Due to the similar formulation and neural network architecture, the PINN performance assessment insights from Chapter 3 (Section 3.5) for the deformation-only cases should be applicable here.

weights and biases in the neurons of the network, are denoted by the vector $\boldsymbol{\theta}$. The PINN formulation can then be written as

$$(\hat{\mathbf{u}}, \hat{\mathbf{P}}, \hat{\psi}) = \mathcal{NN}(\mathbf{X}, t, \boldsymbol{\theta}), \quad (4.6)$$

with $\hat{\cdot}$ denoting direct outputs of the neural network. Specifically, $\hat{\mathbf{u}} = \{u_1, u_2\}$, $\hat{\mathbf{P}} = \{P_{11}, P_{12}, P_{21}, P_{22}\}$. $\hat{\psi}$ is the approximated undamaged free energy field. Although not necessary for the problems considered here, $\hat{\psi}$ will be needed to model damage for potential future applications and is therefore retained.

Loss function construction

The construction of the loss function is again similar to that for the PINN for fracture in Chapter 3 (Section 3.3). It can be summarized as:

$$\mathcal{L}_{Total} = \alpha_{BLM}\mathcal{L}_{BLM} + \alpha_{BAM}\mathcal{L}_{BAM} + \alpha_{BC}\mathcal{L}_{BC} + \alpha_C\mathcal{L}_C + \alpha_{CNT}\mathcal{L}_{CNT}. \quad (4.7)$$

$\mathcal{L}_{Total}$, $\mathcal{L}_{BLM}$, $\mathcal{L}_{BAM}$, $\mathcal{L}_{BC}$, $\mathcal{L}_C$ denote the total, individual loss functions for the balance of linear momentum, the balance of angular momentum, boundary conditions, constitutive relations, and contact conditions respectively. α 's are the corresponding weights. The spatial and temporal discretization scheme from Chapter 3 (Section 3.3) is used. During PINN training for a time instant t^j , the network parameters $\boldsymbol{\theta}^j$ are optimized to minimize the total loss function ($\mathcal{L}_{Total}$),

$$\check{\boldsymbol{\theta}}^j = \underbrace{\operatorname{argmin}}_{\boldsymbol{\theta}^j} \mathcal{L}_{Total}(\boldsymbol{\theta}^j). \quad (4.8)$$

The expanded form of the contact loss function that penalizes errors in the collocation equations in the least-squares sense can be written as (forms for the *BLM*, *BAM*, *BC*, and *C* losses are the same as in Chapter 3 [Section 3.3])

$$\begin{aligned}
\mathcal{L}_{CNT} &= \alpha_G \mathcal{L}_G + \alpha_{FRL} \mathcal{L}_{FRL} \\
&= \frac{\alpha_G}{N_{CNT}} \sum_{i=1}^{N_{CNT}} |\mathbf{X}_{CNT}^i + \hat{\mathbf{u}}(\mathbf{X}_{CNT}^i, t^j, \boldsymbol{\theta}^j) - \tilde{\mathbf{x}}_c(\mathbf{X}_{CNT}^i, t^j, \boldsymbol{\theta}^j)|^2 \\
&\quad + \frac{\alpha_{FRL}}{N_{CNT}} \sum_{i=1}^{N_{CNT}} |[\hat{\mathbf{P}}(\mathbf{X}_{CNT}^i, t^j, \boldsymbol{\theta}^j) \mathbf{n}_R(\mathbf{X}_{CNT}^i)] \cdot \tilde{\mathbf{e}}_t(\mathbf{X}_{CNT}^i, t^j, \boldsymbol{\theta}^j)|^2,
\end{aligned} \tag{4.9}$$

with subscripts *G*, *FRL* denoting terms associated with the gap function (penetration), and frictionless contact condition, respectively. $\mathbf{X}_{CNT}$ are the points on Γ_{df}^{pen} and N_{CNT} denoting their number. The approximate PINN solution for the deformable body at a referential point $\mathbf{X}$ and time instant t^j can then be evaluated for post-processing following the process discussed in Chapter 3 (Section 3.3). The architecture of the PINN is summarized in Figure 4.3. For the problems considered here, the inertial terms are negligible and are therefore neglected. In the baseline FEM solutions, we have incorporated the inertial terms and show that this simplification has negligible effects. The same neural network is trained at each time step without re-initialization, i.e., the trained network parameters for a time step are used as the initial values for the next time step.

4.4 Numerical examples

We apply the PINN to four rigid body - deformable body plane strain geometries under contact. The chosen geometries consider different combinations of outer surface curvatures: (i) flat-flat, (ii) convex-flat, (iii) flat-concave, and (iv) concave-convex to rigorously test the PINN's predictive capabilities. The material parameters and neural network structure (number of layers, number of neurons in each layer, and other base

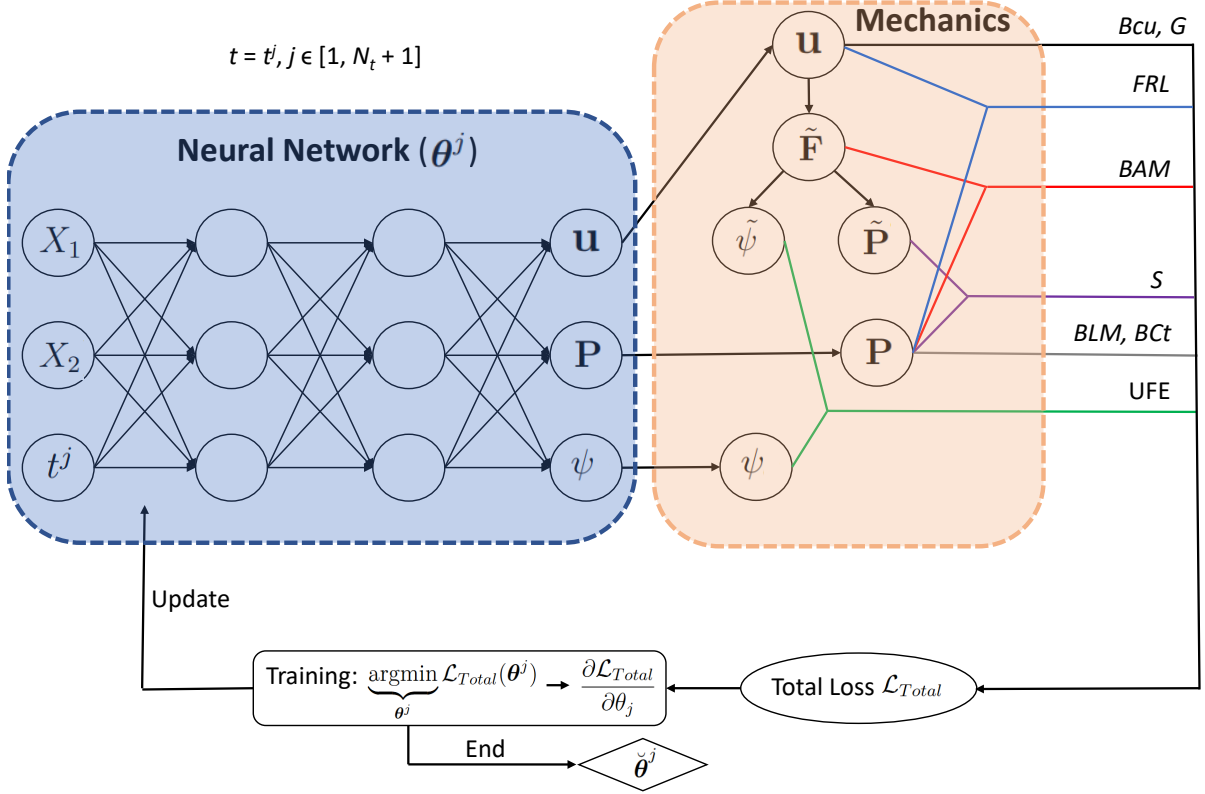


Figure 4.3: **The architecture of the PINN for large deformation contact modeling.** The formulation is based on temporal discretization into N_t time steps. The loss function formulation and training process for a time instant t^j , $j \in [1, N_t + 1]$ is summarized. Mechanics-related information of boundary conditions (BCu , BCt), governing equations (BLM , BAM), constitutive relations (S , UFE), and contact conditions (G , FRL) is encoded in the total loss function $\mathcal{L}_{Total}$. BLM and BAM denote the individual loss functions corresponding to the balance of linear momentum and angular momentum, respectively. BCu and BCt are the individual loss functions corresponding to displacement and traction boundary conditions, respectively. S and UFE are the individual loss functions for stress and undamaged free energy density constitutive relations, respectively. G and FRL are the individual loss functions for the gap function (penetration) and the frictionless contact condition, respectively.

hyperparameter values) from Chapter 3 (Section 3.4) are used. The loss weights α_{BLM} , α_C , α_{BC} , α_{CNT} are set equal to 1. The training is performed with GPU acceleration on the computation cluster provided by the Center for Computation and Visualization (CCV). It is important to note that, because of hyperelasticity, training can begin at any time. This helps reduce the computation time. Hence, we employ the PINN (one-time step) approach discussed in Chapter 3 (Section 3.5).

The finite element software ABAQUS/Explicit is used to obtain the baseline solutions for the PINN validation. The VUMAT from Chapter 3 was used. The number of elements used in the finite element meshes for each deformable body geometry is sufficiently large ($\sim 10,000$) to ensure good accuracy of the baseline FEM solutions. The rigid bodies are modeled as rigid analytical surfaces. Point-wise and global L_2 errors are evaluated for quantitative comparison (refer to the definitions in Chapter 3 [Section 3.4]). The solutions of the PINN after training were evaluated on a 1×1 testing grid consisting of 1000×1000 points to test its extrapolation capabilities. For L_2 error evaluation and contour comparison, nodal data exported from ABAQUS were interpolated on the testing grid.

4.4.1 Flat-flat configuration

First, the simplest possible problem (flat contacting surfaces) is considered to troubleshoot any issues. A 1×1 elastomer plate is compressed using rigid, flat platens (see Figure 4.4(A)) and is free to expand laterally. The top platen is displaced down at a speed of 2 while the bottom platen is held fixed. Only the left half of the plate is simulated, exploiting the problem's symmetry. The collocation grid used for PINN and the mesh used for the baseline FEM solution are shown in Figure 4.4(B). $\mathbf{e}_t$ was evaluated using the dy/dx approach (the ideal tangent approach can be applied to this problem). The PINN solution is evaluated directly at $t = 0.1$ s using the one-time step approach with 100000 epochs training. Good agreement between the PINN-predicted displacement contours and the baseline FEM solution is evident in Figure 4.4(C). The L_2 errors for displacement and stress are low, and the PINN solution has converged in the range of training epochs considered, as shown in Figure 4.4(D). The PINN's strong performance on this simplest contact configuration instills confidence in its application to more complex scenarios.

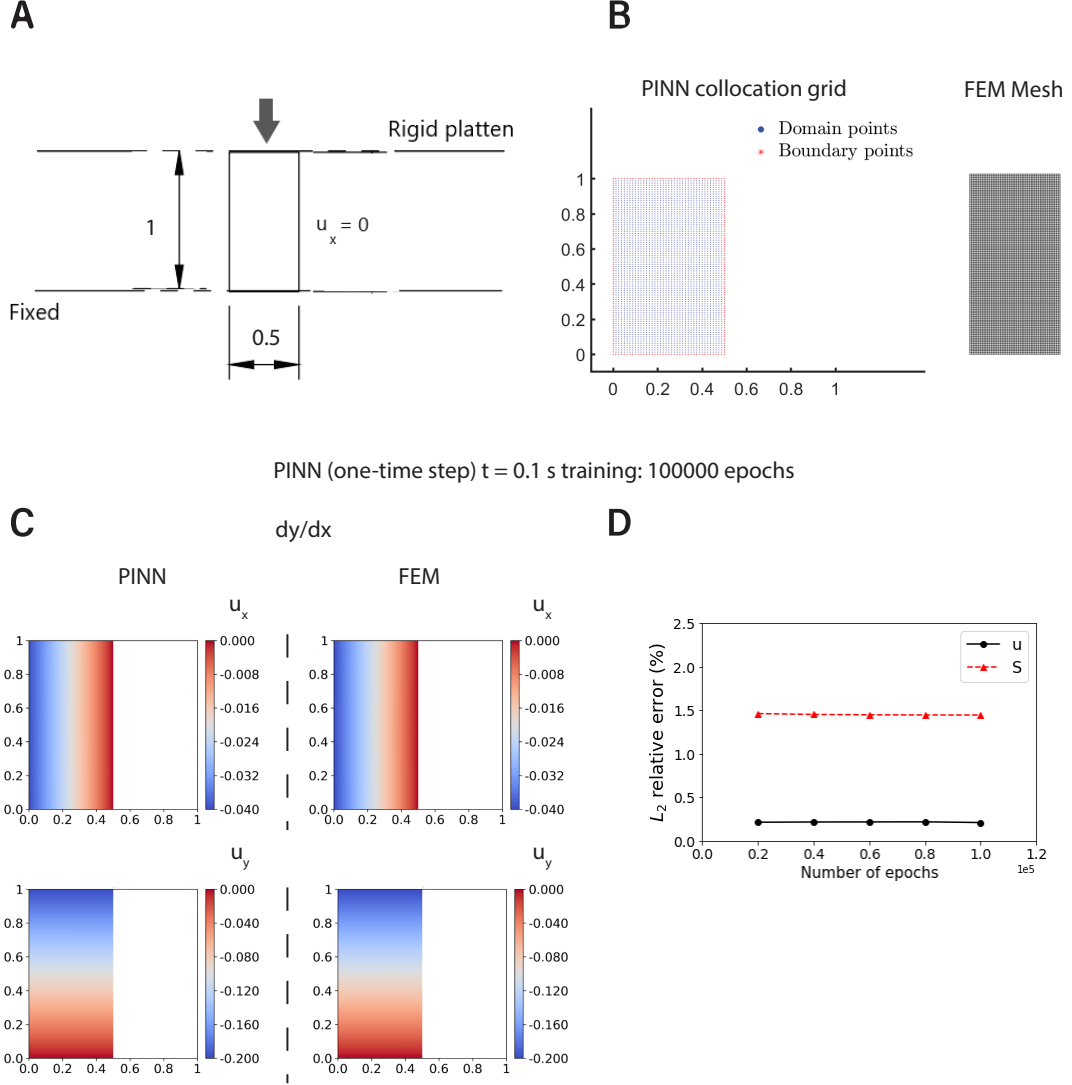


Figure 4.4: **Flat-flat configuration: schematic, comparison of PINN predictions with baseline FEM solution.** (A) Schematic for an elastomer block subject to simple compression using rigid platens under plane strain conditions. The bottom platen is fixed, while the top platen moves downward at a speed of 2. The contacting surfaces are flat, which is the simplest possible configuration and is useful for troubleshooting. (B) The collocation grid and mesh used for PINN and FEM, respectively. (C) Comparison of displacement contour (u_x, u_y) predictions from PINN (one-time step) [solution evaluated at $t = 0.1$ s, 100000 epochs training] with baseline FEM solution showing good agreement. $\mathbf{e}_t$ was evaluated using the dy/dx approach (the ideal tangent approach can be applied to this problem). (D) L_2 relative error evolution during training. The displacement and stress errors are low, and the PINN solution has converged in the range of training epochs considered.

4.4.2 Convex-flat configuration

Next, we consider a 1 x 1 elastomer block indented with a rigid cylinder of radius 0.25 (see Figure 4.5(A)). The indenter is displaced downward at a speed of 2, while the block's bottom surface is fixed. The rigid body contacting surface is convex, whereas the deformable body surface is flat. Figure 4.5(B) shows the collocation grid and mesh used for PINN and FEM, respectively. $\mathbf{e}_t$ was evaluated using all three proposed approaches. The PINN (one-time step) solution is evaluated directly at $t = 0.2$ s with 200000 epochs training. The comparison of the T_{yy} contour predictions from PINN on the deformed shape from the three approaches with the baseline FEM solution is shown in Figure 4.6(A). The dy/dx and Ftang approaches result in good agreement. Because the circular rigid body's surface lacks a globally unique tangent, the ideal tangent approach is unsuitable. Figure 4.6(B) shows the L_2 error evolution during training for the three approaches. The dy/dx and Ftang approaches yield low errors in displacement and stress and converge, whereas the ideal tangent approach does not, as anticipated. The PINN performs well for this contact configuration.

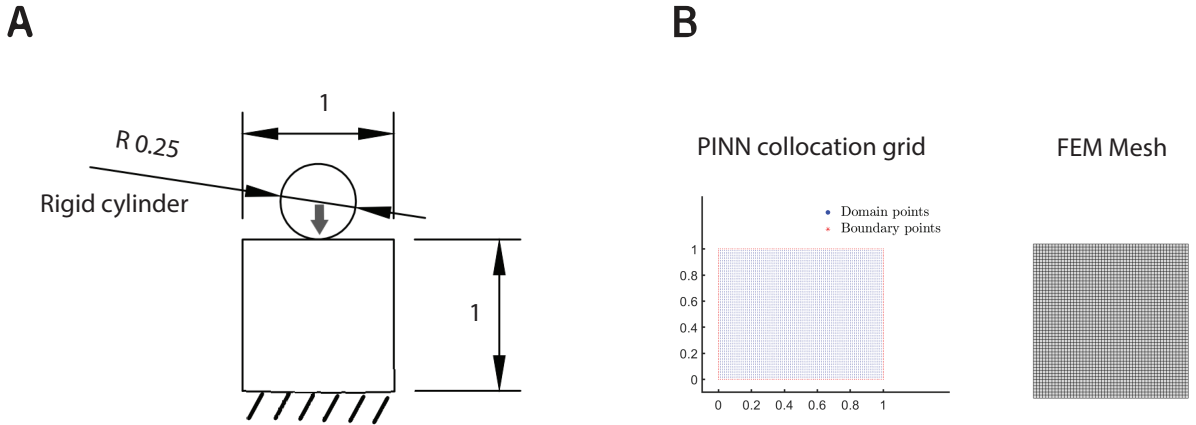
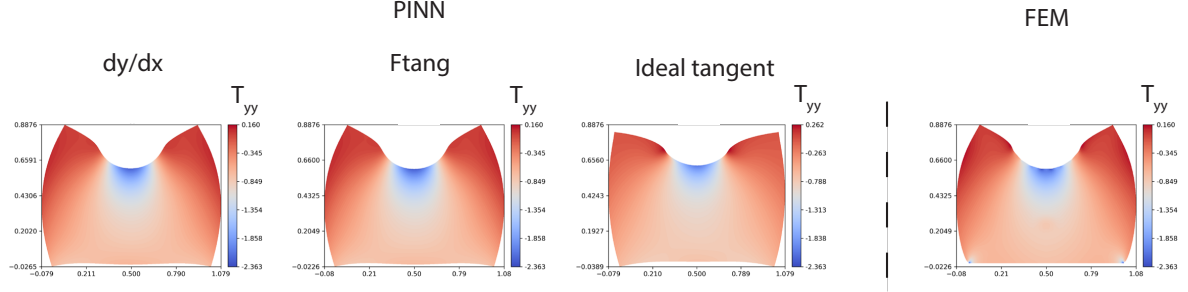


Figure 4.5: **Convex-flat configuration: schematic.** (A) Schematic for an elastomer block indented with a rigid indenter of radius 0.25 under plane strain conditions. The bottom surface of the block is completely fixed, while the indenter is displaced downward at a speed of 2. (B) The collocation grid and mesh used for PINN and FEM, respectively.

PINN (one-time step) $t = 0.2$ s training: 200000 epochs

A



B

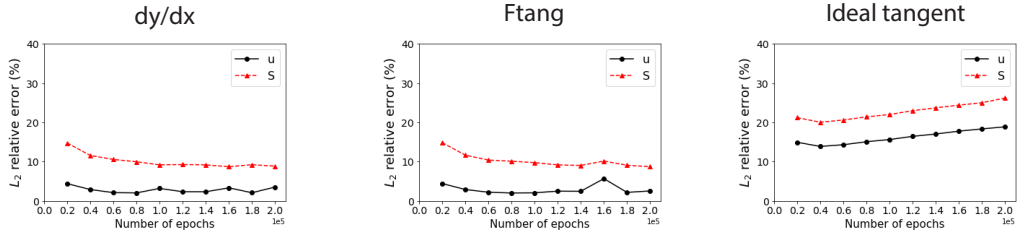


Figure 4.6: **Convex-flat configuration: results.** **(A)** Comparison of T_{yy} contour predictions on the deformed shape from PINN (one-time step) [solution evaluated at $t = 0.2$ s, 200000 epochs training] using the three approaches for $\mathbf{e}_t$ evaluation with the baseline FEM solution. The dy/dx and $Ftang$ approaches result in good agreement. The ideal tangent approach is unsuitable here because the circular rigid body surface lacks a globally unique tangent. **(B)** L_2 relative error evolution during training. The dy/dx and $Ftang$ approaches yield low errors in displacement, stress, and convergence. The ideal tangent approach yields high errors and non-convergence, as anticipated.

4.4.3 Flat-concave configuration

Now we consider an elastomer half cylinder (radius = 0.5) indented with a rigid platten (see Figure 4.7(A)). The platen is displaced downward at a speed of 2, while the bottom surface of the block is constrained in the y -direction. Only the left half is simulated, exploiting the problem's symmetry. The rigid body contacting surface is flat, whereas the deformable body contacting surface is concave. Figure 4.7(B) shows the collocation grid and mesh used for PINN and FEM, respectively. $\mathbf{e}_t$ was evaluated using all three proposed approaches. The PINN (one-time step) solution is evaluated directly at $t = 0.2$ s with

200000 epochs training. The comparison of the T_{yy} contour predictions from PINN on the deformed shape from the three approaches with the baseline FEM solution is shown in Figure 4.8(A). The Ftang approach results in good agreement. The performance of the dy/dx approach for this configuration is unsatisfactory. The rigid body here has a globally unique tangent; however, the ideal tangent approach is unsatisfactory (the solution blows up after 140000 epochs). Figure 4.8(B) shows the L_2 error evolution during training for the three approaches. The Ftang approach yields low displacement and stress errors along with convergence, whereas the dy/dx and ideal tangent approaches do not. The PINN with the Ftang approach performs well for this contact configuration.

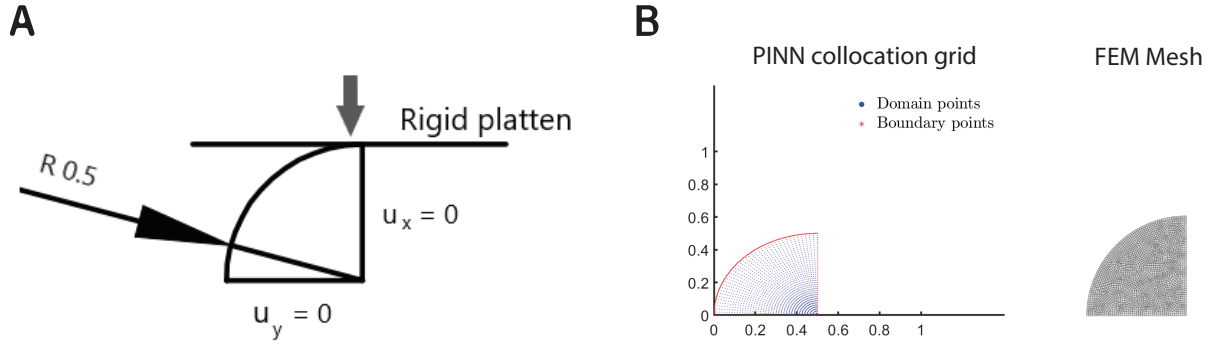


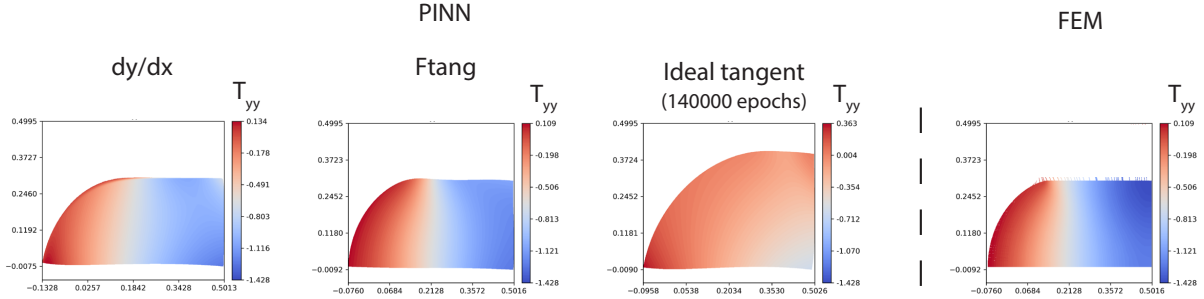
Figure 4.7: **Flat-concave configuration: schematic.** (A) Schematic for an elastomer half cylinder (radius = 0.5) indented with a rigid platen under plane strain conditions. The bottom surface of the half cylinder is constrained in the y direction, while the platen is displaced down at a speed of 2. (B) The collocation grid and mesh used for PINN and FEM, respectively.

4.4.4 Concave-convex configuration

Finally, we consider an elastomer half cylinder (radius = 0.5) being compressed against a rigid half cylinder (radius = 0.5) under plane strain conditions (see Figure 4.9(A)). The rigid half cylinder is completely fixed, while the top surface of the elastomer half cylinder is displaced downward at a speed of 2. Only the left half is simulated, exploiting the problem's symmetry. The rigid body contacting surface is concave, whereas the deformable body contacting surface is convex. Figure 4.9(B) shows the collocation grid and mesh used for PINN and FEM, respectively. $\mathbf{e}_t$ was evaluated using the Ftang approach. The

PINN (one-time step) $t = 0.2$ s training: 200000 epochs

A



B

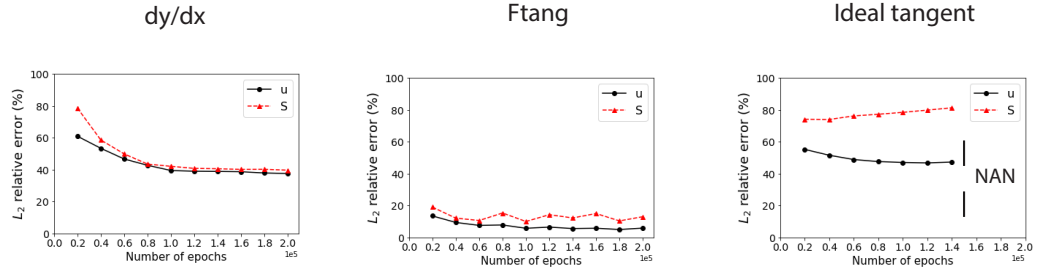


Figure 4.8: **Flat-concave configuration: results.** (A) Comparison of T_{yy} contour predictions on the deformed shape from PINN (one-time step) [solution evaluated at $t = 0.2$ s, 200000 epochs training] using the three approaches for $\mathbf{e}_t$ evaluation with the baseline FEM solution. The Ftang approach results in good agreement. The dy/dx approach performance is not up to par for this configuration. The ideal tangent approach's performance is unsatisfactory (the solution blows up after 140000 epochs), even though the rigid body surface has a unique global tangent. (B) L_2 relative error evolution during training. The Ftang approach yields low displacement and stress errors along with convergence, whereas the dy/dx and ideal tangent approaches do not. NAN - not a number.

PINN (one-time step) solution is evaluated directly at $t = 0.1$ s with 200000 epochs training. The comparison of the T_{yy} contour prediction from PINN on the deformed shape with the baseline FEM solution is shown in Figure 4.9(C). The Ftang approach results in good agreement at lower epochs; however, the solution quality degrades with increasing epochs. Figure 4.9(D) shows the L_2 error evolution during training. The evolution is non-monotonic with a minimum with low displacement and stress errors at 40000 epochs. Along the lines of mixed formulation, the approach of having the tangential

vector components as NN outputs was also tried, but no significant improvements were observed.

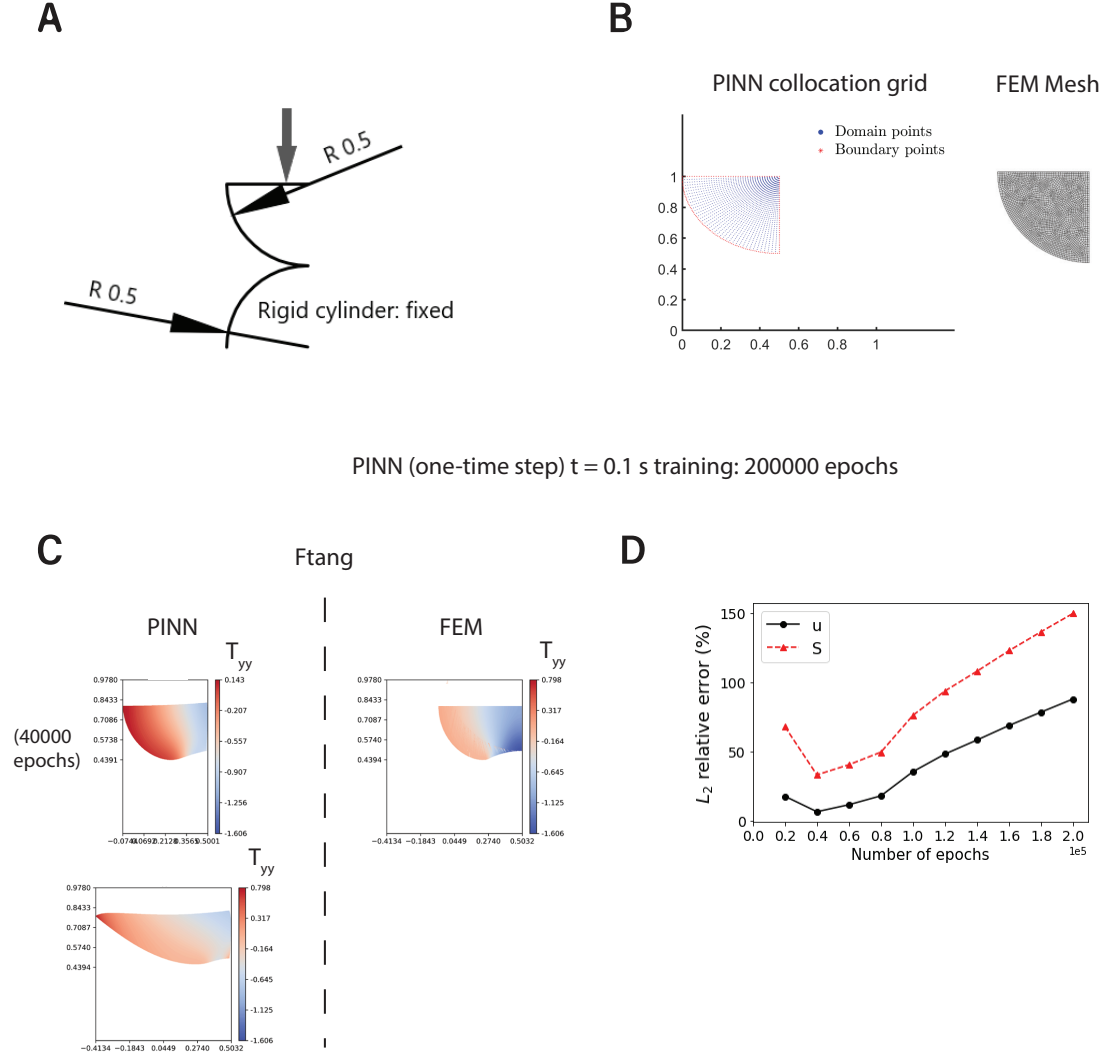


Figure 4.9: Concave-convex configuration: schematic, comparison of PINN predictions with baseline FEM solution. (A) Schematic for an elastomer half cylinder (radius = 0.5) being compressed against a rigid half cylinder (radius = 0.5) under plane strain conditions. The top surface of the elastomer cylinder is displaced down at a speed of 2, while the rigid cylinder is held fixed. The rigid and deformable body contact surfaces are concave and convex, respectively. (B) The collocation grid and mesh used for PINN and FEM, respectively. (C) Comparison of T_{yy} contour prediction on the deformed shape from PINN (one-time step) [solution evaluated at $t = 0.1$ s, 200000 epochs training] using the Ftang approach for $\mathbf{e}_t$ evaluation with the baseline FEM solution. The Ftang approach yields good agreement at lower epochs; however, the solution quality degrades with increasing epochs. (D) L_2 relative error evolution during training. The evolution is non-monotonic with a minimum with low displacement and stress errors at 40000 epochs.

4.5 A preliminary extension for deformable-deformable body contact

Python geometric packages like Shapely will be extremely useful for efficient minimization of penetration during deformable-deformable body contact. However, as discussed earlier, as these do not support Autograd yet, we utilized the more tedious master-slave concept from FEM with point-wise penetration minimization. After assigning master and slave surfaces, regions of interest are first identified as shown in Figure 4.10.

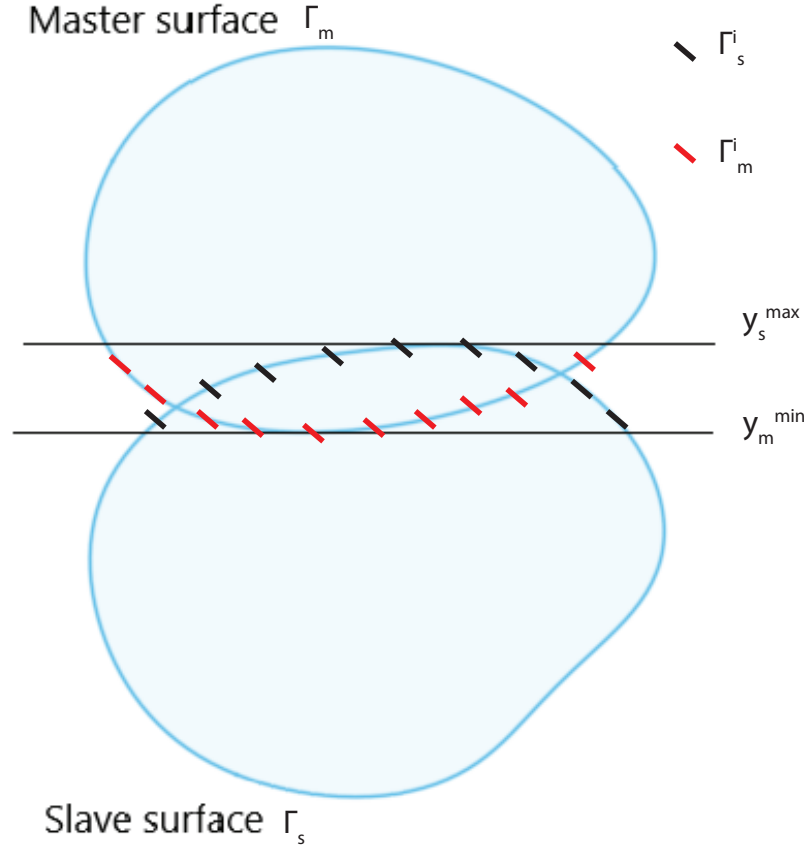


Figure 4.10: **Deformable-deformable body contact: regions of interest.** A possible instant during the solution procedure is shown where a deformable body's outer surface [assigned as slave, Γ_s] penetrates another deformable body's outer surface [assigned as master, Γ_m]. Regions of interest on Γ_s, Γ_m can be identified through first evaluating $y_s^{max}, y_s \in \Gamma_s, y_m^{min}, y_m \in \Gamma_m$ and then $\Gamma_s^i : y_m^{min} < y_s, \Gamma_m^i : y_m < y_s^{max}$.

The subsequent algorithm can be summarized as:

- Divide Γ_m, Γ_s into $\Gamma_m^i, \Gamma_m^{trf}$ and $\Gamma_s^i, \Gamma_s^{trf}$ using the conditions in Figure 4.10 and $\Gamma^{trf} = \Gamma - \Gamma^i$ [trf - traction free]. Also, get $\Gamma_m^{i,ud}, \Gamma_s^{i,ud}$ [ud - undeformed], and szmi, szsi [sz - size of array]. Calculate the normal traction ($\mathbf{t}_s^{i,n}$) on Γ_s^i using $\Gamma_s^{i,ud}$ and the Ftang approach to enforce normal traction continuity (Newton's third law) later.

- For i in range(szsi):

Find neighboring master points for each slave point in Γ_s^i

$$\text{xdiff} = \Gamma_m^i[:,0] - \Gamma_s^i[i,0]$$

$$\text{xdiffneg} = \text{torch.where}((\text{xdiff} < 0))[0]$$

$$\text{xdiffpos} = \text{torch.where}((\text{xdiff} > 0))[0]$$

$$\text{min} = \Gamma_m^i[\text{xdiffneg}]$$

$$\text{mip} = \Gamma_m^i[\text{xdiffpos}]$$

$$\text{minud} = \Gamma_m^{i,ud}[\text{xdiffneg}]$$

$$\text{mipud} = \Gamma_m^{i,ud}[\text{xdiffpos}]$$

$$\text{minudl} = \text{minud}[-1,:]$$

$$\text{mipudf} = \text{mipud}[0,:]$$

Calculate penetration

$$\text{xnm} = \text{min}[-1,0]$$

$$\text{ynm} = \text{min}[-1,1]$$

$$\text{xpm} = \text{mip}[0,0]$$

$$\text{ypm} = \text{mip}[0,1]$$

Interpolated y coordinate of the master surface at the x location of the slave point

$$\text{yintrm} = \text{ynm} + \text{torch.div}((\text{ypm} - \text{ynm}), (\text{xpm} - \text{xnm})) * (\Gamma_s^i[i,0] - \text{xnm})$$

Penetration

$$\text{ydiff}[i] = \Gamma_s^i[i,1] - \text{yintrm}$$

Calculate normal traction ($\mathbf{t}_m^{i,n,l}, \mathbf{t}_m^{i,n,p}$) on neighboring master points using minudl, minpudf, and Ftang approach

Calculate normal traction discontinuity; the plus sign takes into account Newton's third law

$$\mathbf{t}_{diff}^n = \mathbf{t}_s^{i,n} + \frac{(\mathbf{t}_m^{i,n,l} + \mathbf{t}_m^{i,n,p})}{2}$$

Cases of only left or right neighboring master points are handled with the same process

- Not all points in the region of interest are penetrating. We need to separate non-penetrating points to enforce traction-free boundary conditions. For the slave surface:

$$\text{signydiff} = \text{torch.sign}(\text{ydiff})$$

$$\text{sipen} = \text{torch.where}(\text{signydiff} == 1)[0]$$

$$\text{sitrif} = \text{torch.where}(\text{signydiff} != 1)[0]$$

Enforce $\text{ydiff} = 0$, $\mathbf{t}_{diff}^n = \mathbf{0}$, frictionless contact condition (Ftang) for sipen and add sitrf to Γ_s^{trf}

For master surface: assemble neighboring master points for each slave point in sipen to find penetrating points (mipen) in Γ_m^i , enforce frictionless contact condition (Ftang) on mipen, subtract from Γ_m^i to get points (mitrf) to be added to Γ_m^{trf}

Enforce traction-free boundary conditions

- Instead of ydiff, projection height (gap) for penetration quantification was also considered.

4.5.1 Numerical examples

The details from Section 4.4 for neural network parameters, implementation, and results analysis are followed. We considered the convex-flat and concave-convex configurations in this section. Although the PINN predictions using the Ftang approach captured the qualitative behavior (e.g., displacement continuity in the contact region, rigid-deformable body contact-like results for a significant difference in moduli of the bodies) to some extent, the quantitative performance was not satisfactory. Hence, the results are not presented.

4.6 Conclusion

Elastomers, a class of soft polymers, play a critical role in various industries, including automotive tires, industrial seals, footwear, and modern wearable technology. Defined by their high elasticity, these materials frequently experience large deformation contact with rigid surfaces during their applications, an example being sealing. Feasible experiments for material response characterization of very soft materials often involve contact through indentation with a rigid surface. While the conventional finite element method (FEM) has been extensively used for modeling these interactions, physics-informed neural networks (PINNs) have recently gained traction as an alternative numerical method for mechanics problems. PINNs solve governing partial differential equations by embedding physical laws directly into the loss function without a need for training data.

The application of PINNs for contact problems is relatively new and sparse. Although they primarily focus on the small deformation regime, an energy-based PINN for large deformation contact has been formulated using a relatively complex surface contact potential. The underutilization of the mixed PINN formulation, automatic differentiation (AD), and Python geometric packages creates an avenue for relatively simple and efficient PINNs for modeling large deformation contact.

A loss function that also minimizes the intersection area/volume of geometries during the solution procedure can result in a simple PINN-based approach. A crucial role here can be played by the geometric packages in Python (e.g., Shapely). As Autograd is not currently supported for these packages, we used the gap function approach utilized in FEM. A large deformation PINN with a mixed formulation is developed for frictionless contact between rigid and elastomeric deformable bodies. The tangential vector on the deformed body's surface is required to enforce the frictionless condition. Different approaches (dy/dx , F_{tang} , ideal tangent) using AD are presented for its evaluation. Four distinct contact surface configurations (surface curvatures of the rigid and deformable body outer surfaces) were considered for PINN application, enabling rigorous testing of the predictive capabilities. The baseline solutions were obtained using FEM. The dy/dx and F_{tang} approaches appear to perform well only for the flat-flat, convex-flat configurations and flat-flat, convex-flat, flat-concave configurations, respectively. The performance of the F_{tang} approach for the concave-convex configuration is non-monotonic with respect to the number of training epochs, with an optimal point yielding decent results. The simplistic, approximate ideal-tangent approach did not perform well for the only configuration for which it was tested (flat-concave), contrary to expectations. Nevertheless, the simplicity of the proposed PINN for large deformation contact modeling and the demonstrated use of AD for easier, more efficient calculations are promising and should extend to a variety of materials.

A preliminary extension of the formulation to frictionless contact between elastomeric deformable bodies is also presented. Pointwise penetration is minimized through the master-slave concept used in FEM, which can be cumbersome. The results from the preliminary extension are not promising, but the process is documented to aid researchers on this path.

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CHAPTER 5

Polyborosiloxane- polydimethylsiloxane double network polymers and carbon nanotube-filled composites: experimental response and simplified model-based insights

5.1 Abstract

Soft polymers are key engineering materials with broad applications and relatively low elastic modulus in the range of a few kilopascals to tens of megapascals. Conventional polymers with permanent crosslinks exhibit high elasticity and rate-independent response. Significantly rate-dependent behavior and autonomous self-healing can be achieved through dynamic, reversible crosslinks, although with a lack of shape stability. An efficient combination of the contrasting crosslinking mechanisms' desirable properties can be enabled through interpenetrating double network polymers (IDNs). Polyborosiloxane (PBS) is an

important rate-stiffening soft polymer with dynamic crosslinks. PBS-polydimethylsiloxane (PDMS) IDN's significantly rate-dependent, self-healing, and shape retention properties make it a promising matrix candidate for flexible sensing, impact mitigation, and actuation applications. However, the large strain mechanical and electrical response of PBS:PDMS IDNs and filled composites has not been systematically characterized over a range of loading conditions and compositions. This data is crucial for scientific understanding, model development, and practical applications. To address these critical gaps, we have first extensively characterized the rate-dependent mechanical response of select PBS:PDMS (Sylgard 184) IDN compositions (80:20, 60:40, and 20:80 w/w) through large strain tension and compression, stress relaxation, cyclic loading, and creep experiments. The stress-strain responses of the IDNs are significantly nonlinear with respect to composition, strain, and strain rate. The 80:20 composition is shown to be ideal for practical applications in which the properties of PBS are desired without its deficiency in shape stability. A simplified weighted Voigt-Reuss (Hirsch) model captured the PBS:Sylgard 184 IDN response over the compositions and loadings considered with reasonable accuracy. The IDNs were found to be closer to the Reuss idealization, and the model, along with qualitative mechano and photochemical luminescence experiments, can explain the characteristic properties of the 80:20 IDN. The simplified model-based insights should apply to other microstructurally similar IDNs. The 80:20 IDN was used as a rate-dependent matrix for strain and strain-rate sensing, with carbon nanotubes (CNTs) as conductive fillers. The large deformation, rate-dependent mechanical and electrical response was characterized over a range of CNT compositions. Dynamic electrical measurements show that lower strain rates result in higher gauge factors. A simple numerical study suggested that this can be attributed to the corresponding decrease in the stiffness of the rate-stiffening IDN matrix and easier filler separation.

5.2 Introduction

Soft polymers are important engineering materials with chain networks formed through permanent or dynamic chemical or mechanical crosslinks, characterized by a relatively low elastic modulus in the range of a few kilopascals to tens of megapascals. Their applications are diverse, ranging from biomedical (Cianchetti et al., 2018) to structural (Liu et al., 2021). The conventional permanently crosslinked polymers exhibit high elasticity and rate-independent response (Nunes, 2011). Polymers with dynamic, reversible crosslinks are being increasingly explored due to their significantly rate-dependent behavior (Konale et al., 2023) and autonomous self-healing capabilities, enabling reusability (Cheng et al., 2019; Xu et al., 2023). However, they lack shape stability for practical applications (Li et al., 2025a, 2018a). The desirable properties of the contrasting crosslinking mechanisms can be efficiently combined through the approach of interpenetrating double network polymers (IDNs) (Li et al., 2025b; Venkata et al., 2021; Cash et al., 2018).

Polyborosiloxane (PBS) is a key rate-stiffening soft polymer with dynamic boron (B)-oxygen (O) coordinate-bond crosslinks (Zhao et al., 2020a; Kurkin et al., 2021) and broad applications including self-healing sensing and actuation (Narumi et al., 2019) and flexible impact mitigation (Kim et al., 2022; Tu et al., 2023). The large strain, nonlinear, significantly rate-dependent, loading-unloading response of PBS was characterized in (Konale et al., 2023). PBS flows under its own weight over long timescales (hours); that is, it lacks shape stability as is typical for dynamically crosslinked polymers (see Figure 1.1(A)). Open-cell polymeric foams have been used as a supporting structure for PBS (Fan et al., 2022; Sang et al., 2021). The IDN strategy offers several advantages, including one-pot synthesis and uniform dispersion and interpenetration of the networks (Tang et al., 2019a). A few studies have used polydimethylsiloxane (PDMS) to synthesize PBS-based IDNs (Tang et al., 2019a, 2020; Qu et al., 2021; Yeasmin et al., 2023).

Figure 5.1 shows the excellent shape stability of the 80:20 weight/weight (w/w) PBS:PDMS IDN compared to PBS over a timescale of 24 hours.

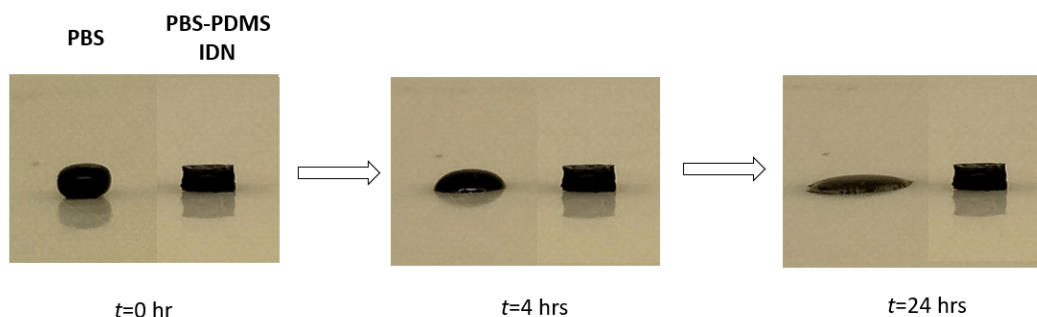


Figure 5.1: **Enhanced shape stability of PBS:PDMS IDN.** Shape stability comparison of PBS and PBS:PDMS (Sylgard 184) IDN (80:20 w/w). The specimens for both materials were initially approximately cylindrical, with diameters and heights of 5 mm and 3 mm, respectively. Snapshots of the specimens under their own weight at $t=0$, 4, and 24 hours are shown. PBS flows like a viscous fluid over these timescales, while PBS:PDMS IDN retains its shape. The specimens were colored for visualization.

PBS:PDMS IDN's significantly rate-dependent, self-healing, and shape retention properties make it a suitable matrix candidate for flexible strain and strain rate sensing (Tang et al., 2019b; Qu et al., 2021; Tang et al., 2020; Wu et al., 2019). These sensors conduct electricity via contacts between conductive particles and via electron tunneling between separated particles. Sensing is achieved by measuring changes in conductivity resulting from mechanical deformation. However, these studies have focused on practical applications, and a comprehensive material response characterization is lacking. In addition to flexible sensing, there is broad applicability of PBS:PDMS and similar IDNs in several other important areas, including engineered tissues (Talebian et al., 2019), flexible lightweight impact mitigators (Zhao et al., 2021a, 2020b; Jiang et al., 2014), and actuators (magnetorheological materials) (Gang et al., 2019; Zhao et al., 2021b; Wang et al., 2014).

Despite this wide range of applications, the large strain mechanical and electrical response of PBS:PDMS IDNs and filled composites has not been systematically characterized over a range of loading conditions and compositions. This data is crucial for scientific understanding of the underlying mechanisms, model development, and practical

applications. To address these critical gaps, the following contributions are presented in this chapter:

(i) Extensive mechanical response characterization of three PBS:PDMS (Sylgard 184) IDN compositions (80:20, 60:40, 20:80 w/w) over three decades of strain rate (0.001 s^{-1} - 1 s^{-1}) through large strain tension and compression, stress relaxation, cyclic loading, and creep experiments. Comparisons of the PBS:PDMS IDN data with that of pure PBS and PDMS are also presented.

(ii) The 80:20 PBS:PDMS IDN exhibits improved shape stability and recovery upon unloading without a significant change in stress-strain curves with respect to pure PBS. A similar observation is also made for another PDMS variant (Ecoflex 00-30). Mechano and photochemical luminescence qualitative experiments highlight the possible underlying mechanisms. The variation in the mechanical response of the IDN with composition is highly nonlinear. Hence, a wide range of mechanical properties can be achieved through careful design of IDNs.

(iii) A basic model based on the weighted Voigt-Reuss (Hirsch) approach is shown to reasonably capture the PBS:Sylgard 184 IDN response over the compositions and loadings considered. The IDNs are closer to the Reuss idealization, and the model can explain the peculiar properties of the 80:20 IDN. The simplified model-based insights should extend to other microstructurally similar IDNs.

(iv) As a model system for flexible strain and strain rate sensing applications, 80:20 PBS:Sylgard 184 IDN filled with carbon nanotubes (CNT) as conductive filler particles is considered. The high aspect ratio of CNTs enables electrical percolation at relatively low filler contents (Jung et al., 2019). The large deformation, rate-dependent (0.01 - 0.1 s^{-1}), stress-strain, and electrical response of 80:20 PBS:Sylgard 184 IDN with different CNT filler compositions is characterized. The mechanical and electrical responses are highly sensitive to the CNT composition, strain, and strain rate. The gauge factor varies

inversely with the strain rate and CNT composition.

5.3 Experiments

All experiments were conducted at 25°C. Three repetitions were performed for each experimental result reported.

5.3.1 Synthesis

PBS

The procedure outlined in Chapter 1 (Section 1.2.1) was followed.

PDMS

Sylgard 184

Sylgard 184 (Dow Inc.) Part A and Part B were mixed in a 10:1 weight ratio, and the stirred mixture was vacuum degassed, followed by heating at 60°C for 5 hours.

Ecoflex 00-30

Ecoflex 00-30 (Smooth-On Inc.) Part A and Part B were mixed in a 1:1 weight ratio, and the stirred mixture was degassed using a vacuum pump, followed by curing at room temperature for 4 hours.

PBS:PDMS IDN

PBS:PDMS IDN was synthesized by mixing PBS and PDMS (Sylgard 184 or Ecoflex 00-30) precursors, in the desired w/w ratio, stirring at room temperature, followed by heating at 120°C for 72 hours. The condensation of hydroxy end groups on boric acid and hydroxy-terminated PDMS melt for PBS (crucial for dynamic crosslinking), and hydrosilylation reaction between the silicon (Si)–hydrogen (H) groups and vinyl groups

(permanent crosslinking) in PDMS occurs simultaneously during the synthesis procedure to form an IDN. Similar backbone structures enable uniform dispersion and penetration of the networks (Tang et al., 2019b). It can be noted that the densities of PBS, Sylgard 184, and Ecoflex 00-30 are close to 1 g/cc; hence, w/w is approximately equivalent to v/v. The IDN compositions are specified as ratios hereon.

80:20 PBS:Sylgard 184 IDN filled with CNT

The PBS:PDMS CNT composite was synthesized through a one-pot process. PBS and Sylgard 184 precursors were mixed in an 80:20 w/w ratio. The desired amount (wt% of total PBS and Sylgard 184 weight) of multi-walled CNT (10-20 nm, Cheap Tubes) was added to the polymer precursor mixture, followed by stirring at room temperature, and heating at 120°C for 72 hours. Higher CNT concentrations resulted in more viscous precursor mixtures; therefore, appropriate mixing methods should be employed to ensure uniform dispersion. The composite compositions are specified hereon as 80:20 PBS:Sylgard 184 z CNT, where z is the wt% of CNT relative to the total weight of the polymer precursor mixture.

5.3.2 Mechanical response characterization

The mechanical testing experiments were carried out using the UniVert Mechanical Test System (CellScale). The custom 3D printed setup discussed for tensile testing in Chapter 2 (Section 2.2) was used. The tension (cuboidal) and compression (cylindrical) specimens had dimensions of 20 ± 2 mm (length) x 6 ± 1 mm (width) x 3.175 ± 1 mm [gauge dimensions], and 4 ± 1 mm x 3.175 ± 1 mm (height), respectively. For the compression tests, the platten-specimen surfaces were lubricated to prevent barreling. The small strain rheology experiments were performed on an ARES G2 Rheometer to 1% strain using a cone and plate geometry (40 mm diameter and 0.04 rad cone angle).

5.3.3 Electrical response characterization

The cuboidal specimens of 80:20 PBS:Sylgard 184 IDN z CNT had gauge dimensions of 30 ± 2 mm x 30 ± 2 mm (width) x 3.175 ± 1 mm. To measure simultaneous changes in resistance during deformation, two metallic strips were secured in the designed slots of custom 3D-printed stretching fixtures. This ensured good electrical contact with the specimen's top and bottom (along the length) surfaces, which are constrained by the fixtures and do not deform. Alligator clips attached to the strips were connected to a Keysight 34465A Digital multimeter. For static measurements, the metallic strips placed flush with the top and bottom specimen surfaces were slightly compressed by a clamp to ensure electrical contact. Resistance measurements were taken every 30 minutes over 2 hours, and the average values are reported.

5.4 Results - PBS:PDMS IDN

5.4.1 Stress-strain curves

The tensile and compressive engineering stress-strain curves for 80:20 and 60:40 PBS:Sylgard 184 IDNs over three decades of strain rate from 0.001 to 1 s^{-1} are shown in Figures 5.2(A) and (B), respectively. Both compositions exhibit rate-dependent response, with the degree of rate dependence decreasing with decreasing PBS content. This is consistent with the significantly rate-dependent and approximately rate-independent behaviors of PBS (Konale et al., 2023) and Sylgard 184 (see Figure 5.2(C)), respectively. The significant tension-compression asymmetry inherent to PBS (Konale et al., 2023; Konale and Srivastava, 2025) and Sylgard 184 (<https://www.sandia.gov/polymer-properties/sylgard-184-youngs-modulus-at-23c/>, 2026) is also seen for the 80:20 and 60:40 compositions in Figures 5.2(A), (B).

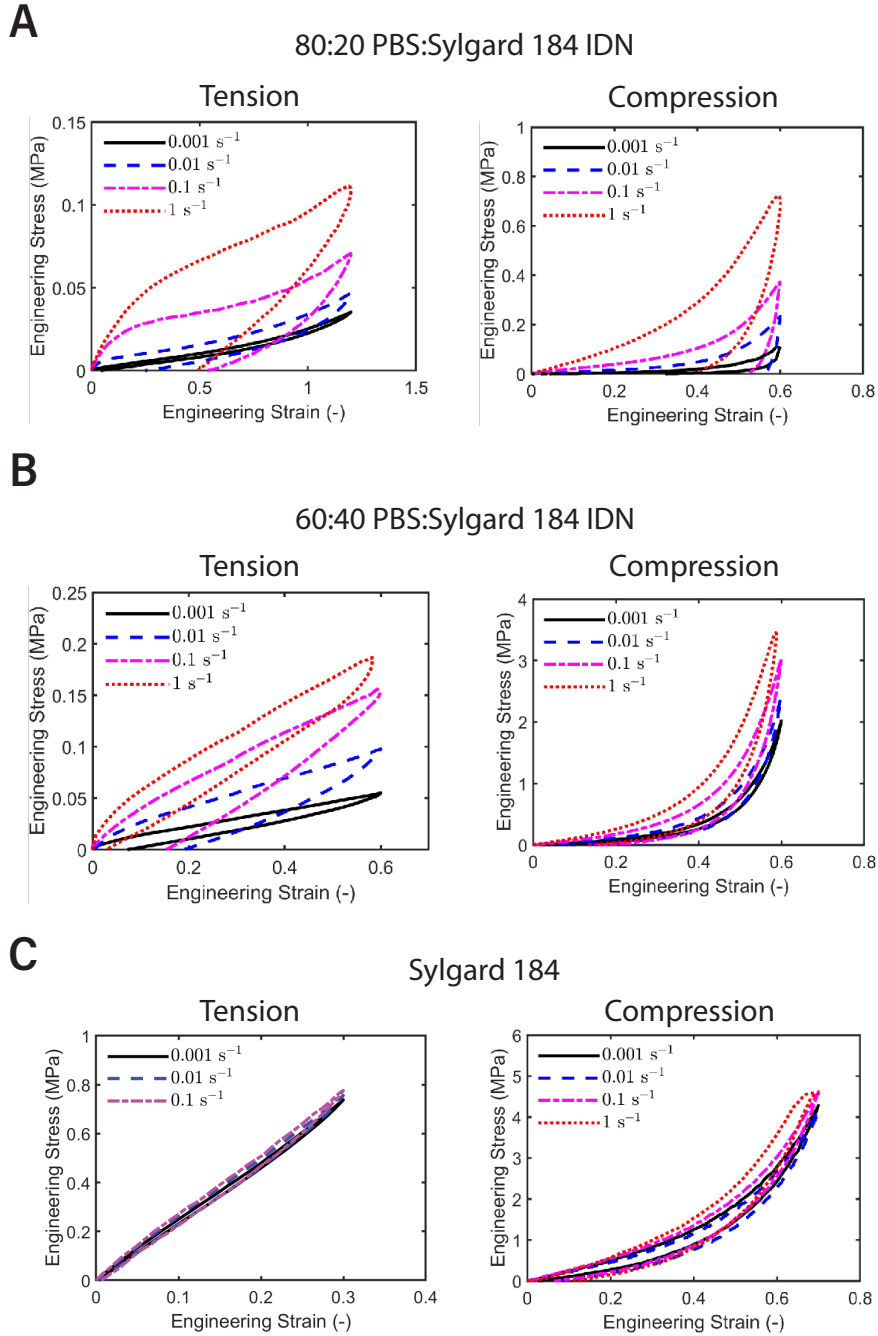


Figure 5.2: Tensile and compressive stress-strain curves for 80:20, 60:40 PBS:Sylgard 184 IDNs and Sylgard 184 over a range of strain rates. Stress-strain curves under tension and compression for **(A)** 80:20 and **(B)** 60:40 PBS:Sylgard 184 IDNs over three decades of strain rate (0.001 - 1 s^{-1}). Both compositions exhibit rate-dependent behaviour, with the degree of rate-dependence decreasing with decreasing PBS content. The 80:20 and 60:40 compositions exhibit significant tension-compression asymmetry inherent to PBS and Sylgard 184. **(C)** Engineering stress-strain curves for Sylgard 184 under tension and compression over the 0.001 - 1 s^{-1} strain rate range. The approximately rate-independent response of Sylgard 184 is highlighted. This also explains the reduced rate-dependency of the 60:40 IDN composition.

Comparison of IDN stress-strain response with pure PBS and Sylgard 184

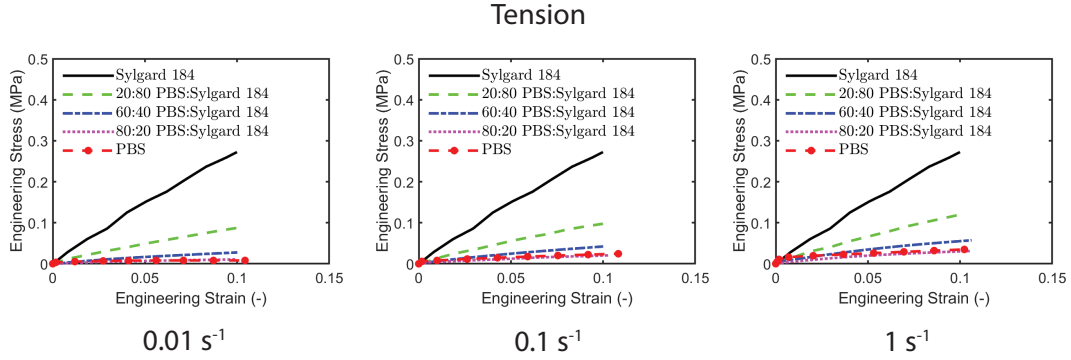
The tensile engineering stress-strain curves of PBS:Sylgard 184 IDNs with three different compositions (80:20, 60:40, 20:80) are compared with that of pure PBS and Sylgard 184 at 0.01, 0.1, and 1 s⁻¹ strain rates in Figure 5.3(A). Sylgard 184 is significantly stiffer than PBS. Additionally, the curves for 80:20 IDN and PBS overlap across all three strain rates, indicating that the addition of 20% Sylgard 184 does not significantly alter the stress-strain curves relative to PBS. This is emphasized in Figure 5.3(B), with comparison of 80:20 IDN and PBS stress-strain curves under tension and compression for the three rates specified earlier. The degree of similarity increases with higher strain rates. The significant deviations at lower strain rates can be attributed to a small rate-independent contribution from Sylgard 184 in the IDN.

5.4.2 Stress relaxation

Tensile and compressive stress-relaxation curves (60% engineering strain) for 80:20 and 60:40 PBS:Sylgard 184 IDNs for the 0.001-1 s⁻¹ are shown in Figures 5.4(A) and (B), respectively. Since PBS relaxes completely over the timescales considered here (Konale et al., 2023), the relaxed stress level, which is approximately constant with respect to strain rate for both compositions, can be attributed to the stress carried by the approximately rate-independent Sylgard 184 network. It can be noted that the Sylgard 184 network stress contribution in the 80:20 IDN is relatively small, consistent with the observation in Section 5.4.1.

Figure 5.5(A) compares the relaxed stress levels in tension and compression from the Voigt model and experiments. Deviations from the experimental results are more pronounced in tension than in compression, as seen for the 60:40 composition. However, for the 80:20 composition, the deviations are significant in both tension and compression, with the experimental values being much smaller than the Voigt model predictions. The tensile Voigt model predictions are consistently higher than the experimental values.

A



B

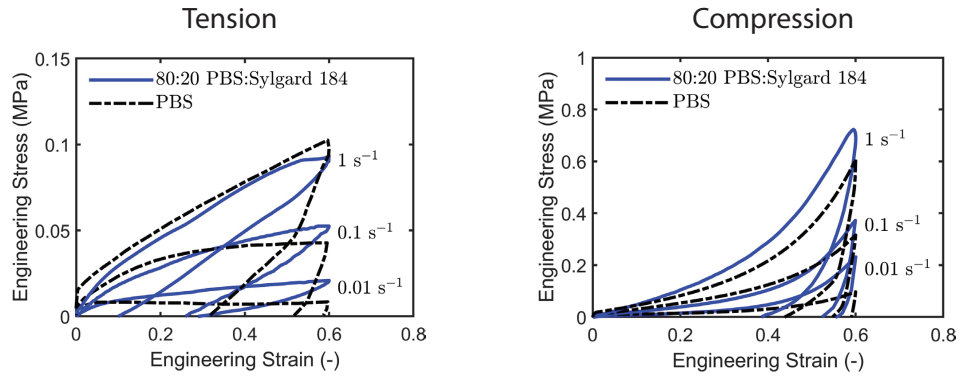


Figure 5.3: Comparison of IDN stress-strain curves with pure PBS and Sylgard 184: range of compositions and strain rates. (A) Tensile engineering stress-strain curves for PBS, 80:20 IDN, 60:40 IDN, 20:80 IDN, Sylgard 184 at 0.01, 0.1, and 1 s⁻¹ strain rates. The curves for 80:20 IDN and PBS overlap, indicating no significant change upon addition of 20% Sylgard 184. **(B)** Tensile and compressive engineering stress-strain curves of 80:20 IDN and PBS for the three strain rates specified earlier. The degree of similarity between the 80:20 IDN and PBS curves increases with increasing strain rate. A relatively small, rate-independent contribution from Sylgard 184 in the IDN led to significant deviations at lower strain rates.

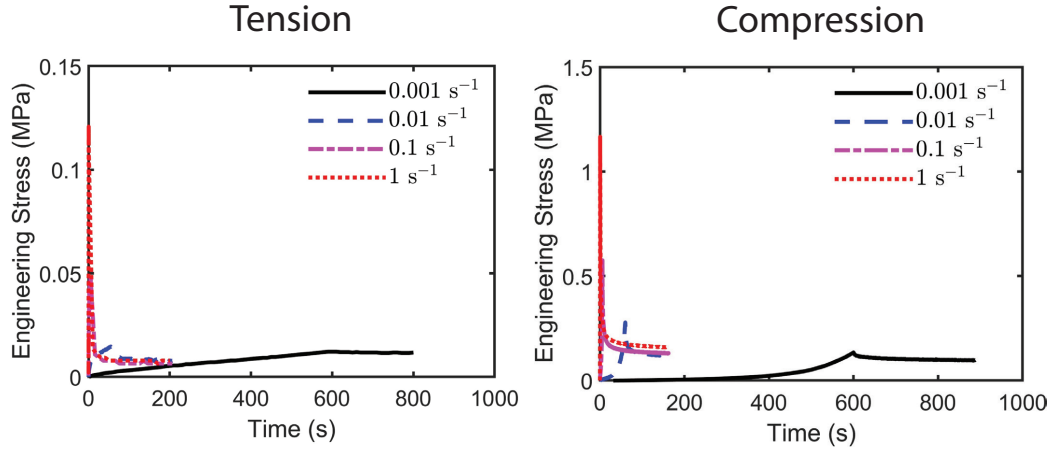
Similar observations are also made for the 80:20 PBS:Ecoflex 00-30 IDN as seen in Figure 5.5(B).

5.4.3 Cyclic tensile loading

The 80:20 and 60:40 PBS:Sylgard 184 IDNs were subjected to cyclic tensile loading until 60% engineering strain at 0.01 s⁻¹ strain rate, with a 10-minute rest period between cycles. The good repeatability over three cycles for both compositions is shown in Figure 5.6. It can be noted that the specimens recovered plastic strain during the rest time.

A

80:20 PBS-Sylgard 184 IDN
60% engineering strain

**B**

60:40 PBS:Sylgard 184 IDN
60% engineering strain

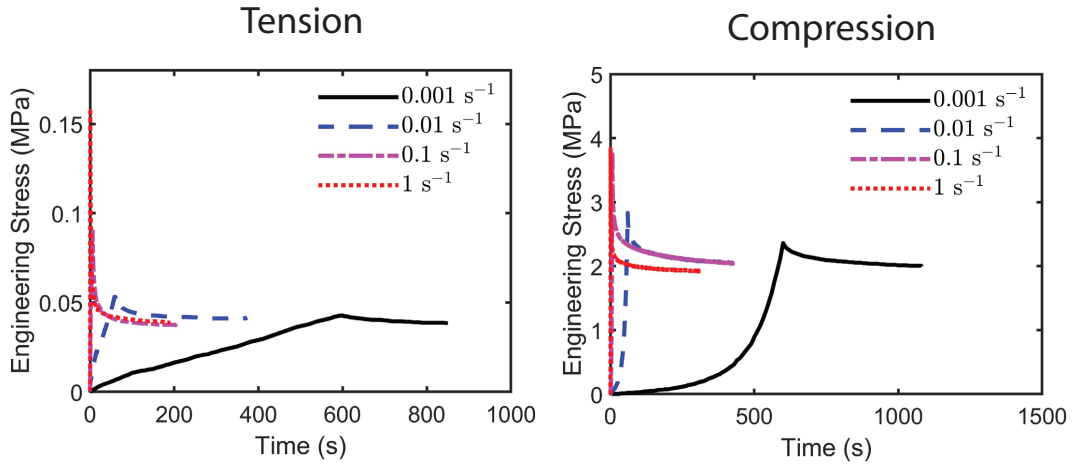


Figure 5.4: Tensile and compressive stress relaxation curves for 80:20 and 60:40 PBS:Sylgard 184 IDNs over a range of strain rates. Engineering stress relaxation curves under tension and compression for (A) 80:20 and (B) 60:40 PBS:Sylgard 184 IDNs over three decades of strain rate (0.001 - 1 s^{-1}) and subjected to 60% engineering strain. The approximately constant relaxed stress levels for both compositions can be attributed to the stress carried by the approximately rate-independent Sylgard 184 network. PBS completely relaxes over these timescales. The relatively small Sylgard 184 network stress contribution for the 80:20 IDN is consistent with the observation in Section 5.4.1.

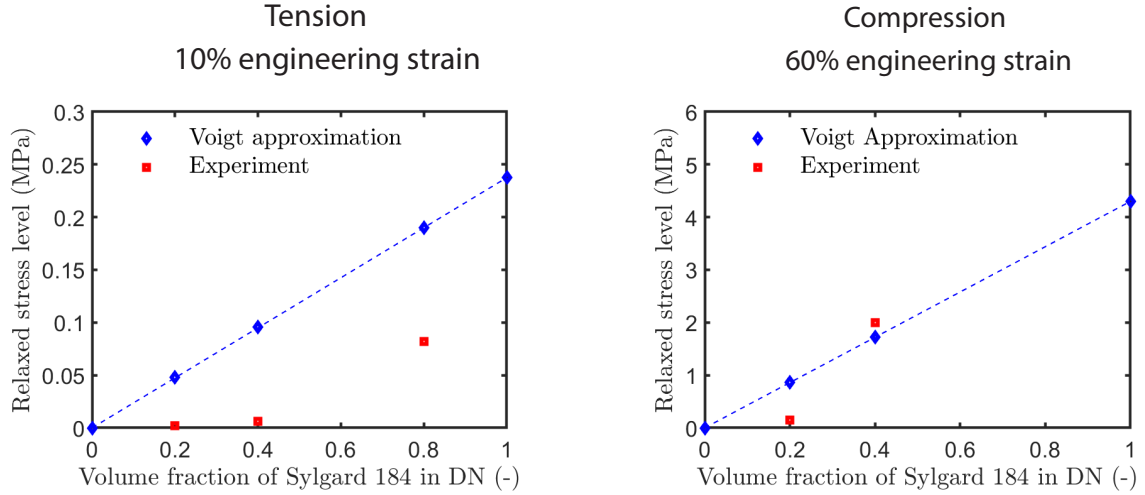
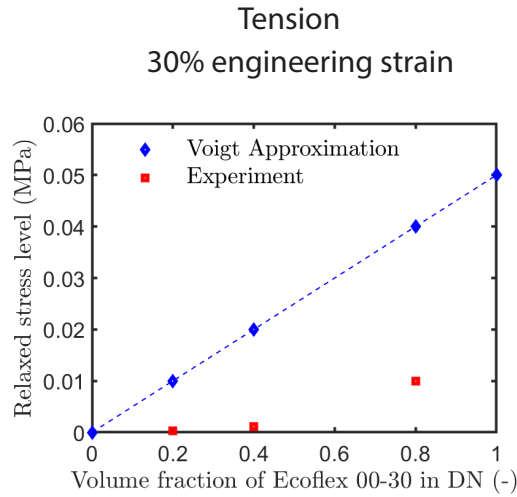
A**PBS-Sylgard 184 IDN****B****PBS-Ecoflex 00-30 IDN**

Figure 5.5: **Comparison of Voigt model predictions for relaxed stress levels under tension and compression with experimental results.** (A) Relaxed stress level predictions under tension (10% engineering strain) and compression (60% engineering strain) from the Voigt model compared with experimental results. Deviations of the Voigt model predictions from the experiments are more pronounced in tension than in compression, as observed for the 60:40 IDN. For the 80:20 IDN, the deviations are significant in both tension and compression, with the experimental values being much smaller than the Voigt model predictions. (B) Similar observations for the 80:20 PBS:Ecoflex 00-30 IDN.

PBS could not recover the plastic strain during the rest period and, therefore, could not be subjected to cyclic loading.

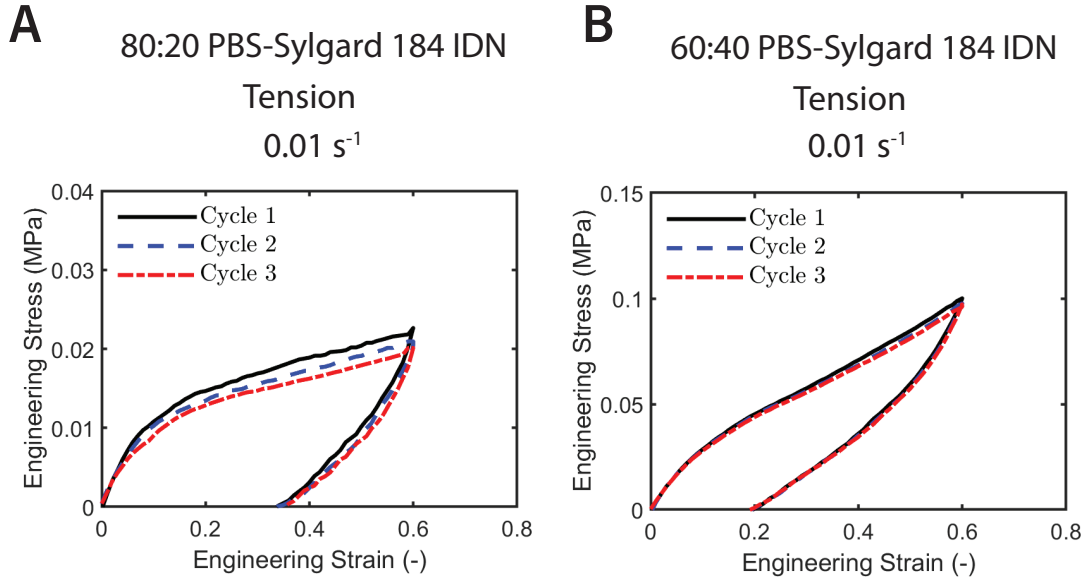


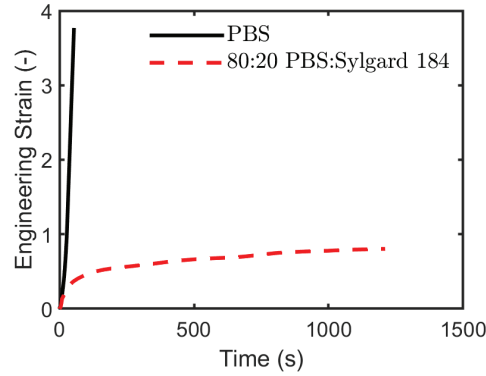
Figure 5.6: **Cyclic tensile loading of 80:20 and 60:40 PBS:Sylgard 184 IDNs.** Engineering stress-strain curves from cyclic tensile loading of (A) 80:20 and (B) 60:40 IDNs till 60% engineering strain and at 0.01 s^{-1} strain rate. The rest time between the three cycles was 10 minutes. Both compositions exhibited good repeatability across the three cycles, and the specimens recovered plastic strain during the rest period. PBS cannot be subjected to cyclic loading as it cannot recover the plastic strain during the rest period.

5.4.4 Creep

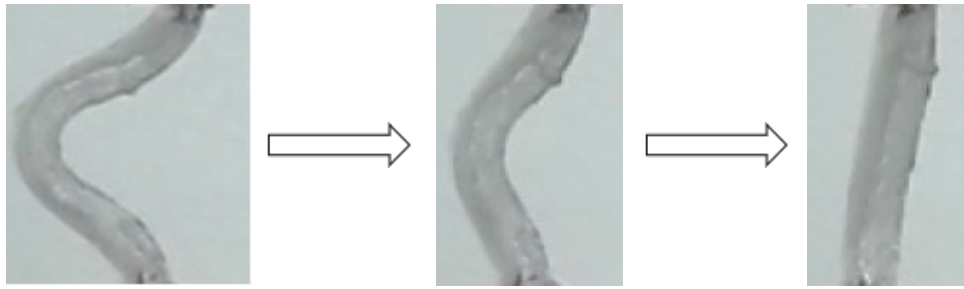
The 80:20 PBS:Sylgard 184 IDN and PBS were subjected to creep at a fixed engineering stress of 0.01 MPa, achieved in 10 s. The engineering strain vs. time curves in Figure 5.7(A) show that the 80:20 IDN has significantly higher creep resistance than PBS. A similar observation for a different IDN system was reported in (Li et al., 2018b). The fixture was moved back to the initial specimen length at the end of the creep tests. Figure 5.7(B) shows the plastic strain recovery of the 80:20 IDN at different stages. PBS showed no plastic strain recovery.

A

Creep: Fixed engineering stress of 0.01 MPa
achieved in 10 s

**B**

80:20 PBS-Sylgard 184 IDN



Fixture brought to initial specimen
length after the creep test end

Figure 5.7: **Enhanced creep resistance of 80:20 PBS:Sylgard 184 IDN compared to PBS.** (A) The engineering strain vs. time curves for 80:20 IDN and PBS subjected to creep with the fixed engineering stress of 0.01 MPa achieved in 10 s. The creep resistance of the 80:20 IDN is significantly higher than that of PBS. (B) Plastic strain recovery of the 80:20 IDN upon bringing the fixture back to the initial specimen length. PBS showed no plastic strain recovery.

5.4.5 Fracture resistance W_{cr} of the 80:20 IDN

The fracture resistance of the 80:20 IDN, W_{cr} , was evaluated using the experimental setup and details described in Chapter 2 (Sections 2.2 and 2.3). Figure 5.8 shows the force-displacement curve for the ASTM Type V dogbone specimen stretched uniaxially at an approximate strain rate of 4 s^{-1} . W_{cr} evaluated using this curve is equal to 0.63 MPa, which is very close to the value for PBS (0.6 MPa). This aligns with the stress-strain

response of the 80:20 IDN being approximately similar to that of PBS, especially at the higher strain rates in Section 5.4.1.

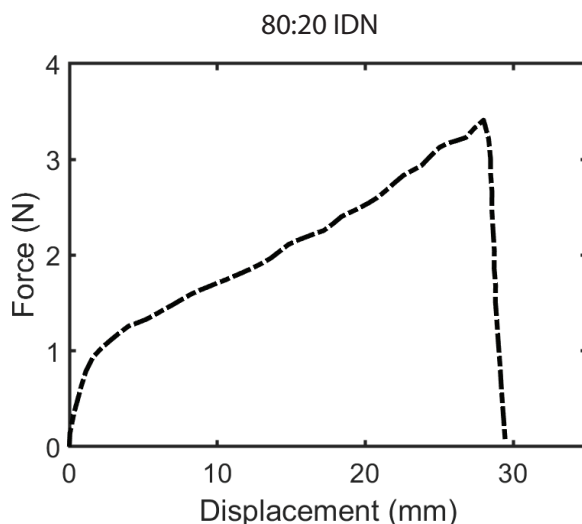


Figure 5.8: W_{cr} of the 80:20 IDN: force-displacement curve. The experimental setup and details in Chapter 2 (Sections 2.2 and 2.3) were used to evaluate W_{cr} of the 80:20 IDN. Force vs. displacement curve from uniaxial tension test (4 s^{-1}) of ASTM Type V dogbone specimen. W_{cr} of the 80:20 IDN (0.63 MPa) is very close to that of PBS (0.63 MPa), in line with their similar stress-strain responses.

5.4.6 Mechano and photochemical luminescence to visualize the Sylgard 184 network in the 80:20 IDN

Spiropyran (SP) has been used in the literature as a mechanochromic probe for visualizing the deformation of Sylgard 184 (Brown et al., 2017; Zhang et al., 2014). SP covalently attaches only to Sylgard 184 chains as crosslinks. Upon exposure to UV light or mechanical force, SP undergoes a reversible reaction that generates visible purple light. In a polymer with Sylgard 184 and SP, mechanoluminescence will be observed only if the Sylgard 184 network carries significant stress. SP (CAS No.: 1554286-40-6, Ambeed Inc.) was added in a small amount to Sylgard 184 (Sylgard 184 curing agent w/SP w = 1/15) and 80:20 IDN (Sylgard 184 curing agent w/SP w = 1/60). Figures 5.9(A) and (B) show the effect of UV light exposure and mechanical forces (compression in a vise grip) on Sylgard 184 and 80:20 IDN cylindrical specimens with SP, respectively. Photochemical

luminescence upon UV exposure is observed in both materials, confirming the presence of Sylgard 184 constituents. However, no significant mechanoluminescence is seen for the 80:20 IDN. This suggests that the Sylgard 184 network is in the initial stages of percolation or is well connected but carries substantially lower stresses, even though the corresponding monomer and crosslinker were added in a stoichiometric ratio.

5.4.7 Frequency sweep response of the 80:20 and 60:40 IDNs and comparison with PBS

The comparison of the small strain (1% strain) frequency sweep response of the 80:20 and 60:40 IDN with that of PBS (Konale et al., 2023) is shown in Figure 5.10. The rate-independent response of Sylgard 184 implies its storage (G') and loss moduli (G'') will be approximately constant with respect to frequency. PBS's G' and G'' , on the other hand, vary significantly with frequency due to its rate-stiffening behavior. The 80:20 IDN's G' and G'' curves have characteristics similar to those of PBS. This aligns with the results in Section 5.4.1, highlighting that the 80:20 IDN exhibits properties similar to those of PBS. G' , G'' for the 60:40 IDN have reduced frequency dependence, inline with the observation in Section 5.4.1.

5.5 Basic modeling

5.5.1 Comparison of 80:20 IDN stress-strain response with Voigt model predictions

A simple model for composites, particularly IDNs, is the Voigt (equal strain) approach, where the total response is modeled as the sum of the pure constituent response scaled by the corresponding volume fractions (Zhao, 2012; Lavoie et al., 2019; Mao et al., 2017). Figure 5.11 compares the stress-strain curves from the Voigt model (addition of pure PBS and pure Sylgard 184 responses scaled by the corresponding volume fractions) and

A

Sylgard 184 with SP



After UV exposure

After compression
in a vise gripSubsequent exposure
to white light (10 mins)**B**

80:20 IDN with SP



After UV exposure

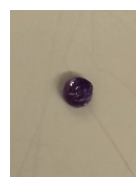
After compression
in a vise gripSubsequent exposure
to UV light

Figure 5.9: **Sylgard 184 network visualization through mechano and photochemical luminescence.** SP, which specifically crosslinks Sylgard 184 chains, was used. SP emits visible, purple light upon exposure to UV light or application of mechanical forces. The effect of UV light exposure and mechanical forces (compression in a vise grip) on **(A)** Sylgard 184 and **(B)** 80:20 IDN cylindrical specimens with SP. The presence of Sylgard 184 constituents is confirmed by photoluminescence in both materials. The absence of mechanoluminescence in the 80:20 IDN suggests that the Sylgard 184 network is in the initial stages of percolation or is experiencing significantly lower stresses.

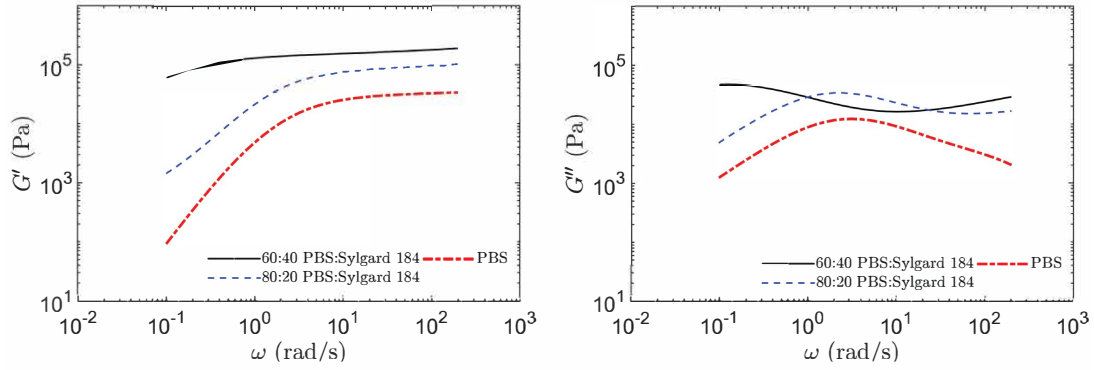


Figure 5.10: **Frequency sweep response: Comparison of 80:20 and 60:40 IDNs with PBS.** G' and G'' curves for the 80:20 and 60:40 IDNs and PBS. PBS and Sylgard 184 exhibit significantly rate-dependent and approximately rate-independent responses, resulting in G' and G'' varying significantly and insignificantly with frequency, respectively. The 80:20 IDN's storage and loss modulus curves have characteristics similar to PBS, as anticipated through observations in Section 5.4.1. 60:40 IDN's G' , G'' show reduced frequency dependency consistent with the observation in Section 5.4.1.

experiments for the 80:20 IDN under tension. The experimental values are significantly smaller than the predictions of the Voigt model. Possible explanations can be that the Sylgard 184 network in the 80:20 IDN is in the initial stages of percolation or experiences significantly lower stress as suggested in Section 5.4.6.

5.5.2 Voigt model: corrected PDMS volume fraction

The actual quantity of crosslinked PDMS in the IDN can be accounted for through a corrected PDMS volume fraction in the Voigt model. Specifically, the ratio of the relaxed stress level to the corresponding stress in pure PDMS can be used as a correction factor for the PDMS volume fraction. Figure 5.12 shows the Voigt model predictions for the 80:20 IDN under compression with the corrected volume Sylgard 184 fraction. Reasonable agreement is seen with the experimental results. It should be noted that this approach did not yield acceptable results under tension and under both tension and compression for 60:40 and 20:80 IDNs. Hence, the Voigt idealization is not suitable for modeling the PBS:Sylgard 184 IDN.

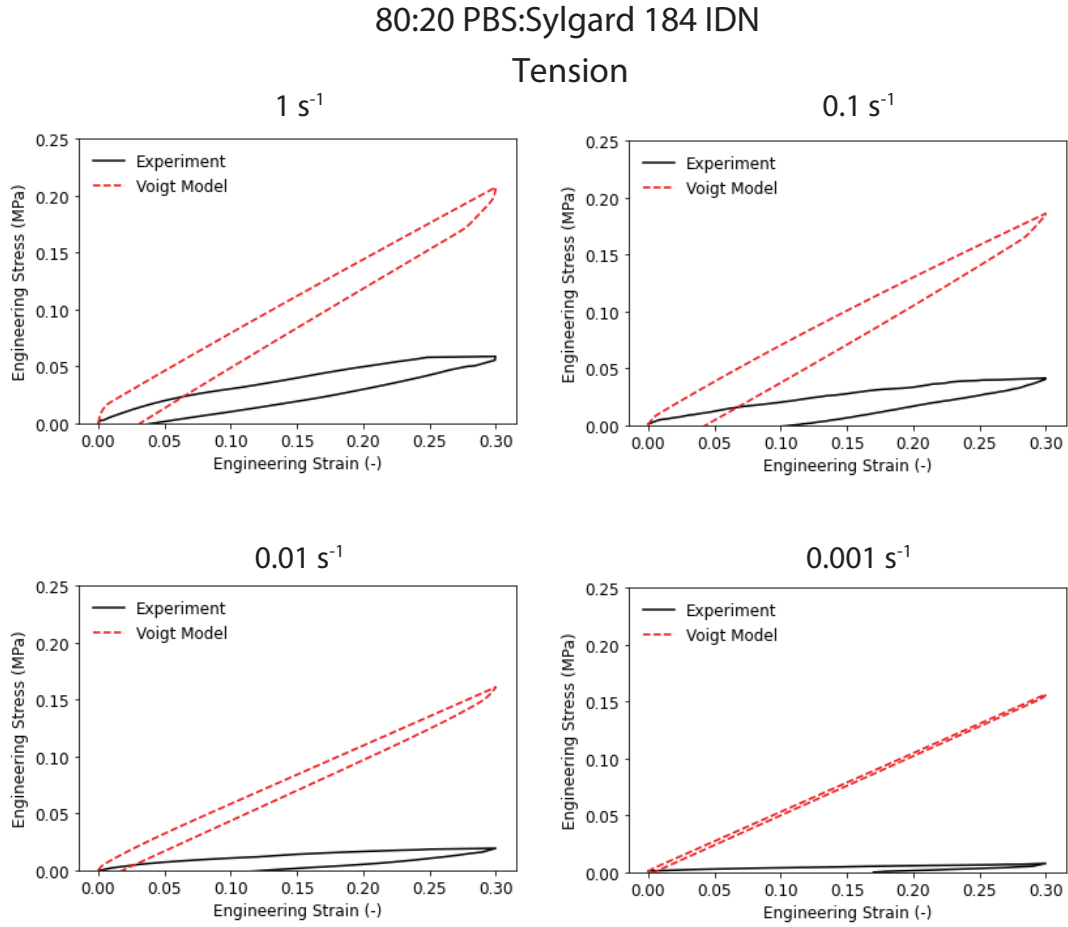


Figure 5.11: **Comparison of IDN stress-strain response with Voigt model predictions.** Comparison of the tensile stress-strain curves from the Voigt model (addition of pure PBS and pure Sylgard 184 responses scaled by the corresponding volume fractions) and experiments for the 80:20 IDN. The experimental values are significantly smaller than the Voigt model predictions. This can be attributed to the Sylgard 184 network in the 80:20 IDN being in the initial stages of percolation or carrying substantially lower stresses as suggested in Section 5.4.6.

5.5.3 Hirsch (weighted Voigt-Reuss) model

Several studies in the literature have proposed the equal stress (Reuss) approach (the opposite of the Voigt approach) as a more suitable method for modeling IDNs (Tauber et al., 2021; Hu et al., 2012; von Lockette et al., 2002; Okumura, 2004). The response of several composites is best captured by a combination of Voigt and Reuss idealizations (Luo, 2022; NIKLAS, 1992; Christensen and and, 2015). An example is the Hirsch model (Chihaoui et al., 2020) where the response is taken as a weighted average of the Voigt and

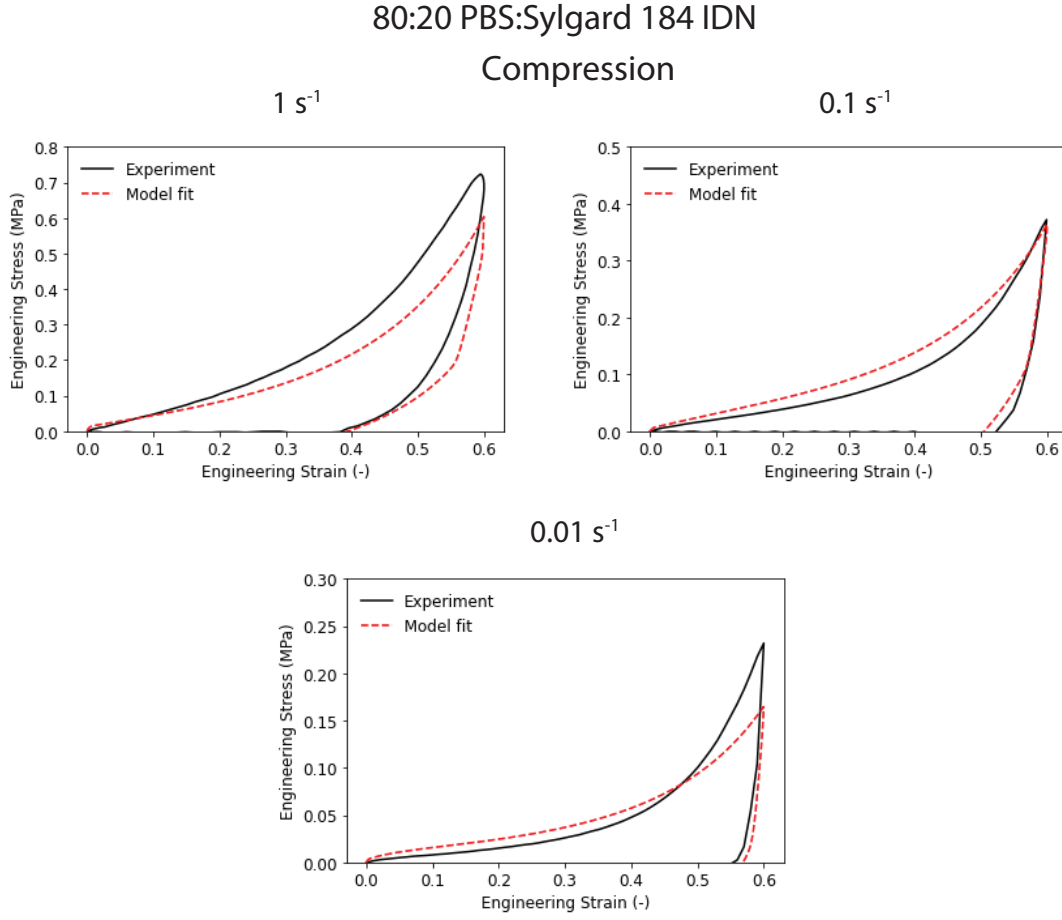


Figure 5.12: **Voigt model stress-strain response predictions: corrected PDMS volume fraction.** The ratio of the relaxed stress level to the corresponding stress in pure PDMS can be interpreted as a corrected PDMS volume fraction. Comparison of Voigt model predictions with corrected Sylgard 184 volume fraction with experimental results for the 80:20 IDN under compression, showing reasonably good agreement. This approach did not yield acceptable results under tension and both tension and compression for 60:40 and 20:80 IDNs. The PBS:Sylgard 184 IDN response can not be modeled using the Voigt idealization.

Reuss predictions, i.e.,

$$\sigma = \chi \sigma_{Voigt} + (1 - \chi) \sigma_{Reuss}. \quad (5.1)$$

χ is an adjustable weight, with values closer to 1 or 0 indicating that the system is closer to the Voigt or Reuss idealization, respectively. An alternative interpretation is that χ represents the stress transfer between the phases. The Hirsch model is applied to the PBS:Sylgard 184 IDNs. Evaluation of the σ_{Voigt} is straightforward as discussed in Section

5.5.1. For the σ_{Reuss} evaluation, the following strategy is adopted. Let $\Delta\varepsilon_{pbs}$, $\Delta\varepsilon_{pdms}$ be the strain increments in PBS and Sylgard 184 networks in the Reuss idealization. The stress increments are calculated approximately by using the local stiffnesses $\left(E(\varepsilon) = \frac{\partial\sigma}{\partial\varepsilon}\right)$ of the pure components at the strain rate of interest from the experimental data (Figure 5.2 and (Konale et al., 2023)). The inherent assumption here is that the imposed global strain rate and the strain rates experienced by the components are of the same order of magnitude. The Reuss (equal stress) idealization requires that

$$\sigma_{total} = \sigma_{pbs} = \sigma_{pdms} \implies \Delta\sigma_{total} = \Delta\sigma_{pbs} = \Delta\sigma_{pdms}. \quad (5.2)$$

For sufficiently small strain increments, the above equation can be rewritten as

$$E_{pbs}\Delta\varepsilon_{pbs} = E_{pdms}\Delta\varepsilon_{pdms}. \quad (5.3)$$

Since a relatively simple model can capture the response of Sylgard 184, it is beneficial to evaluate $\Delta\varepsilon_{pdms}$ for evaluating $\Delta\sigma_{total}$. For a total strain increment of $\Delta\varepsilon_{total}$ and volume fraction ϕ of PDMS, $\Delta\varepsilon_{pdms}$ can be approximately evaluated using $\Delta\varepsilon_{total} = \phi\Delta\varepsilon_{pdms} + (1 - \phi)\Delta\varepsilon_{pbs}$ and (5.3) as

$$\Delta\varepsilon_{pdms} = \frac{\Delta\varepsilon_{total}}{\phi + \frac{E_{pdms}}{E_{pbs}}(1 - \phi)}. \quad (5.4)$$

σ_{total} can be then be calculated as

$$\sigma_{total} = \sum \Delta\sigma_{total} = \sum E_{pdms}\Delta\varepsilon_{pdms}. \quad (5.5)$$

χ values can be obtained by fitting the Hirsch model predictions to the corresponding IDN data. Figures 5.13(A), (B), and (C) show the good Hirsch model fit to the experimental

tensile stress-strain curves for the 80:20, 60:40, and 20:80 IDNs, respectively, over the 0.001-1 s⁻¹ strain rate range, and the corresponding χ values. Two trends can be noted-(i) the χ values are closer to 0, i.e., the IDNs are closer to the Reuss idealization, and (ii) the χ values increase with increasing PDMS content (ϕ). The interactions within the polymer networks are therefore a function of composition and strain rate. A few IDNs closer to the Reuss idealization have been reported in the literature (Yoo et al., 2006).

The model can be validated by comparing its predictions for the relaxed stress level with the experimental results in Section 5.4.2. Relaxed stress predictions can be obtained by noting that the Reuss part relaxes completely. Further, in the Voigt part, only the stress contribution from the Sylgard 184 network is included, since PBS relaxes completely. Reasonably good agreement of the tensile relaxed stress predictions with the experimental results for the 80:20 (30% strain), 60:40 (30% strain), and 20:80 (10% strain) IDNs over the 0.001-1 s⁻¹ strain rate range can be seen in Figures 5.14(A), (B), and (C), respectively, validating the model.

The model can also be applied to the small strain frequency sweep data in Section 5.4.7, specifically for G' . For a shear strain input $\gamma(t) = \gamma_o \sin(\omega t)$, the shear stress $\tau(t)$ is equal to

$$\tau(t) = G' \gamma_o \sin(\omega t) + G'' \gamma_o \cos(\omega t). \quad (5.6)$$

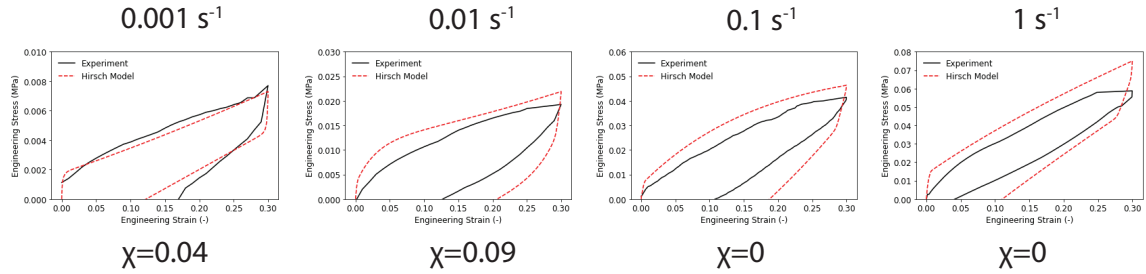
G'_{Voigt} calculation is straightforward, i.e. summation of G'_{Voigt} of each phase scaled by their corresponding volume fraction. G'_{Reuss} for the IDN can be defined following (5.3) as equal to $\gamma_o^{pdms} G'_{pdms} / \gamma_o$, where γ_o^{pdms} can be obtained following (5.4). The simplified expression for G'_{Reuss} is

$$G'_{Reuss} = \frac{G'_{pdms} G'_{pbs}}{(1 - \phi) G'_{pdms} + \phi G'_{pbs}}. \quad (5.7)$$

A

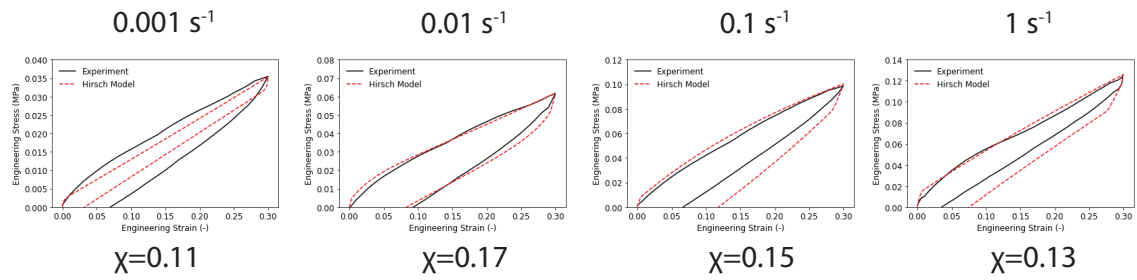
80:20 PBS:Sylgard 184 IDN

Tension

**B**

60:40 PBS:Sylgard 184 IDN

Tension

**C**

20:80 PBS:Sylgard 184 IDN

Tension

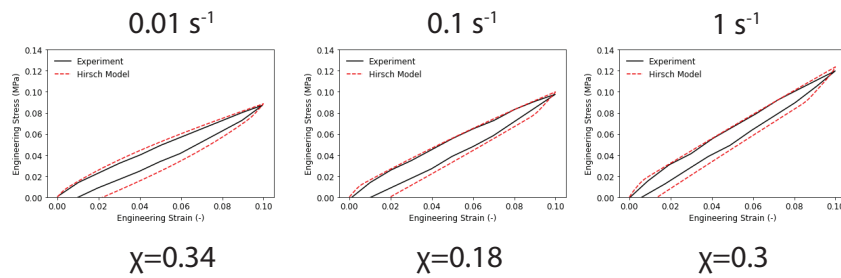


Figure 5.13: **Hirsch (weighted Voigt-Reuss) model fit.** Good fit of the Hirsch model to the experimental tensile stress-strain curves over the 0.001-1 s⁻¹ strain rate range for **(A)** 80:20, **(B)** 60:40 and **(C)** 20:80 IDNs. The corresponding χ values are also shown. The χ values are closer to 0, indicating the IDNs are closer to the Reuss idealization. Further, the χ values increase with increasing PDMS content. Composition and strain rate affect interactions within the polymer networks.

A

80:20 PBS:Sylgard 184 IDN

Tension

30% engineering strain

Relaxed stress (MPa)	Experiment	Hirsch Model
0.001 s ⁻¹	0.003	0.006
0.01 s ⁻¹	0.005	0.014
0.1 s ⁻¹	0.005	0
1 s ⁻¹	0.003	0

B

60:40 PBS:Sylgard 184 IDN

Tension

30% engineering strain

Relaxed stress (MPa)	Experiment	Hirsch Model
0.001 s ⁻¹	0.026	0.034
0.01 s ⁻¹	0.025	0.053
0.1 s ⁻¹	0.025	0.046
1 s ⁻¹	0.025	0.040

C

20:80 PBS:Sylgard 184 IDN

Tension

10% engineering strain

Relaxed stress (MPa)	Experiment	Hirsch Model
0.01 s ⁻¹	0.078	0.070
0.1 s ⁻¹	0.087	0.037

Figure 5.14: **Hirsch (weighted Voigt-Reuss) model validation: stress relaxation.** The tensile relaxed stress predictions from the Hirsch model agree reasonably well with the experimental results over the 0.001-1 s⁻¹ strain rate range for **(A)** 80:20 (30% strain), **(B)** 60:40 (30% strain) and **(C)** 20:80 (10% strain) IDNs, validating the model.

G' for the IDN can then be evaluated following (5.1) as

$$G' = \chi G'_{Voigt} + (1 - \chi) G'_{Reuss}. \quad (5.8)$$

G'_{pdm_s} was approximated using the tensile Young's modulus from the stress-strain curve in Figure 5.2 and assuming ν (Poisson's ratio) = 0.5 along with $G' = G$ (since Sylgard 184 is approximately rate-independent and elastic). Figure 5.15 shows the Hirsch model G' predictions using the average large strain fit χ (values in Figure 5.13 for the 80 : 20 and 60 : 40 IDNs). Good qualitative agreement with the experimental results is evident, further validating the model.

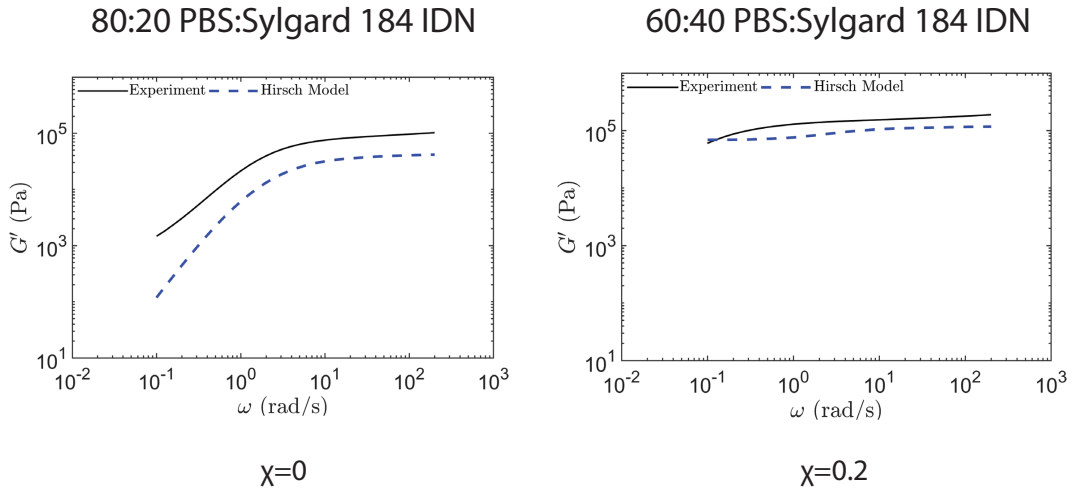


Figure 5.15: **Hirsch (weighted Voigt-Reuss) model validation: small strain frequency sweep.** Good qualitative agreement of the Hirsch model G' predictions with the experimental results for the 80:20 and 60:40 IDNs in Figure 5.10, further validating the model. Average χ values fit to the large strain response in Figure 5.13 were used.

A detailed understanding of the polymer network interactions is needed to develop comprehensive models for PBS:Sylgard 184 IDNs. The results in Section 5.4.1 showed that Sylgard 184 is significantly stiffer than PBS, which exhibits a substantial rate-stiffening behavior. The high degree of similarity between the 80:20 IDN and PBS stress-strain responses at the higher strain rates (0.1 and 1 s^{-1}) in Section 5.4.1 can be attributed to (i) χ being closer to 0 - Reuss idealization. This is equivalent to two springs in series,

with the effective response being closer to the softer spring, i.e., PBS, and (ii) σ_{Voigt} being insignificant compared to σ_{Reuss} in (5.1). The deviations at lower strain rates (0.01 s^{-1}) can be attributed to (i) σ_{Reuss} being small due to the significantly small stiffness of PBS at these rates and (ii) σ_{Reuss} being insignificant compared to σ_{Voigt} . The shape stability in Figure 5.1 and enhanced creep resistance in Figure 5.7 of the 80:20 IDN are a result of the non-zero σ_{Voigt} (non-zero χ). Thus, the basic weighted Voigt-Reuss (Hirsch) model can reasonably capture the PBS:Sylgard 184 IDN's response over the compositions and loadings considered and explain the particular properties of the 80:20 IDN. The simplified model-based insights should extend to other microstructurally similar IDNs.

5.6 Results - PBS:PDMS IDN filled with CNT

5.6.1 Stress-strain curves

The CNT composition ($z=5, 10, 20$ at 0.01 s^{-1}) and rate-dependent ($0.001 - 1 \text{ s}^{-1}$) stress-strain response of 80:20 PBS:Sylgard 184 IDN zz CNT is shown in Figures 5.16(A) and (B), respectively. $z=20$ resulted in a relatively stiffer and more brittle response than the lower CNT compositions. The 80:20 PBS:Sylgard 184 IDN 5 CNT exhibits a significantly rate-dependent response, which is crucial for strain rate sensing.

5.6.2 Electrical response

Static measurements

The electrical response of 80:20 Sylgard 184 IDN z CNT ($z=5, 10, 20$) was measured under static (undeformed) conditions. Specifically, the specimens' resistance was measured. Resistivity, a material property, was evaluated as the product of the specimen's resistance and cross-sectional area, divided by its length. The resistivity decreases sharply with increasing CNT composition (i.e., increasing z values), as shown in Figure 5.17. This variation suggests that $z=5, 10, 20$ CNT compositions lie in the electrical percolation

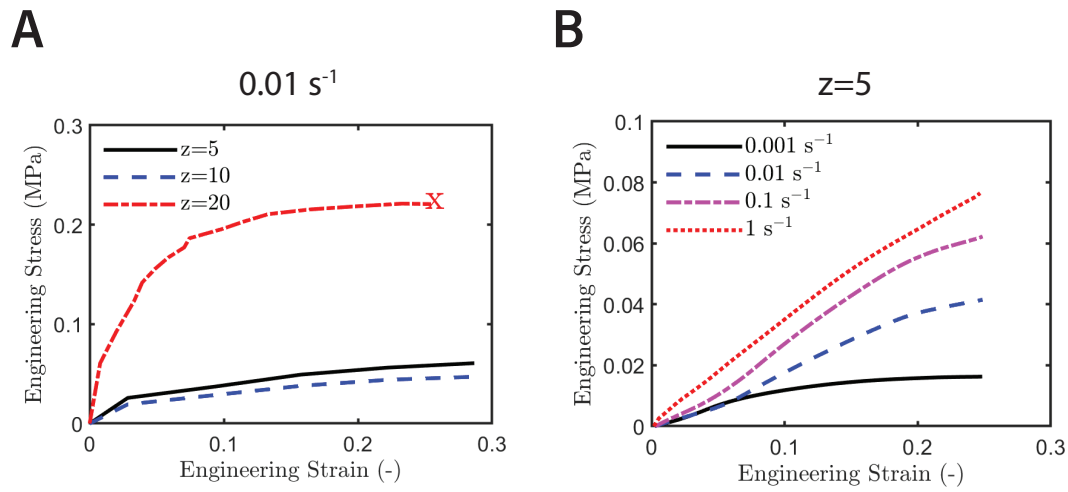


Figure 5.16: **Filler composition and rate-dependent stress-strain response of 80:20 PBS:Sylgard 184 IDN z CNT.** The engineering stress-strain curves for (A) different CNT compositions ($z=5, 10, 20$) at 0.01 s^{-1} strain rate and (B) at different strain rates ($0.001 - 1 \text{ s}^{-1}$) for $z=5$. Higher CNT compositions ($z=20$) yield a relatively stiffer and more brittle response. $z=5$ CNT composition exhibits a significantly rate-dependent response crucial for rate sensing.

regime.

Static measurements

z (wt% of CNT)	Resistance (k Ω)	Resistivity (Ω -mm)
5	300	544,285
10	2.5	4535
20	0.021	38

Figure 5.17: **Variation of static electrical properties of 80:20 PBS:Sylgard 184 IDN z CNT with CNT composition.** Resistance of the specimens and material resistivity under static conditions for $z=5, 10, 20$. Increasing CNT composition (z values) resulted in a pronounced nonlinear decrease in resistivity.

Dynamic measurements

Relative change in resistance ($\frac{\Delta R}{R_o}$) and gauge factor (relative change in resistance divided by engineering strain) measurements were performed during deformation of specimens with different CNT compositions ($z=5, 10, 20$ at 0.01 s^{-1}) and at different strain rates ($0.01 - 1 \text{ s}^{-1}$ for $z=5$). The lower CNT composition ($z=5$) resulted in a higher gauge factor and, hence, greater strain sensitivity, as shown in Figure 5.18(A). For a fixed CNT composition ($z=5$), Figure 5.18(B) shows that the sensitivity to strain decreases with increasing strain rates. It can be noted here that higher sensitivity to strain implies higher separation (along the direction of loading) between fillers upon deformation. Several studies in the literature have shown that the gauge factor can increase as matrix stiffness decreases (Patidar et al., 2025; Narongthong et al., 2020). These works suggest that lower matrix stiffness can lead to higher filler mobility and relatively easier separation upon deformation. Since the PBS:PDMS IDN matrix is rate-stiffening, reduced matrix stiffness at lower rates may have led to higher gauge factors.

5.6.3 Effect of matrix stiffness on filler separation upon deformation: simple numerical study

The experimental data in Section 5.6.2 highlighted that in filled PBS:PDMS IDN, the separation between filler particles upon deformation is rate-dependent. Specifically, the separation increased with decreasing strain rates. It was suggested that this was possibly due to the corresponding decreasing stiffness (number of elastically active crosslinks) of the rate-stiffening PBS:PDMS IDN matrix. A simple numerical study was performed in ABAQUS/Explicit to qualitatively elucidate the correlation between matrix stiffness and filler separation under deformation. Figure 5.19(A) shows a schematic of the two-dimensional model used. The circular filler particles considered were modeled as rigid. The only form of interaction considered was hard, frictionless contact between filler and matrix. To model strain rate-dependent variations in matrix stiffness, the segments (chains) at

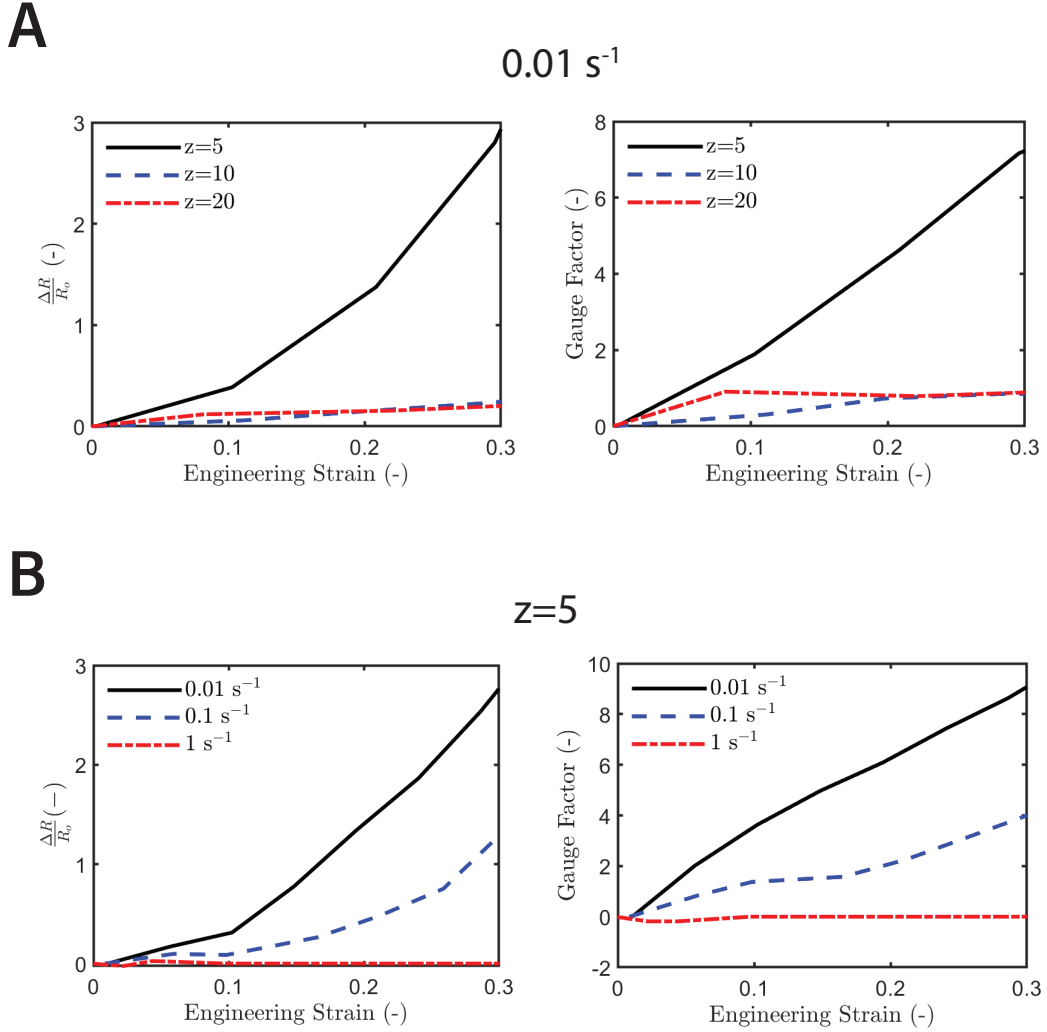


Figure 5.18: **Filler composition and rate-dependent gauge factor (sensitivity to strain) of 80:20 PBS:Sylgard 184 IDN z CNT.** The relative change in resistance ($\frac{\Delta R}{R_o}$) and gauge factor vs. engineering strain curves for **(A)** different CNT compositions ($z=5, 10, 20$) at 0.01 s^{-1} strain rate and **(B)** at different strain rates ($0.001 - 1 \text{ s}^{-1}$ for $z=5$). Lower CNT composition ($z=5$) and lower strain rates (for a fixed z) resulted in higher gauge factors and, hence, greater sensitivity to strain.

the center of the 1x1 IDN polymer mesh were connected (crosslinked) for high strain rates and unconnected (uncrosslinked) for low strain rates. The matrix was modeled as a neo-Hookean material ($C10=1$, $D1=0$ in ABAQUS). The finite element mesh of the matrix consisted of 10543 CPS4R (four-node, reduced integration plane stress) elements. The bottom edge of the matrix was fixed, and the top edge was displaced at a pseudo-speed of 1 for both stiffness cases. Although the filler placement was mostly symmetric, some

stochasticity was introduced. Figure 5.19(B) shows the evolution of filler separation with deformation for the uncrosslinked and crosslinked matrices. Filler separation along the direction of loading is higher for the uncrosslinked matrix (corresponding to low strain rates). The absence of a crosslink at the center facilitated easier separation of filler particles near the centerline. This simple numerical study supports the hypothesis that lower matrix stiffness at lower strain rates leads to greater filler separation during deformation. However, more detailed and physically accurate models are needed to fully validate this hypothesis.

5.7 Conclusion

Soft polymers, characterized by a relatively low elastic modulus in the range of a few kilopascals to tens of megapascals, have broad applications. Traditional polymers with permanent crosslinks are rate-independent with high elasticity. Dynamic, reversible crosslinks result in a significantly rate-dependent response and autonomous self-healing, although with a lack of shape stability. Interpenetrating double network polymers (IDNs) enable an efficient combination of the contrasting crosslinking mechanisms' desirable properties. Polyborosiloxane (PBS) is an important rate-stiffening soft polymer with dynamic crosslinks and diverse applications. PBS-polydimethylsiloxane (PDMS) IDNs are a promising matrix candidate for flexible sensing, impact mitigation, and actuation due to their significantly rate-dependent, self-healing, and shape retention properties. However, the large strain mechanical response of PBS:PDMS IDNs has not been systematically characterized over a range of loading conditions and compositions. This data is crucial for scientific understanding of the underlying mechanisms, model development, and practical applications.

To address these critical gaps, we have first extensively characterized the rate-dependent mechanical response of select PBS:PDMS (Sylgard 184) IDN compositions (80:20, 60:40, and 20:80 w/w) through large strain tension and compression, stress relaxation, cyclic

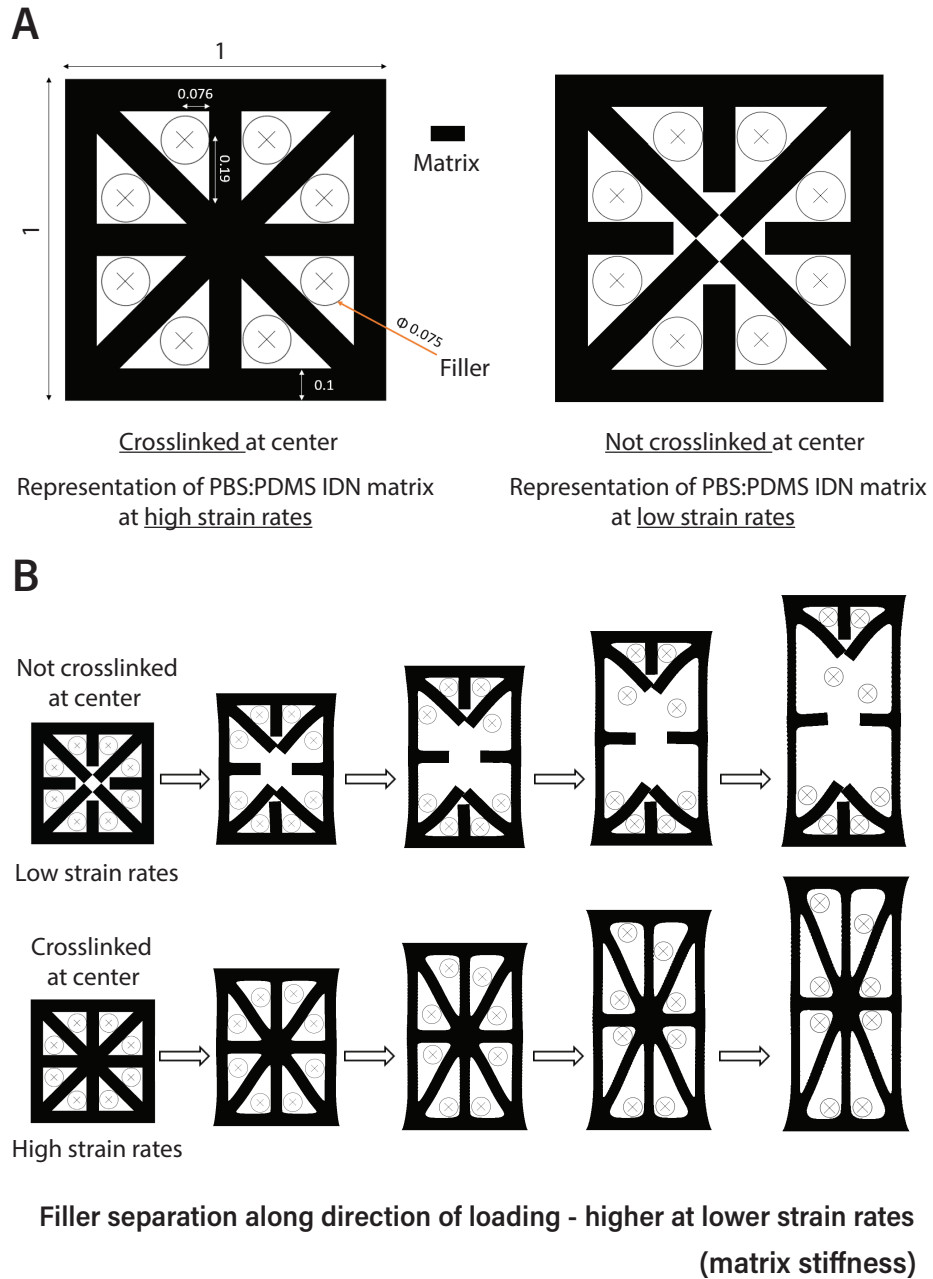


Figure 5.19: **Simple numerical study to understand the effect of matrix stiffness on filler separation upon deformation.** (A) Schematic of the simple two-dimensional model considered. To model the rate-dependent stiffness of the PBS:PDMS IDN matrix, the segments (chains) were either connected (crosslinked) or left unconnected (not crosslinked), corresponding to high and low strain rates, respectively. The circular filler particles considered were modeled as rigid, and the contact between filler and matrix was assumed to be hard and frictionless. The bottom edge of the matrix was fixed, whereas the top edge was displaced at a pseudo-speed of 1 in both stiffness cases. (B) Filler separation evolution with deformation for both matrix variations. The absence of a crosslink at the center (low matrix stiffness at low strain rates) facilitated easier separation of the filler particles along the direction of loading.

loading, and creep experiments. The stress-strain responses of the IDNs do not follow the commonly used Voigt model predictions and are significantly nonlinear with respect to composition, strain, and strain rate. The stress contribution from the approximately rate-independent Sylgard 184 network is, in general, significantly lower than the Voigt model predictions. Similar observations were also made for 80:20 PBS:Ecoflex 00-30 (another PDMS variant) IDN. For the 80:20 PBS:Sylgard 184 IDN, the stress-strain curves are not significantly different from those of PBS. The 80:20 IDN showed significant improvements in creep resistance and shape recovery upon unloading compared to PBS. Hence, this composition is ideal for practical applications in which the properties of PBS are desired without its deficiency in shape stability. The desirable, peculiar properties of the 80:20 IDN can be attributed to the Sylgard 184 network being in the initial stages of percolation or experiencing significantly lower stresses, as shown through the qualitative mechano and photochemical luminescence experiments. A basic weighted Voigt-Reuss (Hirsch) model captured the PBS:Sylgard 184 IDN response over the compositions and loadings considered with reasonable accuracy. The IDNs were found to be closer to the Reuss idealization, and the model can explain the characteristic properties of the 80:20 IDN. The simplified model-based insights should apply to other microstructurally similar IDNs. The weights depend on composition and strain rate, and a detailed understanding of polymer network interactions is required for a comprehensive model development.

The 80:20 IDN was used as a rate-dependent matrix for strain and strain-rate sensing, with carbon nanotubes (CNTs) as conductive fillers. The large deformation stress-strain response of 80:20 IDN z CNT (z is the wt% of the total IDN precursor mixture) was characterized over a range of compositions ($z=5, 10, 20$) and strain rates ($0.001-1 \text{ s}^{-1}$). A high CNT composition ($z=20$) resulted in a relatively stiffer and more brittle response. $z=5$ CNT composition exhibited a significantly rate-dependent response, crucial for strain rate sensing. Static electrical measurements showed that the resistivity decreased in a highly nonlinear fashion with increasing CNT composition. Dynamic electrical measurements

during deformation revealed that lower CNT composition ($z=5$) and lower strain rates resulted in higher gauge factors, i.e., greater sensitivity to strain. The 80:20 IDN is therefore a suitable matrix candidate for flexible strain and strain rate sensing. A simple numerical study suggested that the increase in the gauge factor with decreasing strain rate can be attributed to the corresponding decrease in the stiffness of the rate-stiffening IDN matrix, thereby facilitating easier filler separation upon deformation. Significant variations in mechanical and electrical properties can be achieved through careful design of IDNs and filled composites.

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CHAPTER 6

Modeling damage and fracture in polymeric architected materials and structures

6.1 Abstract

Architected triply periodic minimal surface (TPMS) structures offer superior performance beyond the limits of bulk materials. While recent advancements have established rate-dependent viscoplastic constitutive models to capture the complex nonlinear deformation response of polymeric TPMS structures, a significant research gap remains in predicting their failure under complex loading conditions. This work addresses this critical limitation from both scientific and practical perspectives by developing a damage and fracture model for polymeric TPMS structures. We first implement a high-fidelity viscoplastic constitutive deformation-only model from (Ma et al., 2025) into the finite element (FE) software ABAQUS by writing user subroutines, motivated by the FE method with gradient-damage being a suitable numerical framework for fracture modeling. We then propose a damage initiation criterion based on a measure of stored elastic energy

(elastic part of stress work or free energy density) and an equivalent plastic strain for amorphous polymers. The gradient-damage framework specialization and the numerical solution procedure from our recent work (Konale and Srivastava, 2025) are appropriately mapped. The developed damage constitutive model and numerical simulation capability are quantitatively and qualitatively validated through application to lattices under tension and a unit cell subjected to non-monotonic loading. The proposed fracture model can enable the comprehensive design of polymeric architected materials and structures and provide valuable physical insights.

6.2 Introduction

Superior performance—beyond the limits of bulk materials—can be achieved by exploiting topology in architected structures (Kladovasilakis et al., 2022; Greer and Deshpande, 2019; Barthelat, 2015). Among these, triply periodic minimal surface (TPMS) designs are particularly promising due to their smooth geometry, high connectivity, and inherent tunability (Qiu et al., 2024; Feng et al., 2022; Yu et al., 2019). Utilizing polymers as a base material offers additional advantages, including low density, environmental stability, and biocompatibility (Cao et al., 2023). However, predicting the performance of polymeric TPMS structures is difficult due to: (i) geometric complexity and fabrication variability, and (ii) nonlinear, strain-rate-dependent behavior of the base polymers. Ke Ma, Abhishek Gupta, and co-workers addressed these challenges by developing a rate-dependent viscoplastic constitutive model with multiple internal mechanisms (Ma et al., 2025). Material characterization, calibration, and experimental validation were hierarchically performed using dog-bone, unit-cell, and lattice specimens fabricated via stereolithography (SLA). Further, efficient large-deformation computations were enabled by Isogeometric (IGA) Kirchhoff–Love (KL) thin-shell analysis. The work thus takes a significant step toward the efficient and reliable design of lightweight, tunable polymeric architected materials and structures.

Damage and fracture modeling of polymeric TPMS structures is also important from both scientific (for physics-based insights) and practical application (for completeness of the design process) perspectives. However, there is a significant corresponding research gap in the literature. As discussed in Chapter 2 (Section 2.1), the finite element method (FEM) with gradient-damage is a suitable numerical framework for this purpose, and we have developed relevant capabilities. We make the following contributions here:

(i) Implementation of the deformation-only constitutive model from (Ma et al., 2025) in the FE software ABAQUS through user subroutines. Mapped application of the damage and fracture modeling approach for soft polymers from Chapter 2 through - development of relevant damage initiation criterion for amorphous polymers, mapping of the gradient-damage framework specialization, and numerical implementation in FEM.

(ii) Experimental validation of the developed damage constitutive model and numerical simulation capability through application to lattices under tension and a unit cell subjected to non-monotonic loading, along with quantitative and qualitative comparisons. The proposed fracture model can enable the comprehensive design of polymeric architected materials and structures and provide valuable physical insights.

6.3 Methods and results

6.3.1 TPMS unit cell geometry, fabrication, and experimental setup

Figure 6.1 summarizes the TPMS geometry for a unit cell, along with the experimental setup for unit cells and lattices (4x4x4, 2x2x2) subjected to compression and tension (Ma et al., 2025). The 5% target relative density for the unit cells and lattices from Ma et al. (2025) was considered. Solid platens on the two opposite faces normal to the printing direction were added for both the unit cell and lattice to facilitate planar loading

and to impose fixed boundary conditions at the top and bottom. The lattice fixtures were redesigned for tension experiments (see Figure 6.1(C)) to ensure that failure occurs within the lattice. Although applications of TPMS lattices typically involve compression, tensile experiments, which are seldom performed, are necessary for a comprehensive characterization and understanding of their response. Dr. Abhishek Gupta performed the experiments. Three repetitions were performed for each experimental result reported. Formlabs Form 3+ SLA 3D printer with Clear resin V4 was used for the fabrication of the specimens. Interested readers can refer to (Ma et al., 2025) for further details.

6.3.2 STL files conversion for FEM

As the specimens were fabricated using SLA and the IGA simulations utilized shell elements, we were provided with STL files. Although shell elements would be the ideal choice even for FEM, continuum elements were used, keeping in mind the restrictions of the ABAQUS FE software-based damage modeling approach in Chapter 2.1 (Section 2.6.1). Hence, the STL files had to be converted to an independently meshable solid body. We lay out the conversion process here using Autodesk Fusion 360 for the unit cells and lattices:

Unit cell

- Import the STL file in Fusion 360
- Go to ‘Mesh’ module - ‘Prepare’ - ‘Generate Face groups’
- ‘Modify’ - ‘Convert Mesh’ - Method: ‘Organic’ - Choose conversion accuracy as needed
- Export solid body as STEP file
- Import STEP file into ABAQUS

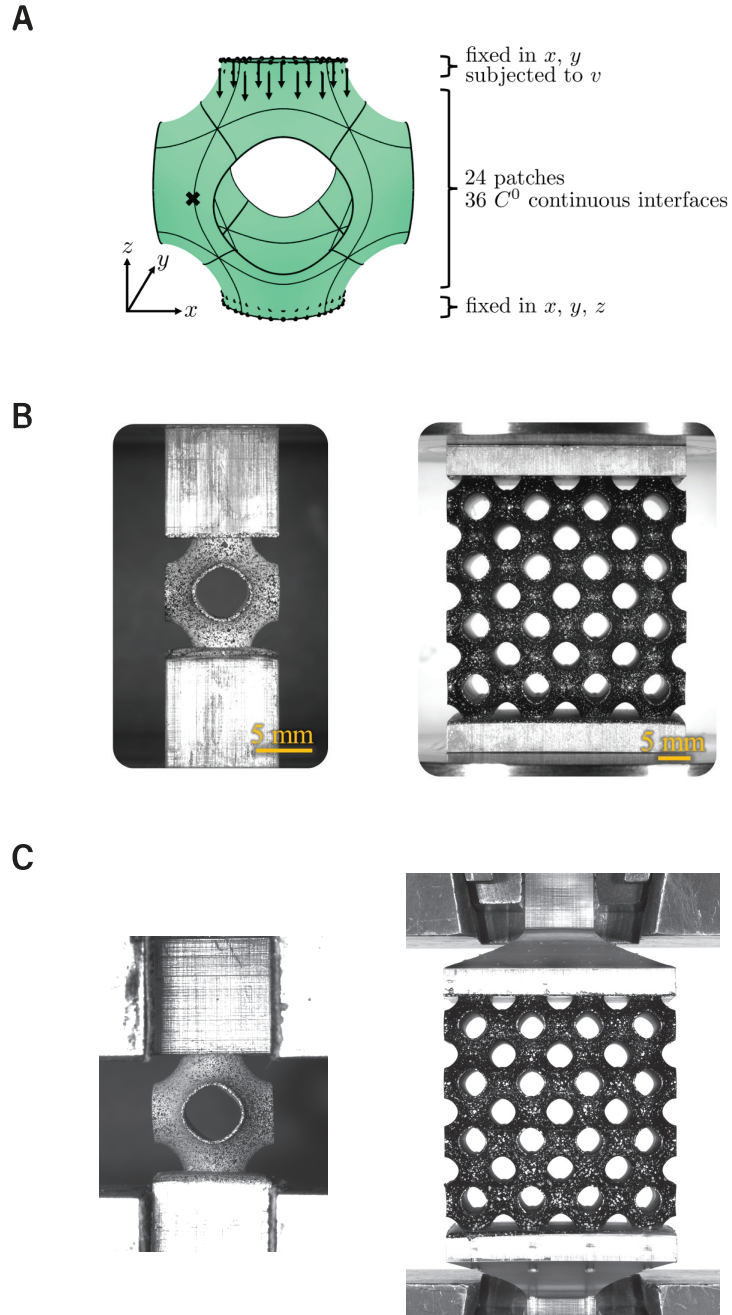


Figure 6.1: **TPMS unit cell geometry and experimental setup.** (A) TPMS unit cell geometry and Non-Uniform Rational B-Splines (NURBS) patch arrangement (Ma et al., 2025). Experimental setup for compression (Ma et al., 2025) (B) and tension (C) tests on unit cells and lattices.

- Go to 'Mesh' module in ABAQUS - 'Virtual Topology' - 'Combine faces' - select the entire part, follow the prompts
- Now the part can be meshed using the standard procedure in ABAQUS

Lattice

- Import the STL file in Fusion 360
- Reduce number of triangular elements if needed to reduce conversion time, 'Mesh' module - 'Reduce'
- 'Repair' - 'Stitch and Remove'
- 'Modify' - 'Convert Mesh' - Method: 'Organic' - Choose conversion accuracy as needed
- 'Solid' - 'Split Body' - Create datum planes to split each unit cell into eight equal parts. Partitioning in ABAQUS is impractical due to the very large computation time
- Export solid body as STEP file
- Import STEP file into ABAQUS, Choose 'Combine into single part', 'Merge solid regions', 'Retain intersection boundaries' in the dialog box.
- Go to 'Mesh' module in ABAQUS - 'Virtual Topology' - 'Combine faces' - select the entire part, follow the prompts
- There will be a 1/8th part of a unit cell that is not assigned sweep mesh control. Locate it, go to 'Tools' - 'Partition' - 'Face' - Select the red face - Use 'Shortest distance between two points' - Select the two points on that face which are not connected by a line. Now the sweep mesh control will be assigned to it
- Now the part can be meshed using the standard procedure in ABAQUS

6.3.3 Implementation of the deformation-only model from (Ma et al., 2025) in FEM

As the first step towards developing a FEM-based damage and fracture modeling tool for polymeric architected materials, we implemented the deformation-only model from (Ma et al., 2025) in ABAQUS. The model was implemented using both implicit and explicit time integration schemes via material subroutines (UMAT and VUMAT, respectively) to accommodate both slow and fast loading scenarios. For UMAT, the consistent tangent matrix reported in (Ma et al., 2025) was used directly. The implementations are comprehensively validated using single-element (uniaxial tension) and multi-element (unit cell and lattice) simulations. Figure 6.2 shows the good agreement of the single element (UMAT-C3D8, VUMAT-C3D8R) FEM stress-strain curve predictions for uniaxial tension (using the material parameters from (Ma et al., 2025)) over four decades of strain rate with the baseline MATLAB predictions from (Ma et al., 2025). We increased the mass scaling factor (MSF) for the explicit simulations (0.1 s^{-1} : $\text{MSF} = 10^6$, 0.01 s^{-1} : $\text{MSF} = 10^8$, 0.0001 s^{-1} : $\text{MSF} = 10^{10}$, 0.00001 s^{-1} : $\text{MSF} = 10^{12}$) to reduce the computation time (increase the time step size), particularly for the slower strain rate. The predictions are stable and accurate even for very large MSFs, highlighting the model's robustness.

Next, we modeled the compression of a unit cell (loading schematic described in Figure 6.1(A)). It was observed that the UMAT failed to converge in these multi-element simulations; the simulations aborted almost instantly. We also utilized a tangent obtained numerically using the perturbation method. The convergence issue was resolved; however, the predictions exhibited significant deviations from baseline results. Henceforth, we resorted to the VUMAT. Three elements were meshed through the thickness, providing a good trade-off between accuracy and computational cost. Self-contact (assumed frictionless) of the unit cell upon compression was modeled using the inbuilt ABAQUS tools. Figure 6.3 shows the FE mesh and comparison of the FEM predictions for the nominal

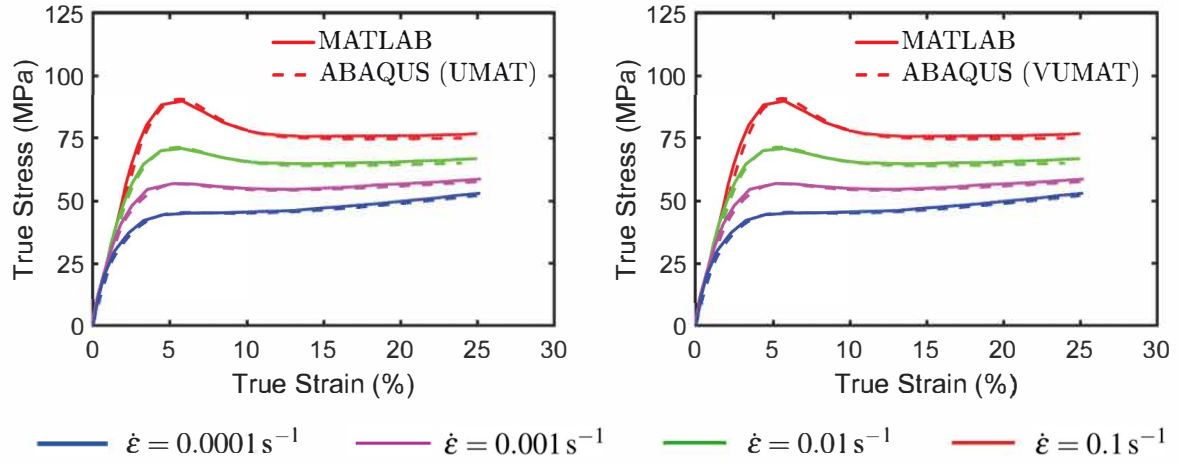


Figure 6.2: **Single element uniaxial tension: Comparison of UMAT and VUMAT predictions with baseline MATLAB results.** Good agreement of single-element UMAT and VUMAT uniaxial tension predictions with baseline MATLAB results from (Ma et al., 2025) over four decades of strain rates. For VUMAT, following MSFs were used - 0.1 s^{-1} : $\text{MSF} = 10^6$, 0.01 s^{-1} : $\text{MSF} = 10^8$, 0.0001 s^{-1} : $\text{MSF} = 10^{10}$, 0.00001 s^{-1} : $\text{MSF} = 10^{12}$. The predictions are stable and accurate even for very large MSFs, highlighting the model's robustness.

stress-strain response with the baseline IGA (uniform thickness) results for the four strain rates (0.1 s^{-1} : $\text{MSF} = 10^6$, 0.01 s^{-1} : $\text{MSF} = 10^8$, 0.0001 s^{-1} : $\text{MSF} = 10^{10}$, 0.00001 s^{-1} : $\text{MSF} = 10^{12}$). Good agreement is observed, with deviations in the densification stage attributable to differences in the contact algorithms used in FEM and IGA.

Further, we modeled the compression of a $4 \times 4 \times 4$ lattice (loading schematic described in Figure 6.1). If the unit cell mesh density in Figure 6.3(A) is used, the number of elements for a full lattice simulation will make computation time intensive. Only $1/8$ th of the lattice (front view-bottom right) was hence modeled using appropriate symmetry boundary conditions to reduce the overall computation time. A necessary modification to obtain physical results is the inclusion of a rigid surface at the top of the $1/8$ th lattice. This accounts for the self-contact with its symmetric counterpart above (see Figure 6.4(A)). The bottom platten and the additional rigid surface were modeled as analytical rigid surfaces, and the contact between them and the $1/8$ th lattice was assumed to be frictionless. These contacts and the self-contact of the $1/8$ th lattice upon compression were again modeled using the built-in ABAQUS tools. Figures 6.4(B) and (C) show the good agreement of the

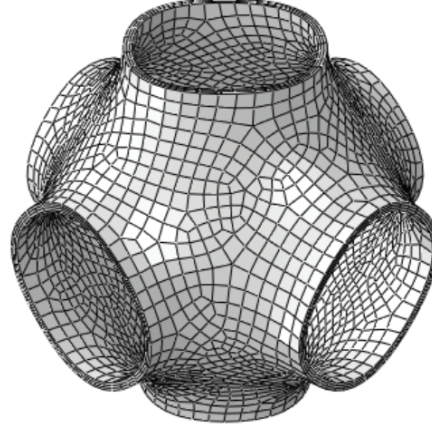
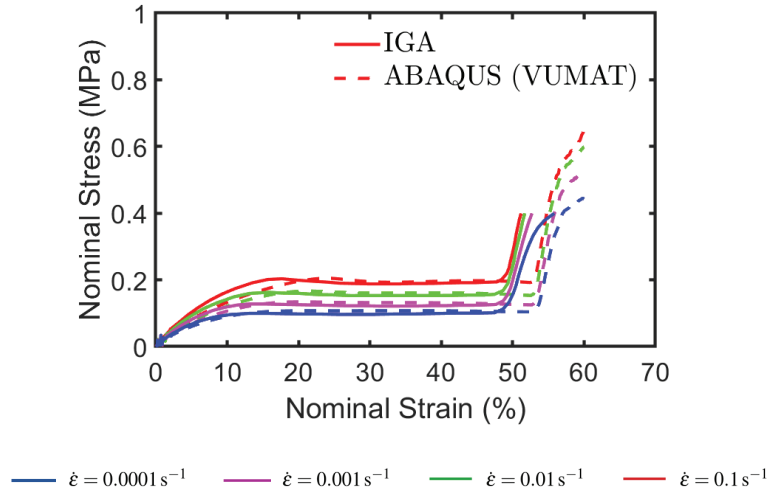
A**B**

Figure 6.3: **Unit cell compression: Comparison of VUMAT predictions with baseline IGA results.** (A) FE mesh used for the unit cell with 6201 C3D8R elements and 3 elements through the thickness. (B) Good agreement of VUMAT predictions with baseline IGA (uniform thickness) results from (Ma et al., 2025) over four decades of strain rates. The following MSFs were used - 0.1 s^{-1} : $\text{MSF} = 10^6$, 0.01 s^{-1} : $\text{MSF} = 10^8$, 0.0001 s^{-1} : $\text{MSF} = 10^{10}$, 0.00001 s^{-1} : $\text{MSF} = 10^{12}$. Deviations in the densification stage are attributable to the difference in contact algorithms used in FEM and IGA.

FEM predictions with the baseline IGA (uniform thickness) results for the 0.01 s^{-1} strain rate ($\text{MSF} = 10^8$). The mismatch in the nominal stress-strain curves after the softening regime can be attributed to differences in the contact algorithms used in FEM and IGA.

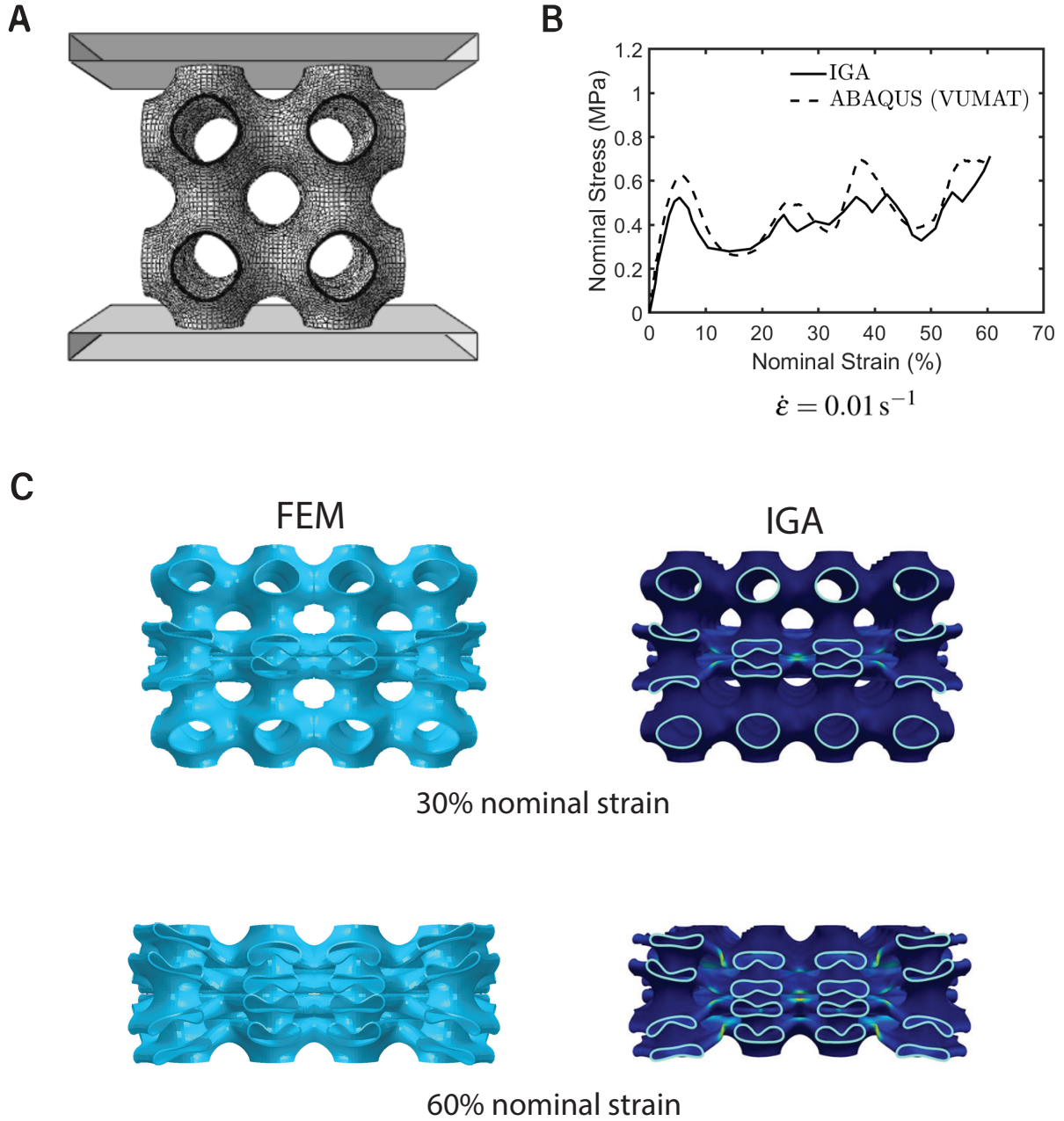


Figure 6.4: **4x4x4 lattice compression: Comparison of VUMAT predictions with baseline IGA results.** (A) FE mesh used for the 1/8 lattice (front view-bottom right) with 44649 C3D8R elements and 3 elements through the thickness. The top rigid surface accounts for the self-contact with its symmetric counterpart above. (B) Good agreement of VUMAT predictions with baseline IGA (uniform thickness) results from (Ma et al., 2025) for 0.01 s^{-1} strain rate ($\text{MSF} = 10^8$). Deviations after the softening regime can be attributed to differences in the contact algorithms used in FEM and IGA. (C) Qualitative agreement of the deformed lattice shapes (30% and 60% nominal strain) predicted by FEM with baseline IGA results.

6.3.4 Damage initiation criterion development

As the model in (Ma et al., 2025) is developed in the rate/incremental form, a free energy density function cannot be fundamentally defined, and the damage initiation criterion in Chapter 2 (Section 2.5.3) cannot be used directly. However, another measure quantifying stored elastic energy can be used, specifically the elastic part of stress work (W^e) defined as:

$$W^e = \int_0^t J \mathbf{T}_o : \mathbf{D}^e dt, \quad (6.1)$$

where $J, \mathbf{T}_o, \mathbf{D}^e$ are the determinant of the deformation gradient $\mathbf{F}$, undamaged Cauchy stress and elastic part of the stretching tensor $\mathbf{D}$, respectively. Following the arguments in Chapter 2 (Section 2.5.3), damage initiation can now be evaluated using W^e . Figure 6.5(A) shows the model-predicted W_{cr}^e vs. strain rate curve. W_{cr}^e is not constant and increases with strain rate. The variation of W_{cr}^e with another variable of interest, i.e., the equivalent plastic strain $\bar{\varepsilon}^p$, is shown in Figure 6.5(B). For amorphous polymers, plasticity could be one of the predictors of damage initiation (Narayan and Anand, 2021). W_{cr}^e decreases with increasing $\bar{\varepsilon}^p$. A possible physical interpretation is that as plastic strain increases, the stored elastic energy required to break the load-bearing bonds, leading to damage initiation, decreases. A simple relation between W_{cr}^e and $\bar{\varepsilon}^p$ can be obtained by fitting the four data points linearly as

$$W_{cr}^e = -4.39\bar{\varepsilon}^p + 1.13 \text{ MPa}, \quad W_{cr}^{e,dis} = -3.65\bar{\varepsilon}^p + 0.94 \text{ MPa}, \quad (6.2)$$

where *dis* indicates the distortional part, which is needed to ensure that damage only degrades the distortional and tensile dilatational parts of material stiffness ($\mathbf{T} = g(d)\mathbf{T}_o^+, g(d) = (1 - d)^2$).

The damage initiation criterion can then be written as

$$\text{Damage initiates when: } \begin{cases} W^e = W_{cr}^e & \text{if } J > 1 \\ W^{e,dis} = W_{cr}^{e,dis} & \text{if } J \leq 1. \end{cases} \quad (6.3)$$

We believe that damage initiation criteria like the one proposed here, based on a combination of W^e/ψ and $\bar{\varepsilon}^p$, should apply to other amorphous polymers as well. Figures 6.5(C), (D) show the model fit (FEM [UMAT, VUMAT] single element computations). Since fracture was observed to be approximately instantaneous in the experiments, it was approximated to occur in FEM (using the deformation only model) when $W^e = W_{cr}^e$.

6.3.5 Damage growth modeling

The gradient-damage framework specialization in Chapter 2 (see (2.33)) can be directly used by replacing ψ with W^e as shown below

$$\begin{aligned} W^{e+} &= \begin{cases} W^e & \text{if } J > 1, \\ W^{e,dis} & \text{if } J \leq 1, \end{cases} \\ \mathcal{H}(t) &\stackrel{\text{def}}{=} \max_{s \in [0,t]} [\langle W^{e+}(s) - W_{cr}^{e+} \rangle], \\ \langle W^{e+}(s) - W_{cr}^{e+} \rangle &= \begin{cases} 0 & \text{if } W^{e+}(s) - W_{cr}^{e+} < 0, \\ W^{e+}(s) - W_{cr}^{e+} & \text{if } W^{e+}(s) - W_{cr}^{e+} \geq 0, \end{cases} \\ \zeta \dot{d} &= 2(1-d)\mathcal{H} - 2W_*^e(d - l^2 \Delta d). \end{aligned} \quad (6.4)$$

$+$ denotes the distortional and tensile dilatational part of the corresponding quantity. The values of the damage growth related parameters are taken as $l = 2 \text{ mm}$, $\zeta = 1 \text{ kPa.s}$, $W_*^e = 4 \text{ kPa}$.

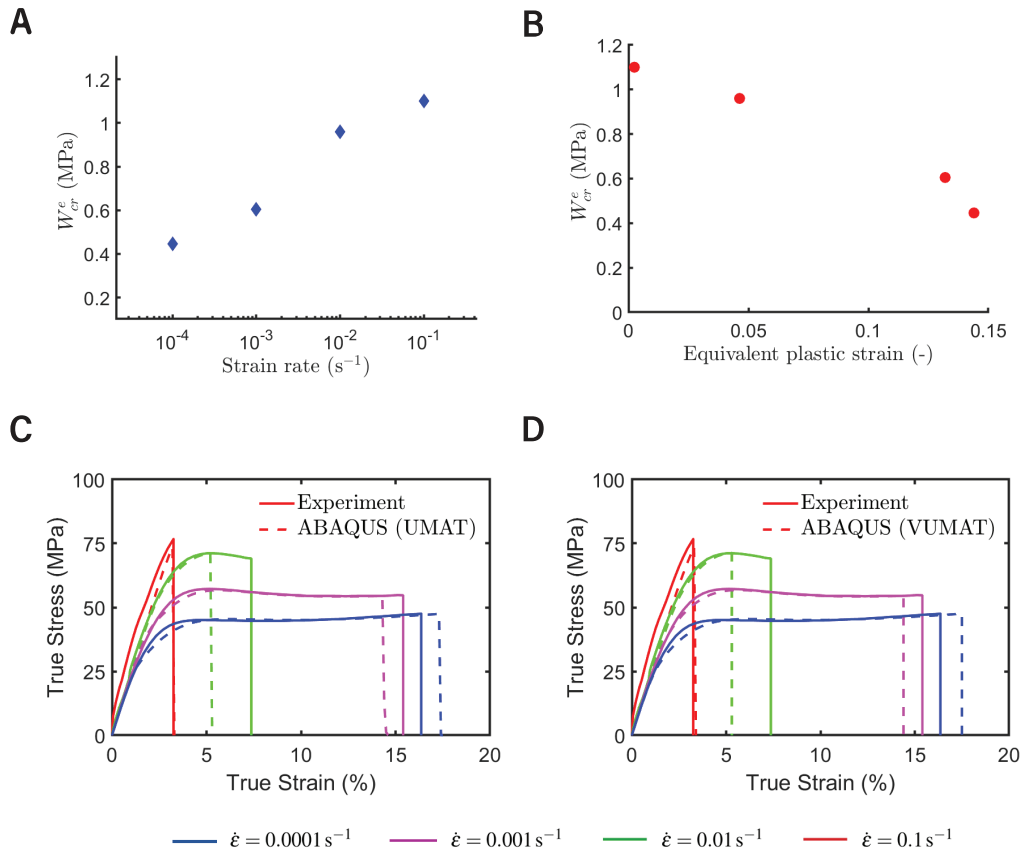


Figure 6.5: **Variation of W_{cr}^e with key parameters and model fit (fracture under uniaxial tension).** Variation of W_{cr}^e with (A) strain rate and (B) $\bar{\epsilon}^p$. Model fit [(C) UMAT and (D) VUMAT single element computations] over the four decades of strain rate. Fracture was observed to be approximately instantaneous in the experiments. Hence, it was approximated to occur in FEM (using the deformation only model) when $W^e = W_{cr}^e$.

l is treated as a numerical regularization parameter and is chosen as equal to 5 times the typical element size to avoid mesh dependency. The VUMAT-VUEL-VUSDFLD numerical solution approach for ABAQUS, discussed in Chapter 2 (Section 2.6.1), is used here. ζ (can be treated as a regularization parameter, can be reasonably changed for numerical convergence), W^{e*} were fit to the parts of the true stress-strain curves for uniaxial tension corresponding to fracture.

6.3.6 Damage initiation criterion recalibration: unit cell

While 3D-printed dog-bone specimens and unit cells share the same base material, their mechanical responses can vary slightly due to geometry-specific subtle variations in the printing process (Ma et al., 2025). Recalibrating the damage initiation criterion parameters using unit-cell experiments can account for such geometry-dependent effects and improve the model's overall predictive ability. Note that this exercise has been performed for the deformation-related model parameters in (Ma et al., 2025), and the corresponding values have been used for unit cell and lattice computations here as well. Once an element is completely damaged ($d = 1$), its stiffness is degraded to nearly zero (a small conditioning number is usually added for numerical stability), and it can undergo severe deformation. This is particularly relevant in multi-element simulations where a single severely distorted element can terminate a simulation. Also, due to the explicit scheme, damage values can be nonphysical. Hence, d from VUEL is processed as follows before using it in VUMAT to degrade the undamaged stress:

$$d^{VUMAT} = \begin{cases} 0.95 & \text{if } d^{VUEL} > 0.95, \\ 0 & \text{if } d^{VUEL} < 0, \end{cases} \quad (6.5)$$

with the subscripts $VUMAT$, $VUEL$ distinguishing d in the two subroutines. Note that the maximum value of d^{VUMAT} is capped at 0.95, so that the element retains a small but sufficient stiffness to avoid severe distortion and aid in numerical convergence. This d^{VUEL}

processing is applied alongside the default element distortion settings in ABAQUS. The parameters in the assumed linear relationship between W_{cr}^e and $\bar{\varepsilon}^p$ [(6.2)] are recalibrated to tensile experiments on unit cells as

$$W_{cr}^e = -1.782\bar{\varepsilon}^p + 1.497 \text{ MPa}, W_{cr}^{e,dis} = -0.883\bar{\varepsilon}^p + 0.857 \text{ MPa}. \quad (6.6)$$

The model fit over the four decades of strain rate is shown in Figure 6.6(A) through nominal stress-strain response predictions till complete failure. The FE mesh in Figure 6.3(A) was used. Also, the model predictions for specimen geometry at complete failure and fracture locations in 6.6(B) show good qualitative agreement with the experimental results (considering the expected variability and uncertainty of fracture measurements for polymers). Elements with $d = 0.95$ are removed for easier visualization.

6.3.7 Fracture model and numerical simulation capability validation

We now apply the recalibrated fracture model and numerical simulation capability to unseen geometries and loading scenarios involving inhomogeneous three-dimensional deformations and compare the results with experimental data for validation.

4x4x4 lattices subjected to uniaxial tension

It can be noted that experimental fracture measurements for polymers exhibit significant variability and uncertainty. Further, variations due to the printing process across the relatively large (4x4x4) lattice can exacerbate this issue. Figure 6.7 shows the model-predicted nominal stress-strain curves till complete failure. The FE mesh in Figure 6.4(A) was used. Good agreement is observed for the two higher strain rates, as the overall deformation extent to fracture is small and the probability of printing-related variations affecting the experimental results is minimal. On the other hand, for the two slower strain

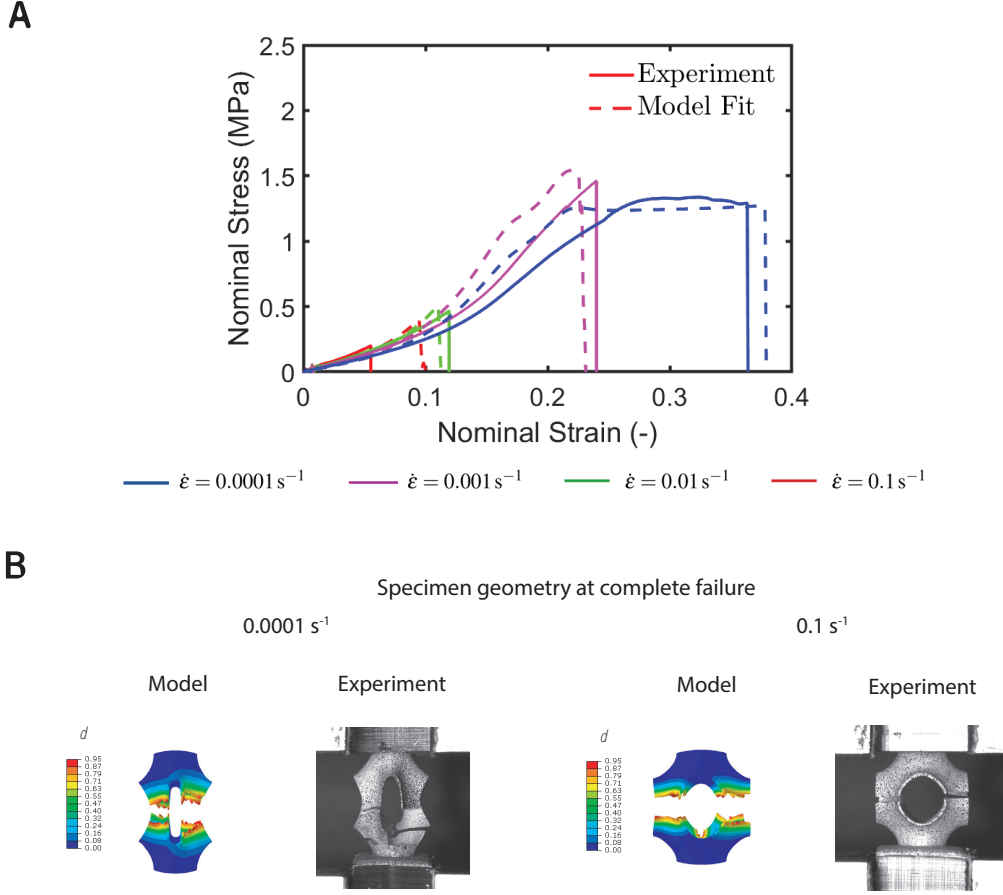


Figure 6.6: **Damage initiation criterion recalibration for unit cell and model fit (fracture of unit cell under uniaxial tension).** The 3D-printed dog-bone specimens and unit cells may exhibit slightly different material responses due to subtle variations in the printing process associated with different geometries. The damage initiation criterion parameters are hence recalibrated using unit-cell tension experiments to eliminate such geometry-dependent effects and improve the model's overall predictive ability. **(A)** The model's fit over the four decades of strain rate shown through nominal stress-strain response predictions till complete failure. The FE mesh in Figure 6.3(A) was used. **(B)** Good qualitative agreement of model-predicted specimen geometry at complete failure and fracture locations with experimental results for 0.0001 and 0.1 s⁻¹ strain rates. Elements with $d = 0.95$ are removed for easier visualization.

rates, the higher level of deformation till failure implies only a reasonable agreement can be expected (considering the factors discussed earlier), as seen.

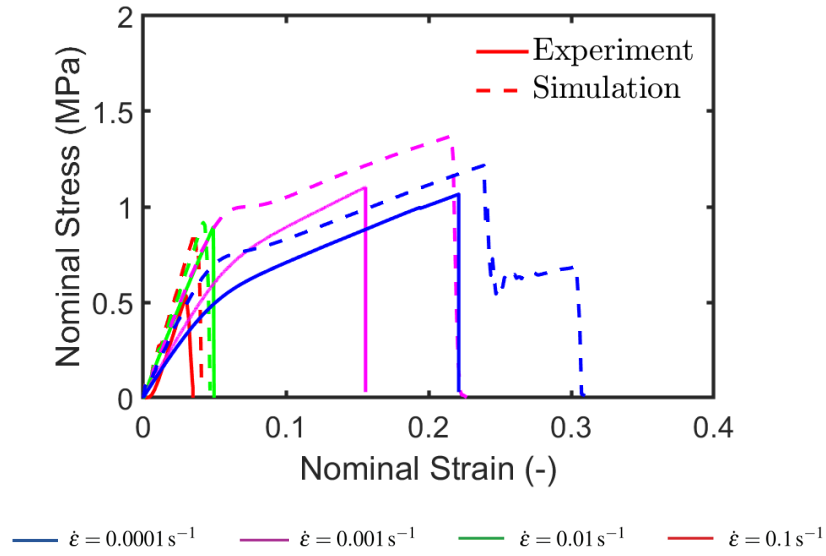


Figure 6.7: **Model validation: fracture of 4x4x4 lattice under uniaxial tension.** Experimental fracture measurements for polymers exhibit significant variability and uncertainty. Reasonable agreement (considering the additional variations due to the printing process across the relatively large lattice) of the model-predicted nominal stress-strain curves till complete failure with the experimental results over the four decades of strain rate. The FE mesh in Figure 6.4(A) was used. At the two slower strain rates, the higher level of deformation to failure suggests that the probability of printing-related variations affecting the experimental results is high, as seen.

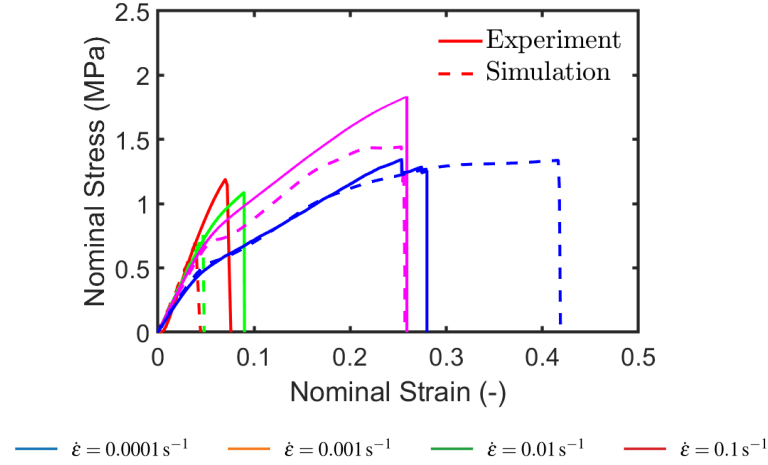
2x2x2 lattices subjected to uniaxial tension

A simpler lattice system (2x2x2) was considered next to reduce the controllable (printing-related) experimental variations. Figure 6.8(A) shows the model-predicted nominal stress-strain curves to complete failure, in good agreement (considering the experimental results variation during polymer fracture discussed earlier) with the experimental results. For the slowest strain rate, the deviation is expected following the previous discussion for the 4x4x4 lattice. The FE mesh in Figure 6.3(A) was used for a unit cell as the 1/8th part (front view-bottom right) of the lattice with appropriate symmetry boundary conditions. The specimen shapes at complete failure, and the fracture locations also qualitatively agree with the experiments, as shown in Figure 6.8(B) for 0.001 and 0.1 s^{-1} . Elements with $d = 0.95$ are removed for visualization. The fracture model, hence, provides reasonably accurate quantitative predictions while capturing the key physical phenomena.

Unit cell subjected to non-monotonic loading

For further validation, especially of the proposed damage initiation criterion in (6.6), we consider a unit cell subjected to a non-monotonic loading profile (shown in Figure 6.9(A)). During unloading, W^e will become zero in the regions of interest at some time instant while $\bar{\epsilon}^p$ is non-zero, allowing a more detailed probing of damage's dependence on these two variables. Good quantitative and qualitative agreement (considering the variations in experimental results for polymer fracture discussed earlier) of model predictions and experimental results is shown in Figures 6.9(B) and (C). The FE mesh in Figure 6.3(A) was used. Elements with $d = 0.95$ are removed, and positive stress values are plotted for visualization. The damage initiation criterion, hence, captures the dependence of damage on both stored elastic energy and plastic strain.

A



B

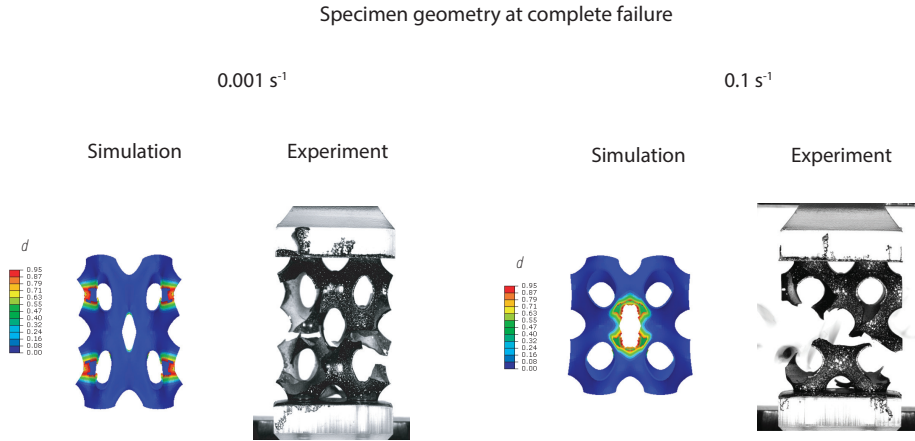


Figure 6.8: **Model validation: fracture of 2x2x2 lattice under uniaxial tension.** (A) Good agreement of the model-predicted nominal stress-strain curves till complete failure with the experimental results (considering the variations in experimental results for polymer fracture). The deviation is expected for the slowest strain rate following the previous discussion for the 4x4x4 lattice. The FE mesh in Figure 6.3(A) was used for a unit cell as the 1/8th part (front view-bottom right) of the lattice with appropriate symmetry boundary conditions. (B) The model correctly predicts specimen shapes at complete failure along with fracture locations for 0.001 and 0.1 s^{-1} . Elements with $d = 0.95$ are removed for visualization. Along with providing reasonably accurate quantitative predictions, the fracture model captures the key physical phenomenon.

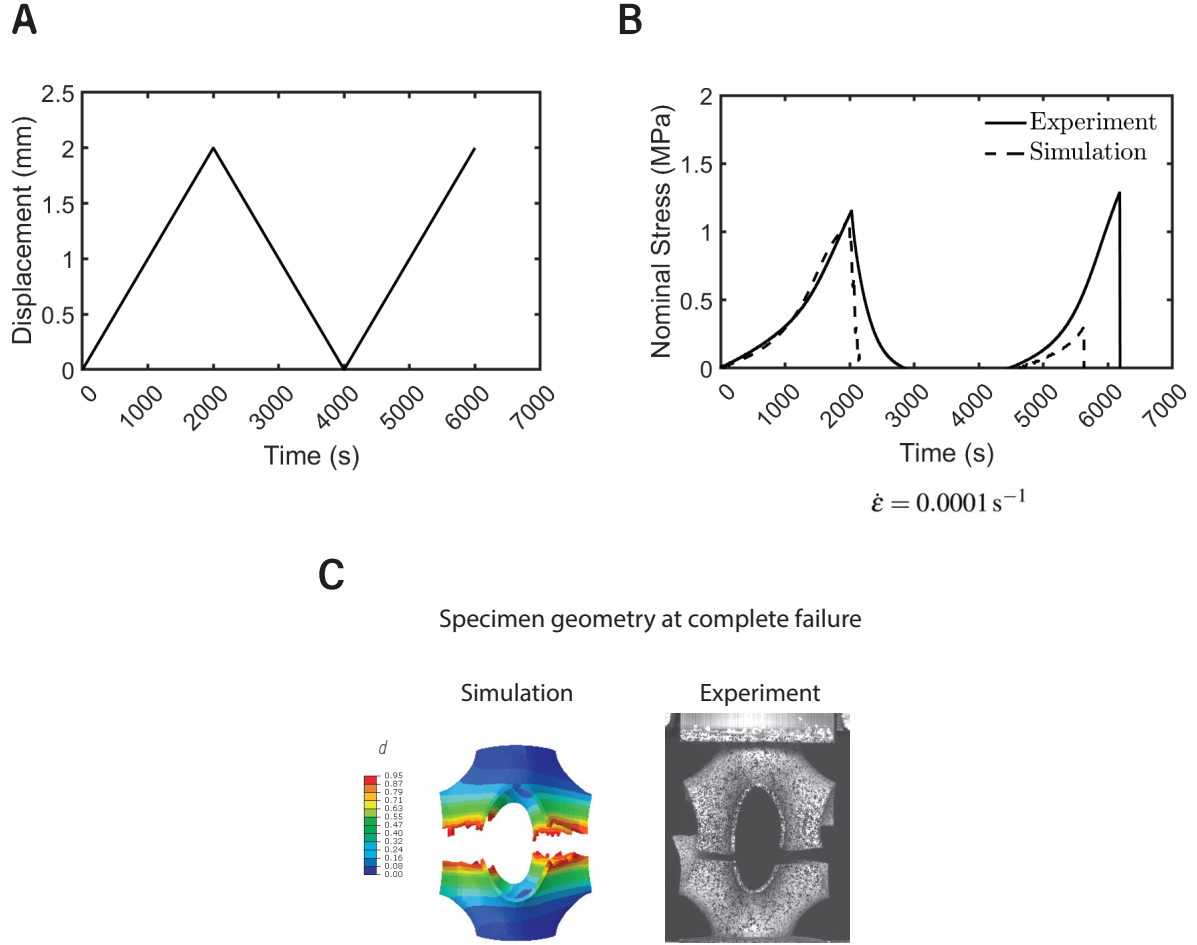


Figure 6.9: **Model validation: fracture of unit cell under non-monotonic loading.** (A) Non-monotonic loading profile. Good quantitative (A) and qualitative (B) agreement of the model predictions with the experimental results (considering the variations in experimental results for polymer fracture). The FE mesh in Figure 6.3(A) was used. Elements with $d = 0.95$ are removed, and positive stress values are plotted for visualization. The proposed damage initiation criterion captures damage dependence on both stored elastic energy and plastic strain.

6.4 Conclusion

Polymeric triply periodic minimal surface (TPMS) architected materials and structures offer an efficient route for obtaining superior performance while maintaining low density and environmental stability. The significant challenge of modeling the complex deformation response of polymeric TPMS designs has been addressed in a recent work (Ma et al., 2025). However, the scientifically and practically important aspect of damage and fracture modeling has a significant research gap in the literature. We attempt to fill this critical gap by leveraging the damage and fracture model and numerical simulation capability for soft polymers developed in Chapter 2.

We began by implementing the deformation-only model from (Ma et al., 2025) in the finite element software ABAQUS through user subroutines. The implementation is robustly tested by applying it to single-element (uniaxial tension) and multi-element (unit cell and lattice) simulations, and by comparing the results with the baseline. Next, we proposed a damage initiation criterion based on a measure of stored elastic energy (elastic part of stress work or free energy density) and an equivalent plastic strain for amorphous polymers. We then appropriately mapped the gradient-damage framework specialization and the numerical solution procedure from Chapter 2. The fracture model was then applied to loading scenarios involving three-dimensional, inhomogeneous deformation (4x4x4, 2x2x2 lattices subjected to uniaxial tension, unit cell under non-monotonic loading), followed by comparison with quantitative and qualitative experimental results for validation. The developed fracture model and numerical simulation capability can enable the comprehensive design of polymeric architected materials and structures and provide valuable physical insights.

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CHAPTER 7

Closing remarks and future work

7.1 Closing remarks

Soft polymers are important materials with applications ranging from biomedical to structural. The mechanical properties of these polymers can vary significantly depending on the underlying crosslinking mechanisms. Currently, deformation, damage, and fracture models that could predict the strongly rate-dependent, non-linear response of a broad class of soft polymers are lacking. Towards this important research gap, in this dissertation, we have experimentally characterized the large strain response of a representative, key rate-stiffening soft polymer PBS; developed a three-dimensional, large deformation, rate-dependent, elastic-viscoplastic constitutive model for PBS and similar dynamically crosslinked polymers; developed a model to predict damage initiation, damage growth, and complete failure in soft polymers using a multi-mechanism generalized gradient-damage framework; implemented these models using the traditional FEM and emerging ML-based approaches for validation and predictive simulations. The highlights and research findings of each chapter are summarized as follows:

In *Chapter 1*, we have experimentally characterized the large strain compression response of PBS up to a true strain of $\sim 125\%$ across a broad strain rate range from 10^{-3} s^{-1} to 10^3 s^{-1} . To describe this behavior, we have developed a three-dimensional, large deformation, rate-dependent, elastic-viscoplastic constitutive model. Motivated by microstructural physics, this model is designed for PBS and similar soft polymers or gels consisting of dynamic crosslinks. The mathematically straightforward framework accounts for dynamic crosslink kinetics and the dominant deformation mechanisms in PBS. Specifically, it captures the transition from dissipative resistance due to chain sliding, which dominates the mechanical response at low strain rates, to the primary contribution of network stretching resistance at high strain rates. The model accurately predicts PBS's response across six orders of magnitude in strain rate (10^{-3} s^{-1} to 10^3 s^{-1}) and has been validated through inhomogeneous deformation experiments. Based on the macroscopic mechanical response, we propose two distinct types of crosslinks with significantly different relaxation timescales (τ) based on the macroscopic mechanical response, namely B:O coordinate-bond dynamic crosslinks ($\tau \approx 3 \text{ s}$), and temporary entanglement lockups at high strain rates acting as crosslinks ($\tau \approx 0.0005 \text{ s}$). A numerical procedure is also outlined to evaluate convolution-like integrals arising from dynamic crosslink kinetics for model implementation in FE softwares.

In *Chapter 2*, we have proposed and demonstrated that a critical value of stress work, W_{cr} , serves as an approximately unique measure of fracture resistance for a certain class of soft polymers. This parameter quantifies the total energy dissipation per unit volume required for the complete failure of an arbitrary material point under a given loading mode, provided the loading rates are sufficient to initiate damage. Here, “complete failure” refers to the inability to support mechanical stress. While W_{cr} can be numerically evaluated for general deformations, it is directly obtainable from experimental data in the special case of homogeneous deformations. We have developed a model to predict damage initiation, damage growth, and complete failure in soft polymers using a multi-mechanism generalized

gradient-damage framework based on the method of virtual power, which accounts for material strength. Following the microstructural physics motivation from Chapter 1, the model accounts for the time-varying number of chain segments contributing to the free energy and their individual stretches, as governed by the underlying crosslink kinetics. The damage initiation criterion, ψ_{cr}^+ (representing the energetic contribution to W_{cr}), can be evaluated using a one-dimensional numerical implementation of the deformation-only constitutive model alongside results from homogeneous deformation experiments, such as simple tension tests. The predictive capabilities of this fracture model were validated for three elastomers (EPDM, SBR, NR), a viscous soft polymer (EPS25 vitrimer), and PBS.

In *Chapter 3*, we have proposed a PINN without gradient-damage to model fracture. Specifically, it does not require training data and does not have the complexities associated with the numerical and mathematical implementation of the gradient-damage method. The PINN is formulated to model large deformation fracture in elastomers. We have validated the predictive capabilities of the PINN (PINN without gradient-damage) for various defect configurations using baseline solutions obtained from FEM with gradient-damage. The proposed modeling framework and neural network architecture should motivate PINNs without gradient-damage to model damage and fracture in a wide range of materials. This is demonstrated through the PINN’s successful application to a biological tissue. The PINN, as a fracture modeling method, is evaluated by systematically varying key parameters. The PINN crack path predictions are approximately insensitive to the distribution of collocation points. We have also shown that relatively computationally inexpensive one-time step PINN solutions yield reasonably accurate crack paths for rate-independent materials under monotonic loading. The results indicate that in elastomer-like materials, fracture progression and path evolution are primarily governed by the damage initiation criterion.

In *Chapter 4*, we continue the theme of utilizing PINNs as an alternative numerical method for soft polymer mechanics. Specifically, we develop a PINN for modeling large deformation frictionless contact between elastomers and rigid bodies. Various automatic differentiation (AD)-based approaches are presented and assessed for evaluating the tangential vector on the deformed body's surface necessary to enforce the frictionless conditions. The PINN's predictive capabilities have been validated for various contact surface configurations using baseline solutions from FEM. The approach using the deformation gradient for evaluating the deformed tangential vector produces the best results. A preliminary extension of the formulation to elastomeric deformable-deformable body frictionless contact is also discussed, although not promising. The proposed simplistic modeling framework and neural network architecture should motivate PINNs to model large deformation contact in a variety of materials.

In *Chapter 5*, with the initial aim of addressing PBS's lack of shape stability for practical applications, we have characterized the large strain tensile and compressive stress-strain and stress relaxation response of PBS:PDMS IDNs over three decades of strain rate (10^{-3} s^{-1} - 10^0 s^{-1}). The 80:20 w/w PBS:PDMS IDN exhibits improved shape stability and recovery upon unloading, without a significant change in the stress-strain response compared to pure PBS. A basic model is shown to reasonably capture the IDN response and can explain the desirable, peculiar properties of the 80:20 IDN. The large deformation, rate-dependent (10^{-2} - 10^{-1} s^{-1}), stress-strain, and electrical response of 80:20 IDN - CNT composites as a model system for flexible strain and strain rate sensing applications is also characterized. A wide range of mechanical and electrical properties can be achieved through careful design of IDNs.

In *Chapter 6*, we map the damage and fracture modeling approach for soft polymers from Chapter 2 for polymeric architected materials. We first implement a high-fidelity rate-dependent viscoplastic constitutive deformation-only model from (Ma et al., 2025) into the FEM framework, motivated by the FE method with gradient-damage being a suitable numerical framework for fracture modeling. We then propose a damage initiation criterion based on a measure of stored elastic energy (elastic part of stress work or free energy density) and an equivalent plastic strain for amorphous polymers. The gradient-damage framework specialization and the numerical solution procedure from Chapter 2 are appropriately mapped. The developed damage constitutive model and numerical simulation capability are quantitatively and qualitatively validated through application to lattices under tension and a unit cell subjected to non-monotonic loading. The proposed fracture model can enable the comprehensive design of polymeric architected materials and provide valuable physical insights.

In the *Appendix*, we have presented the pseudo codes for the fracture model (Chapters 2 and 6) implementation in the FE software ABAQUS.

7.2 Outlook on future research

In this dissertation, we have attempted to make several meaningful contributions to the field of soft polymer mechanics. The presented experimental data, developed deformation and fracture models, and their implementations in traditional as well as emerging numerical methods can enable comprehensive end-to-end design (structure geometry optimization for a given function) of soft polymer-based products and devices (e.g., flexible impact mitigation and sensing using PBS). Although this work establishes a comprehensive framework, it also highlights many promising avenues for future exploration. We recognize that certain limitations remain and that several intriguing questions have yet to be fully addressed. To assist future researchers, we have outlined the key considerations and potential next steps for each chapter below:

Chapter 1: New microstructural experimental techniques in the future, which enable direct measurement of dynamic crosslink formation, relaxation timescales, and associated subchain lengths and densities, will allow for validation of the estimates for PBS provided in this study through a fitting process to the macroscopic mechanical response. Material response characterization in the challenging intermediate strain rate regime (10^0 s^{-1} - 10^3 s^{-1}) is an active area of research. Novel experimental methods under development for this purpose will enable characterization of the material response of PBS in this important intermediate strain rate regime. Future intermediate and high strain rate experiments on PBS involving inhomogeneous deformations will enable more robust validation of the model.

Chapter 2: In the future, more experiments with complex specimen geometries, loading rates, and loading modes can be performed on a variety of distinct soft polymers to further study the approximate independence of the critical stress work from these variables. The field of damage and failure of soft materials is challenging and continuously evolving. Accurate and useful damage and fracture models applicable to a wide range of materials require robust constitutive models that capture all important physics of damage initiation and growth, along with robust computational implementations. Developing concise, physics-based, broadly applicable damage initiation criteria for complex multidimensional loadings is essential, as damage initiation primarily governs fracture progression and path evolution in soft polymers.

Chapter 3: The relatively new nature of PINNs as a numerical method implies that solution quality depends on multiple factors, and problem-specific insights are necessary for successful application. Hence, there is a need for generalizable understanding and guidance for PINNs for solid mechanics problems. However, the relative ease of mathematical formulation, numerical implementation, and mesh-free nature of PINNs are significant advantages. Future work can utilize adaptive collocation point distribution and explore new pre-training and output normalization strategies. The PINN architecture and

implementation can be made more computationally efficient to enable smaller time step sizes and training for a greater number of epochs. Efficient incorporation of physics into the continuously advancing ML-based numerical methods (e.g., DeepONet) can enable robust real-time digital twins and rapid design optimization.

Chapter 4: Further detailed studies are needed to fully understand the applicability of the PINN and the proposed approaches for the deformed tangent vector evaluation. Custom Autograd functions for geometric packages that directly output the intersection measures will enable even easier zero penetration enforcement, especially for contact between deformable bodies. Efficient and robust incorporation of frictional effects is important for accurate contact modeling in practical scenarios.

Chapter 5: More insightful microscopic and macroscopic experiments are needed to develop a comprehensive understanding of the underlying mechanisms. Generally applicable continuum-scale models informed by these experiments to predict the mechanical, electrical, and other functional responses of IDNs and IDN-filler composites for a given composition will be useful from both scientific and practical perspectives. The significant variation in properties with changes in the composition of the stiffer polymer network, as observed for the PBS:PDMS IDN, is related to numerous biological phenomena, e.g., lung fibrosis (Liu et al., 2017), skin aging (Aziz et al., 2016), and embryo phase transition (Petridou et al., 2021). Insights from extensive research on synthetic IDNs should extend to these important phenomena.

Chapter 6: Further model applications are necessary for a detailed assessment of the predictive capabilities. Future work can develop or leverage existing experimental techniques to better understand the damage-associated microstructural mechanisms, as well as to generate macroscopic data for robust model verification, specifically the proposed damage initiation criterion for amorphous polymers under more complex loadings.

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Appendix

A.1 VUMAT-VUEL-VUSDFLD: pseudo input file and subroutines

A.1.1 Input file

A dummy model with the intended geometry and boundary conditions can be created in ABAQUS CAE (Complete ABAQUS Environment). The input file for the dummy model generated by ABAQUS can be modified as follows, with the changes highlighted in bold. For an actual mesh with M elements and m nodes for the VUMAT, a dummy mesh can be created for the VUEL with the same nodal coordinates but element numbers offset by a number $N \geq M$.

*Heading

*Part

*Node

The node numbers and associated coordinates for the mesh created are provided here by ABAQUS.

***Element, type=C3D8R**

Element numbers corresponding to the actual mesh and associated node numbers are written here by ABAQUS.

***User Element, Nodes=8, Type= , Properties= , Coordinates=3, Variables=**

11

***Element, Type=**

Here, the user element to be used for VUEL can be defined. “Type” has to be in the form of “VUn”, where n is any positive integer of choice less than 10000. “Properties” has to be set equal to the number of material properties to be used in the VUEL. “Variables” has to be set equal to the number of state-dependent variables used in the VUEL. The only degree of freedom activated for these elements is “11”. This is the conventional temperature; however, for user elements, it can be any scalar degree of freedom of choice while using the built-in solution procedures for thermo-mechanically coupled problems.

Element numbers corresponding to dummy mesh and associated node numbers should be inserted here.

The sets corresponding to all nodes, the actual mesh for VUMAT, and the dummy mesh for VUEL can now be defined for subsequent use.

***Nset, nset=nodesall, generate**

1, m, 1

***Elset, elset=elvumat, generate**

1, M, 1

***Elset, elset=elvuel, generate**

1+N, M+N, 1

Node and element set definitions for mesh seeds, if any, are written here by ABAQUS.

Now, the section/material for the actual mesh and values of the material properties used by VUEL can be assigned.

***Solid Section, elset=elvumat, material=“Mechanical material”**

***uel property, elset=elvuel**

Assign values to material properties used by VUEL here in the format.

“ ”, “ ”, “ ”, “ ”,

***End Part**

***Assembly, name=**

***Instance, name=, part=**

***End Instance**

A new set of all nodes for assigning damage-related initial conditions can now be defined.

***Nset, nset=nodesalld, instance= , generate**

1, m, 1

The node, element set, and surface definitions corresponding to the dummy model are specified by ABAQUS here.

```
*End Assembly
```

Element controls defined in the dummy model are written by ABAQUS here.

```
*Section Controls, name= , ...
```

Tabular amplitudes in the dummy model; if any are written by ABAQUS here.

```
*Amplitude, name= , ...
```

Now the material “Mechanical material” is defined

```
*Material, name=“Mechanical material”
```

```
*Density
```

```
*Depvar
```

```
*User Material, constants=
```

Assign values to material properties used by VUMAT here in the format.

```
“ ”, “ ”, “ ”, “ ”, ....
```

“Depvar” has to be set equal to the number of state-dependent variables defined in the VUMAT. “Constants” is equal to the number of material properties used by VUMAT.

```
*User Defined Field
```

This line creates a dummy user-defined field calling the subroutine VUSDFLD.

The initial conditions for the damage degree of freedom are prescribed now.

***Initial conditions, type=temperature**

nodesalld,0.0

Initial mechanical boundary conditions are defined in the dummy model and are written now by ABAQUS.

*Boundary

ABAQUS now writes the steps defined in the dummy model.

*Step, name= , nlgeom=YES

***Dynamic Temperature-displacement, Explicit**

This line instructs ABAQUS to evaluate the scalar degree of freedom of choice in VUEL using the in-built solution procedure for temperature in thermo-mechanically coupled problems.

Mechanical loads or boundary conditions applied during the step and defined in the dummy model are written now by ABAQUS.

*Boundary

Finally, the field and history output requests for the step defined in the dummy model are written by ABAQUS.

*Output, field, number interval=

*Node Output

*Element Output

*Output, history, time interval=

*End Step

A.1.2 Subroutines

The discrete version of the evolution equation for damage following the standard Galerkin approximation is given as

$$\mathbf{M}_d \dot{\mathbf{d}}_N^T = \mathbf{P}_d^T, \quad (\text{D.1})$$

where $\mathbf{M}_d$ is the 8x8 “mass” matrix corresponding to damage evolution. $\mathbf{d}_N$ is the 1x8 nodal damage vector so that $d = \mathbf{N}_d \cdot \mathbf{d}_N$ with $\mathbf{N}_d$ being the 1x8 shape function vector. Shape functions used are the same as those used for the C3D8R stress element. With the volume of an undeformed element and its boundary denoted by Ω and $\partial\Omega$ respectively, we have

$$\mathbf{M}_d = \int_{\Omega} \zeta \mathbf{N}_d^T \mathbf{N}_d \, dv_R, \quad (\text{D.2})$$

and $\mathbf{P}_d$, the 1x8 “load” vector corresponding to damage evolution as

$$\mathbf{P}_d = \int_{\Omega} \left[-2(1-d)\mathcal{H}\mathbf{N}_d^T + 2\psi_* d \mathbf{N}_d^T + 2\psi_* l^2 (\text{Grad } \mathbf{N}_d)^T \text{Grad } d \right] dv_R. \quad (\text{D.3})$$

The integrals in (D.2) and (D.3) can be evaluated numerically for the reduced integration user element using Gaussian integration as

$$\mathbf{M}_d = 2\mathcal{J}\zeta \mathbf{N}_d^T \mathbf{N}_d, \quad (\text{D.4})$$

and

$$\mathbf{P}_d = 2\mathcal{J} \left[2(1 - \mathbf{N}_d \cdot \mathbf{d}_N) \mathcal{H} \mathbf{N}_d^T - 2\psi_* (\mathbf{N}_d \cdot \mathbf{d}_N) \mathbf{N}_d^T - 2\psi_* l^2 (\nabla \mathbf{N}_d)^T \nabla (\mathbf{N}_d \cdot \mathbf{d}_N) \right], \quad (\text{D.5})$$

where $\mathbf{N}_d$, $\nabla \mathbf{N}_d$ are evaluated at the center of the element mapped to a unit cube, and $\mathcal{J}$ is the Jacobian of the element under consideration mapping it to a unit cube. It can be seen from (D.5) that only $\mathcal{H}$ from the mechanical problem is needed to evaluate d . We note again that VUMAT only tracks material/integration points and references their numbers during each call. VUEL, on the other hand, stores only the element numbers and references them during each call. VUSDFLD retrieves the element numbers corresponding to the actual mesh and stores them as a state variable in VUMAT. Reduced integration elements imply that the integration/material point number is equal to the element number, ensuring correct integration/material point by integration/material point passing of d and $\mathcal{H}$ between VUMAT and VUEL. The subroutines can now be summarized as follows:

The global variables are defined here

```

module glomodule

parameter (NumEle=M)

real*8,save :: allD(NumEle),allH(NumEle)

end module

```

allD and allH are global arrays with size - Number of elements (NumEle) x 1, which store the damage (d) and damage driving history variable ($\mathcal{H}$) values at the integration points of all elements at a time instant.

VUMAT for the mechanical problem follows

```

subroutine vumat()

use glomodule

```

This line allows the use of global variables in VUMAT.

```
do km = 1,nblock
```

Obtain the element number of the material point called

```
ElemID = stateOld(km,p)
```

p denotes the state variable number used for this purpose

Obtain the evaluated value of d from the VUEL to update the total Cauchy stress

```
d = allD(ElemID)
```

Use the deformation gradient $\mathbf{F}$ at the previous and current time instant provided by ABAQUS to perform explicit integration of the differential equations involved. Specifically, explicitly update the state variables: the total Cauchy stress $\mathbf{T}$, stress work, total undamaged free energy ψ_o , and $\mathcal{H}$.

Store the element number for usage in the next time step

```
stateNew(km,p) = ElemID
```

Store d as a state variable (number q) for damage field visualization

```
stateNew(km,q) = d
```

Pass evaluated $\mathcal{H}$ value to allH for use in VUEL

allH(ElemID) = $\mathcal{H}$

enddo

return

end subroutine vumat

VUEL for damage evolution follows

subroutine vuel()

use glomodule

This line allows the use of global variables in VUEL

do kblock = 1,nblock

Use the undeformed nodal coordinates to evaluate $\mathcal{J}$

Obtain the evaluated value of $\mathcal{H}$ at the current time instant from the VUMAT to calculate $\mathbf{P}_d$

$\mathcal{H} = \text{allH}(\text{jelem}(\text{kblock}) - N)$

Use $\mathcal{J}$, $\mathcal{H}$ and $\mathbf{d}_N$ at the current time instant provided by ABAQUS to evaluate $\mathbf{M}_d$ and $\mathbf{P}_d$ using (D.4) and (D.5).

Fill out amass and rhs required by VUEL

do i=1,8


```
amass(kblock,i,i)= $\mathbf{M}_d(i,i)$ 
```

```
enddo
```

```
do i=1,8
```

```
rhs(kblock,i) =  $\mathbf{P}_d(i,1)$ 
```

```
enddo
```

Pass d at the current time instant to allD for use in VUMAT

```
allD(jelem(kblock) - N) =  $d$ 
```

```
enddo
```

```
return
```

```
end subroutine vuel
```

VUSDFLD for providing element numbers to VUMAT follows

```
subroutine vusdfld()
```

```
use glomodule
```

This line allows the use of global variables in VUEL

Store the element numbers corresponding to the material points of the C3D8R elements (actual mesh) as a state variable for use in VUMAT.

```
do k = 1, nblock
```

```
stateOld(k,p) = jElemUId(k)
```

```
stateNew(k,p) = jElemUid(k)

end do

return

end subroutine vusdfd
```

All utility/helper functions follow