

Model Discovery in Mechanics: From Conventional to Data-Driven Methods

Thesis by
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In Partial Fulfillment of the Requirements for the
Degree of
Doctor of Philosophy



CALIFORNIA INSTITUTE OF TECHNOLOGY
Pasadena, California

2027
Defended July 24, 2026

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ACKNOWLEDGEMENTS

This thesis is the culmination of my work at Caltech over the past five years, and I could not have reached this point without the support and influence of many people. I am deeply grateful to all of them.

First and foremost, I would like to thank my advisor, Kaushik Bhattacharya. He has taught me all the technical aspects of research (mathematics, mechanics, and calculus of variations) and shaped me into the researcher that I am today. He has been the most patient with me whenever I made mistakes and has given me encouragement to push through those difficult times. Beyond research, I am incredibly grateful for his support and encouragement during the more difficult periods of my PhD. Both our professional discussions and our more personal conversations have had a lasting influence on me; this support and our conversations helped me continue moving forward. I will always cherish our meetings and deep conversations, and I know I will miss them as I move on to the next stage of my life. I will always carry the lessons and life outlook I have learned from him long after I leave Caltech.

I would also like to thank Guruswami (Ravi) Ravichandran. I am glad that my time and his time at Caltech happened to overlap. During this time, I learned a great deal from his talks and his classes about experimental mechanics, and from his presence and insights during group meetings, which deepened my understanding of mechanics. Beyond these settings, he always provided both personal encouragement and research guidance for me. I always looked forward to his advice on life, on career outlook, and on research whenever we talked. I am also grateful for his support of my work, current and future.

I would also like to thank Michael Ortiz. Even though Michael spends half a year at Caltech and the rest elsewhere, I still learned mechanics through his presence and discussions during our group meetings. His questions and discussions shape me to be a better critical thinker. Beyond this, his passed-down lecture notes from the classes he taught at Caltech also help me learn continuum mechanics, computational mechanics, constitutive modeling, plasticity, among other things. I am also grateful for his support of my current and future work.

I would also like to thank Andrew Stuart. Andrew taught me math and analysis. I learned initially through his ACM classes, and his extraordinary ability of conveying math to people of different background, made a lasting impact and made me

interested in this direction. In the later years, his works on inverse problems open many exciting new directions for me and I am glad our paths cross before I leave. It is always fun to discuss with him, and I greatly value his perspective.

I would also like to thank Angkur Jyoti Dipanka Shaikeea, who has been helpful through my last year at Caltech with experiments, and with discussions about anything mechanics-related. We have overlapping interests, and it was always fun to talk to him about mechanics, Caltech, and life. Beyond this, he also provided me great emotional support, and I am incredibly grateful for this.

I would also like to thank the entire Kaushik-Ravi-Ortiz group, for the discussions and both technical help and emotional support during different parts of my PhD. There is a long list of members that have come and gone, but I shall try to list them in a loosely-chronological order: Vatsa Gandhi, Andrew Akerson, Shengduo Liu, Mina Karimi, Jacinto Ulloa, Suraj Ravindran, Eric Ocegueda, Aakila Rajan, Sathvik Sanagala, Barry Lawlor, Maximo Cravero Baraja, Mary Agajanian, Theresa Tsaggaris, Carter Cocke, Mayank Raj, Alice Kutsyy, Aditya Shedge, Hi'ileilani Dikilato, Huan Liu, Keyang Yang, Reo Yanagi.

Among the group, I am especially and incredibly grateful for our closer group of friends: Vatsa Gandhi, Shengduo Liu, Aakila Rajan, Sathvik Sanagala, and Barry Lawlor. These people have provided such a deep emotional support through the rough times. I don't think I would have made it through this journey without them. They are always there, ready to help whenever I need anything. They are also my go-to: anytime I have a math-, coding-, or experimental-related questions, I go to them and they always help me out and (for the most part) always have answers. They are an amazing group of friends, and I am happy to know they always have my back. I will always treasure the moments and get-together that we have shared, and looking forward to many more as we move on in life.

I am also thankful for the friends I made in Caltech: Chelsea Fox, Michelle Markarian, Siffi Singh, Chien Yeh Goh, Roshan Naik, Caroline Cardinale, and those in my cohort. They have helped me in countless ways, often without even realizing it. I have always cherished our time together and our conversations about everything and nothing, which made some challenging days better. I am also thankful for our MCE admins who are the most helpful I have ever seen, especially Jenni Campbell and Lynn Seymour. I am also thankful for my friends back from Michigan and Indonesia.

Last and most important of all, I have been incredibly blessed and am forever grateful to have my family and their unwavering support: my mom, my dad, and my brother. They have always been there every step of the way, and it would still be an understatement to say that I owe everything that I have been able to do to them. Being able to write a thesis and obtain a PhD is something I never imagine to be able to achieve, and the fact that I am at this point is really a testament about their extremely sacrificial nature, their forward thinking, their continuous dedication and hard-work for the past 30+ years, day in and day out. No words can truly express how grateful I am for everything they have done for me, or how much I admire the love, strength, and selflessness they have shown throughout my life. This thesis, and everything it represents, is as much a reflection of their perseverance as it is of mine, and I dedicate it to them with all my love and gratitude.

Finally, *"Segala perkara dapat kutanggung di dalam Dia yang memberi kekuatan kepadaku."*— *Filipi 4:13*.

ABSTRACT

The design of the next generation of multifunctional and structural materials requires an understanding of their complex physics. Materials such as soft active polymers, additively manufactured metals, batteries, and biomaterials are crucial for applications including soft robotics, protection, energy storage, and biomechanical implants. Yet for most emerging functional materials, the governing physics remain only partially understood. We present two approaches to learn and develop high-fidelity models for these complex materials.

We first present a more traditional approach that combines experiments and continuum modeling to study a class of multifunctional materials called Liquid Crystal Elastomers (LCEs). Their behavior at large strains and high strain rate regime was characterized, for the first time, using a novel tensile drop-tower experimental setup. This new insight leads to the development of a complete, high-fidelity engineering model that captures their behavior across multiple orders of strain rates while remaining consistent with their underlying multiscale physics.

We then present a second approach that addresses model discovery for new, emerging materials, where physics knowledge is often sparse or incomplete. Advances in imaging now provide rich, full-field experimental data, but traditional inversion techniques fail to fully exploit this raw information. We introduce an image-to-constitutive model (I2C) method that directly infers models from raw experimental images and data, leveraging all the underlying information in the raw data to obtain high-fidelity models. This method is well-posed (the laws of physics regularize the ill-posed image correlation problem without ad hoc filters) and is amenable to any general form of constitutive relations under large deformations, non-pristine test conditions, and complex loadings. This method has been demonstrated to successfully recover the constitutive relation in rubber materials under large deformations.

Finally, this thesis presents an approach that uses the current model and physics knowledge of the material to design optimal experiments that maximize information gain. This approach couples a Bayesian framework with topology optimization to design sample geometries that produce maximally informative data within a single experiment. Together, these contributions create a closed-loop framework between modeling and experimentation for learning and developing next-generation materials.

PUBLISHED CONTENT AND CONTRIBUTIONS

- [1] A. Kutsyy, A. Wihardja, V. Lee, and K. Bhattacharya, “Characterization of the soft behavior of nematic elastomers over a range of temperature and strain rates”, *European Journal of Mechanics-A/Solids*, 106035 (2026) 10.1016/j.euromechsol.2026.106035,
A.W. participated in the conception of the project, development and validation of the experimental methodology, data analysis and validation, and model analysis. A.W. guided the undergraduate first author throughout the experimental, analytical, and writing stages of the project, and contributed to the writing and revision of the manuscript.
- [2] A. Wihardja and K. Bhattacharya, “Specimen design for material parameter identification using topology optimization”, *arXiv preprint arXiv:2607.16865* (2026) 10.48550/arXiv.2607.16865,
A.W. derived the main result of this paper.
- [3] A. Wihardja, J. C. Nieto-Fuentes, D. Rittel, and K. Bhattacharya, “High strain rate behavior of liquid crystal elastomers”, *Journal of the Mechanics and Physics of Solids*, 106501 (2026) 10.1016/j.jmps.2026.106501,
A.W developed the experimental methodology, performed the experiments, produced and processed and analyzed the experimental data, developed model, performed model simulation, and wrote the manuscript.
- [4] J. C. Nieto-Fuentes, A. Wihardja, P. Stovall, T. Wilson, K. Bhattacharya, and D. Rittel, “A new drop weight tensile testing system for soft matter at intermediate strain rates”, *European Journal of Mechanics-A/Solids* **111**, 105507 (2025) 10.1016/j.euromechsol.2024.105507,
A.W participated in developing experimental methodology, performed the experiments, produced and processed and analyzed the experimental data, developed and performed the simulation methodology, and wrote the manuscript.
- [5] A. Wihardja and K. Bhattacharya, “Constitutive relations from images”, *Journal of Applied Mechanics* **92**, 081009 (2025) 10.1115/1.4068870,
A.W derived the main result of this paper.

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Chapter 1

INTRODUCTION

The design of the next generation of multifunctional and structural materials and devices requires an understanding of their complex behavior. Materials such as soft active polymers, hydrogels, architected composites, and bio-inspired matter are important for applications including soft robotics, wearable electronics, impact-mitigating structures, biomechanical implants, and energy storage [1–6]. These materials often exhibit large deformation, nonlinear response, anisotropy, rate dependence, temperature dependence, and coupled multiphysical behavior [7–10]. Their usefulness in engineering design depends on the availability of accurate constitutive models that can predict their response for complex, real engineering applications.

Constitutive models serve as a link between internal material state variables, such as strain, stress, temperature, or microstructural descriptors, and the macroscopic response of a material. For simple or well-established material classes, decades of theory and experiments often provide a clear physics-based form for these models. For most emerging functional materials, however, the governing physics are only partially understood. Moreover, the state variables that appear in a constitutive model are often not directly accessible and therefore cannot be measured in an experiment. The challenge is thus to learn the behavior of complex multifunctional and structural materials from accessible experimental data, while leveraging all the underlying information from data to inform models that can faithfully represent their complexity.

This thesis addresses the challenge of modeling complex materials from two complementary directions. The first direction follows a traditional approach in mechanics, in which high-fidelity constitutive models are developed by designing and performing experiments, and then using the resulting observations to formulate and validate a physics-based model. In this thesis, that approach is demonstrated using liquid crystal elastomers (LCEs), a class of multifunctional and complex soft materials.

The second direction considers the broader problem of constitutive model discovery. As we are exposed to an increasingly data-rich environment, we can obtain more information from experiments with full-field imaging and high-speed measurements,

which thus allows us to readdress the challenge of modeling complex materials. However, despite this increasing stream of data, established approaches struggle to take advantage of all the underlying fine-scale information contained in raw data to infer models. This thesis develops a new data-driven inversion technique that extracts models directly from raw experimental data.

Finally, once a constitutive model and some physics knowledge of the material are available, in the final part of the thesis, we consider how the experiments themselves can be optimally designed to maximize information gain and reduce uncertainty in the inferred model.

1.1 Experiments and modeling of complex materials: Liquid Crystal Elastomers (LCEs)

The first part of this thesis focuses on liquid crystal elastomers (LCEs) as an exemplar of complex, multifunctional materials. LCEs are lightly cross-linked polymer networks containing liquid crystalline mesogens, and their mechanical response is governed by the coupling between rubber elasticity, liquid crystalline order, and deformation [11–15]. This coupling gives rise to unusual behavior, including large spontaneous deformation, and rate- and temperature-dependent response, making LCEs promising for applications such as actuation, energy absorption, and impact mitigation [16–20]. However, despite this promise, the dynamic behavior of LCEs remains poorly understood. Impact absorption applications involve large deformation and high strain rates, yet much of the existing experimental characterization and model development has focused on quasistatic regimes.

This motivates the first part of this thesis, which develops experimental and modeling tools to study the large-strain, high-strain-rate behavior of LCEs. We begin in Chapter 2 by reviewing the relevant physics of LCEs and introducing the previously developed constitutive model that describes the mechanical response of a class of LCEs. This model provides theoretical ground for the subsequent experiments and model extensions. In Chapter 3, we develop a novel tensile drop tower apparatus that enables high strain-rate testing of soft materials. This experimental capability is necessary to access the high strain, high strain-rate deformation regime in LCEs, which is otherwise difficult to access using conventional testing setups. Using this setup, in Chapter 4, we characterize the high strain rate response of LCEs. Motivated by these observations, we develop a macroscopic constitutive model in Chapter 4 from first principles to include polymer viscoelasticity and rate-dependent effects.

In Chapter 5, we further investigate the coupled effects of temperature and strain rate, providing a broader experimental characterization of the unique soft behavior of LCEs over a range of thermomechanical conditions.

More broadly, this first portion of the thesis illustrates the traditional route to constitutive modeling of a complex material when substantial physical knowledge is available. In our particular system, LCEs have been experimentally studied since the 1990s and a large body of literature probing their unique behavior exists [10, 11, 21–26]. In addition, LCEs have microstructural mechanisms that have been well-studied [25, 27–29]. These experimental studies and microstructure knowledge motivate the development of various models for LCEs from first principles [28, 30–33]. As in LCEs, the modeling of complex materials with this traditional approach relies on substantial prior knowledge of the material. However, for many emerging material systems, this knowledge may not yet exist, and a different approach is needed. This motivates the second part of the thesis.

1.2 Data-driven model discovery for complex materials

The LCE studies highlight a broader challenge in mechanics: complex materials require high-fidelity constitutive models, but obtaining such models from first principles alone requires a largely well-established physics knowledge of the material. For most new and emerging materials, such as additively manufactured metals [9, 34], architected materials [1], biological tissues [8, 35], and energy-storage materials [2, 7], the governing mechanisms are often poorly understood. Thus, discovering constitutive relations from experimental data is a challenging inverse problem: we observe experimental data and seek to infer the constitutive law that best explains those observations.

Traditionally, development of constitutive models has relied on carefully designed mechanical tests that produce simple, homogeneous deformation states. Experimental setups such as uniaxial tension, compression, shear, or biaxial loading, along with the corresponding standardized testing specimens, have been established as standard procedures to characterize unknown materials. This approach is successful for many relatively simple materials (e.g., isotropic rubber materials [36, 37]) because it decouples complex responses, assigns some physical intuition to each parameter, and reduces the inverse problem to a simple fitting problem. However, it is a low-throughput approach since each experiment probes only a limited strain, and resulting models can be insufficient when used to predict responses in

real applications that often involve complex coupling and nonuniform deformation. Modern experimental mechanics offers the possibility of improving this traditional approach. Advances in imaging, optics, and rapid measurement diagnostics now allow us to obtain full-field data. The full field data is often in the form of images that contain information about the deformation of the specimen. This creates the potential to leverage complex specimen geometries that can generate heterogeneous strain fields to better inform the model within just one experiment. However, this also creates a more challenging inverse problem, since we seek to infer the constitutive relations, often represented by a set of unknown model parameters, that best explain those full-field measurements.

Current practice typically solves this difficult inverse problem in two steps. First, images are processed using image-matching methods (such as Digital Image Correlation or DIC) to infer displacement fields. Various image-matching algorithms exist for this DIC step [38–40]. Second, the inferred displacement fields are used in an analysis method, such as finite element model updating (FEMU) [41] or the virtual fields method (VFM) [42, 43], to identify material parameters. This is a well-established pipeline and has been used to study the behavior of complex material systems, from biological materials [44, 45], metal plasticity [46], polymers [47, 48], and architected materials and foams [48, 49], among many others.

Although powerful, there are important limitations to the current two-step approach. It is clear that this two-step workflow inherits all the problems associated with nested optimization, such as sensitivity mismatch and error magnification. But more important to note is that the displacement field obtained in the first step itself is not a direct measurement and is processed data. In addition, this first step is done without considering the known physics of the problem or of the material, which leads to information loss. Following this first inversion, when considering the second inversion, to ensure robust model identification from the inferred displacement, one often reverts back to using simple geometry and idealized experiments, instead of leveraging complex material states. This leads to sparse data. In summary, this sequential, two-step approach leads to loss of information, sparse data, and significant uncertainties in complex states of the material. This is not ideal, since complex states of the material are our regimes of interest, which contain the most amount of information to then be used to infer high-fidelity constitutive models.

The second part of this thesis develops a unified, alternative approach that directly integrates experiments, known physics, and model discovery together. In Chapter 7

of this thesis, we present the image-to-constitutive-model (I2C) framework, where constitutive models are determined directly from data in the form of raw images and forces, while already being subject to equilibrium, compatibility, and any known physics. This method is mathematically well-posed, i.e., the laws of physics regularize the ill-posedness of image-matching with no ad hoc filter, and is amenable to any general form of constitutive relations under large deformations, non-pristine test conditions, and complex loadings.

1.3 From model discovery to optimal experiments

The image-to-constitutive-model (I2C) inversion method discussed here allows us to exploit fine-scale information in the raw data that contains the physics of the materials to obtain higher-fidelity models. However, despite this ability, we note that the quality of the inferred model depends strongly on the experiments used to provide data. The resulting model has uncertainties if the experimental data is not informative enough to excite relevant material behavior. Therefore, we seek to leverage all current information about known physics to design the next optimal experiment—optimal in the sense of providing the most informative data to further refine models.

This motivates us to address the more fundamental problem in material characterization and experimental mechanics, which is the optimal design of experiments for constitutive model discovery. In our treatment so far, we addressed the inverse problem of model discovery from a deterministic point of view. Alternatively, this inverse problem can be reposed from a Bayesian perspective, where the model parameters are unknown random variables conditioned upon noisy observations and prior beliefs [50, 51]. According to Bayes’ rule, after data assimilation, the uncertain model parameters are refined and updated by combining prior information and a likelihood model given by the forward problem. Thus, Bayes’ rule gives the posterior, i.e., updated probability density function of the unknown model parameters, with the most probable parameters being the maximizer of this posterior. This Bayesian treatment, therefore, provides a quantitative way to compare possible experiments before seeing any data, enabling a quantitative optimal experimental design framework. Bayesian approach has been used for various applications [52–56] to guide optimal experiments.

We seek to design optimal experiments before seeing any data. In mechanics, experimental design can be performed in terms of choosing boundary conditions, loading

paths, sample geometry, or measurement locations [52, 54, 57–62]. In Chapter 8 of this thesis, we focus on designing complex-geometry specimens to maximize measured information about the material model. This choice is motivated by advances in experimental mechanics that allow full-field measurement. As discussed in the preceding subsection, when combined with full-field measurement, complex geometries generate heterogeneous deformations that contain rich information about the material states in a single test. We combine Bayesian optimal experimental design with topology optimization to design specimens optimized for model discovery. We demonstrate that topology-optimized specimens can outperform conventional standardized specimens by producing richer deformation fields that reduce parameter uncertainty more effectively.

1.4 Thesis vision and organization

In summary, the overall vision of this thesis is to propose an integrated approach to experimental mechanics and constitutive modeling. The remainder of the thesis is organized as follows. The first part, which focuses on a more traditional approach to material modeling, includes Chapters 2,3,4,5. Chapter 2 introduces liquid crystal elastomers, reviews their relevant mechanical behavior, and presents the previously developed constitutive framework used throughout the preceding chapters. Chapter 3 describes the design and validation of a tensile drop tower apparatus for high strain-rate testing of soft materials. Chapter 4 presents high-strain-rate experiments on LCEs and develops a complete constitutive model capable of describing the observed rate-dependent response across multiple orders of strain rates. Chapter 5 extends the experimental characterization to combined temperature-dependent and rate-dependent behavior.

We then turn to the second part of the thesis, which focuses on data-driven approaches to material modeling. We start in Chapter 6 by reviewing currently established inverse methods for material modeling and highlight their limitations. Chapter 7 introduces a novel data-driven inversion technique called the image-to-constitutive-model (I2C) method that infers model parameters directly from raw images and force data. Chapter 8 develops the optimal experimental design framework to design complex-geometry specimens that can provide more accurate models while reducing experimental costs. Finally, Chapter 9 summarizes the main contributions of the thesis and discusses future directions toward a more integrated experimental-inference loop for modeling of complex materials in mechanics.

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Part I

Conventional Method: Constitutive Modeling of Liquid Crystal Elastomers

Chapter 2

BACKGROUND: LIQUID CRYSTAL ELASTOMERS

In this chapter, we introduce and review liquid crystal elastomers as complex materials and their potential in engineering applications. We then delve into current experimental studies and constitutive models of these materials. The model developed in Chapter 4 of this thesis is an extension of a model for LCEs that has been previously developed [1], which is described in Section 2.3 of this chapter.

2.1 Liquid Crystal Elastomers as complex materials

Liquid crystal elastomers (LCEs) consist of stiff nematic mesogens incorporated into a network of polymer chains [2]. The mesogens have long-range orientational order in response to external fields, such as temperature or light. Depending on chemistry and synthesis, LCEs may be nematic, smectic, or cholesteric; they may be monodomain or polydomain; and they may be responsive to temperature, light, electric fields, magnetic fields, or chemical stimuli [2–6]. Of particular interest here are nematic elastomers, in which the mesogens are oriented randomly above a certain transition temperature, but develop orientational order below a transition temperature. Above this transition temperature, T_{ni} , the mesogens are disordered, and their overall configuration is isotropic. Meanwhile, below the transition temperature, the mesogens are uniformly oriented along a particular director, achieving the so-called nematic state. When synthesized, the polymer chains are cross-linked lightly to ensure that these nematic mesogens retain their ability to undergo a phase transition. Therefore, a phase transition from disordered to ordered state leads to significant spontaneous elongation along the director. This interplay between temperature, nematic order, and deformation makes LCE a promising material for engineering applications: from actuation applications [7–9], to mechanical dissipation [10–12], and energy absorption applications [12, 13].

Various types of LCEs exist depending on their synthesis history. Depending on whether the mesogens are aligned during synthesis, they can form a monodomain or a polydomain structure in the nematic phase. In monodomain LCEs, all the mesogens align largely along a global director orientation. Meanwhile, polydomain LCEs (PLCEs) exhibit microstructures composed of multiple domains, each having its own local director, and the mesogens in each domain align along that local director

(Figure 2.1a). One can think of PLCEs as being analogous to a polycrystalline material. Importantly, since PLCEs can be made without requiring any alignment, they are scalable and easy to manufacture, and are therefore desirable as engineering materials [4]. There are two main types of PLCEs: isotropic-genesis PLCEs (I-PLCEs) and nematic-genesis PLCEs (N-PLCEs). I-PLCEs are made via cross-linking in the high-symmetry isotropic phase before cooling down to the nematic phase, whereas N-PLCEs are cross-linked in the nematic phase. This different synthesis gives rise to their unique soft behavior [14], which is discussed next.

One of the most important features of LCEs is their soft behavior, where they can undergo large stretches under low, constant stress [2, 15]. When a monodomain LCE is uniaxially stretched in the direction perpendicular to its director orientation, the director rotates to align towards the stretching direction [15]. This ability of the mesogens to reorient relative to the polymer matrix gives rise to a stress plateau over a large range of stretch, a phenomenon called “soft elasticity,” which can be seen in Figure 2.1b. However, the director orientation is often constrained by imposed boundary conditions (for example, no shear condition during uniaxial extension). Hence, strain-induced microstructures, such as stripe domains, arise to induce the soft deformation yet effectively satisfying compatibility and any imposed boundary conditions [16]. In PLCEs, in addition to any strain-induced microstructure, there is domain microstructure from synthesis. The overall reorientation of microstructure under uniaxial loading is referred to as the polydomain-to-monodomain transition (PMT) [17, 18]. It was soon recognized that while both N-PLCEs and I-PLCEs undergo this PMT transition, only I-PLCEs display the associated soft behavior [14]. Biggins *et al.* attributed this softness unique to I-PLCEs because they are cross-linked in the high-temperature, high-symmetry cross-linking state [19]. As I-PLCEs are cooled down to the nematic phase, any director orientation can be picked arbitrarily, meaning that there exist many low-energy states. Deformations around these ground states and textured deformations constructed from such ground states are soft. Meanwhile, N-PLCEs are cross-linked in the symmetry-broken state and have memory of the directors in this state, resulting in a macroscopically hard behavior. It has further been established that the soft behavior of I-PLCEs is a manifestation of their in-plane liquid-like behavior, i.e., they are unable to sustain shear stresses in the plane [20]. Meaning, when subjected to biaxial loading with differing imposed strains, the two true stresses in I-PLCEs remain equal for a range of strains.

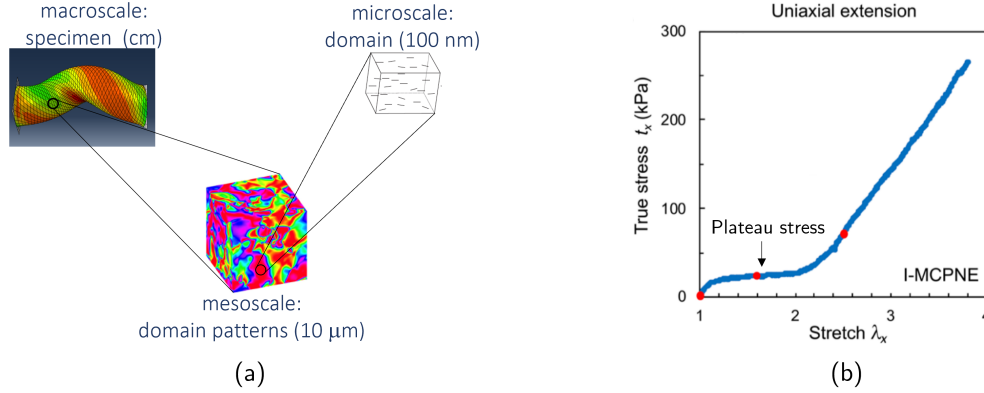


Figure 2.1: (a) Multiple scales in polydomain LCE. (b) Soft behavior of LCE: large stretch at a low plateau stress (adapted from [20]).

This unique behavior of I-PLCEs, as well as ease of synthesis (not needing any global alignment), makes them promising as engineering materials. In addition, they have potential in mechanical damping applications. Both [10] and [11] observed a significant loss behavior in I-PLCE, where the loss tangent is large over a wide range of temperatures and frequencies (75 – 110°C, 1 – 100 Hz) due to internal relaxation of the domains and viscoelasticity of the chains. I-PLCEs are also promising for energy absorption applications. In particular, Jeon *et al.* showed that I-PLCE-based architected material displays extremely large energy absorption density (5MJ/m³) at a high strain rate of 600/s [13]. Given these advantages, we focus on I-PLCEs as engineering materials for the scope of the thesis.

To effectively design I-PLCEs for these damping and impact absorption applications, the dynamics of their domain evolution need to be understood. We discuss current experiments in Section 2.2 and constitutive models in Section 2.3.

2.2 Experiments on Polydomain Liquid Crystal Elastomers

The dynamics of PLCEs are important to consider when using them for energy dissipation applications. Numerous studies have been conducted over the past decades to study the time-dependent behavior of LCEs at various strain rates, frequencies, and temperatures. We note, however, that it is challenging to directly compare the results of different experimental studies due to the different types of PLCEs that were studied (either I-PLCEs or N-PLCEs, side-chain or main-chain, and siloxane-based or acrylate-based). Additionally, they can be synthesized with different methods [4–6], which can lead to different behavior.

The effect of strain rates (10^{-7} to 10^{-3} s^{-1}) on the stress relaxation of side-chain I-PLCEs was first studied by Clarke *et al.* for siloxane-based [21] and by Hotta *et al.* for acrylate-based [22]. Both studies observed that at a certain threshold stress, the mesogens/domains start to reorient towards the direction of extension, resulting in soft elasticity and a stress plateau. The stress response during PMT (polydomain to monodomain transition) is most interesting, where both found that there exists a dual regime of relaxation: short-time and long-time relaxation. Clarke *et al.* attributed the fast relaxation to global domain reorientation and the following slow relaxation to the increase in mechanical resistance as more domains reoriented. Hotta *et al.* also concluded that the fast relaxation occurs due to global domain reorientation. However, they attributed slow relaxation to the relaxation of the polymer network. Both studies observed a transition time of 1000 s. Supporting these findings, Azoug *et al.* [23] observed that both the Young's modulus and plateau stress at PMT are rate independent from (10^{-8} – 10^{-3} s^{-1}) but increase significantly at strain rates higher than 10^{-3} s^{-1} . All three studies indicate that there exists a characteristic time scale associated with global domain rotations. Slower loading than the characteristic time scale would result in weak rate dependency of the stress response.

This time-dependent mechanical behavior is also affected by cross-linking density. As cross-link density is increased, there is a proportional increase in the elastic modulus and a decrease in the recovery time [11, 24]. In addition, the authors of [24] observed that the shape recovery of monodomain LCEs occurs much faster than in polydomain LCEs. This indicates that the relaxation of domains in PLCEs results in slower stress relaxation. Among PLCEs, the shape recovery of N-PLCEs is faster than that of I-PLCEs due to their stronger director correlation from the memory of the cross-linking state.

The effect of domain alignment on the rate-dependent (0.001 - 1 s^{-1}) stress behavior of main-chain acrylate I-PLCE has been investigated by Linares *et al.* [25]. To do so, the networks were pre-stretched to 0.9 strain (corresponding to the end of PMT), and then loaded parallel or perpendicular to the pre-stretch. I-PLCEs loaded in the perpendicular direction exhibit a wider soft stress response since the domain can rotate towards the loading direction. Meanwhile, I-PLCEs loaded in the parallel direction display minimal soft stress response. They also display large hysteresis, suggesting that the polymer viscosity plays a role in the stress response. These results indicate that there is an inherent coupling between liquid crystallinity and network viscosity that affects the mechanical behavior, especially at a dynamic rate.

2.3 Constitutive Model of Liquid Crystal Elastomers

In this section, we describe relevant constitutive models of LCEs. We briefly discuss the classical theory, which is very successful in capturing the quasi-static behavior of LCEs [26]. We then turn to describe a constitutive model recently developed for I-PLCEs that incorporates their multiscale physics and the evolution of microstructures [1].

The classical theory of LCEs was developed by Blandon, Terentjev, and Warner by extending the modeling approach of neo-Hookean elastomers to account for anisotropy [26].

$$W_{\text{BTW}} = \frac{1}{2}\mu \left(\text{tr}(F^T l^{-1} F) - 3 \right), \text{ where} \quad (2.1)$$

$$l = r^{-1/3}((r-1)\hat{n} \otimes \hat{n} + I), \text{ and } r = \frac{1+2Q(T)}{1-Q(T)}. \quad (2.2)$$

Note that the l -tensor here captures anisotropy due to a preferential direction of the mesogens within the domain to align along the director, $\hat{n}$. The degree of this alignment is influenced by the temperature and is captured via $Q(T) \in [0, 1)$. When $Q(T) = 0$, $r = 1$ and the LCE is isotropic; when $r > 1$, the LCE is in a nematic state. This anisotropy results in a spontaneous stretch along the director upon cooling to the nematic state. The resulting nematic energy density is neo-Hookean with respect to this spontaneous deformation state, which is the new ground state. It was recognized that this form of energy is not quasi-convex, leading to fine-scale microstructure formations. To account for such microstructures, Desimone and Dolzmann performed relaxation of the energy [27], which is explicitly computed by minimizing W_{BTW} over the director $\hat{n}$ to obtain

$$W(F) = \min_{\hat{n} \text{ s.t. } |\hat{n}|=1} W_{\text{BTW}}(F, \hat{n}) = \begin{cases} \frac{\mu}{2} \left(r^{1/3} \left(\frac{s^2}{r} + \frac{t^2}{s^2} + \frac{1}{t^2} \right) - 3 \right) & \det F = 1 \\ \infty & \text{else} \end{cases}, \quad (2.3)$$

where s, t are the largest singular values of $F, \text{cof } F$. The relaxed energy is the effective energy of LCE after it has formed the most optimal domain textures. This classical theory successfully captures the soft behavior and stripe domain formations in monodomain LCEs [28]. It is, however, limited to the ideal behavior and neglects impurities such as cross-links and network defects. There have been efforts to include such non-ideality, although relaxation of the non-ideal energy has only been computed numerically [29, 30]. This theory has also successfully described the quasi-static behavior of monodomain LCEs.

Due to their ease of synthesis and inherent kinetics, PLCEs, rather than monodomain LCEs, are more promising for engineering applications [4, 12, 31]. Simulations that consider microstructure evolution on PLCEs to extract macroscopic behavior have been done using parallel computing, though these simulations are very expensive [32]. Additionally, in most design settings, the exact microstructure evolution of our PLCEs is not of priority. For these reasons, recently, an engineering-scale constitutive model for I-PLCEs has been developed [1].

The remainder of this section closely follows the derivation in [1]. We begin by considering the multiscale structure of I-PLCEs (Figure 2.1a). In a specimen of I-PLCE, there exist domain patterns at the mesoscale ($10\mu\text{m}$), with each domain pattern consisting of distinct domains roughly 10-100 nanometers in size. Each domain is composed of mesogens (2 nanometers), all aligning uniformly along a local director. To incorporate such a multiscale setting into the constitutive model, a representative volume element (RVE) is selected at the domain pattern scale. To describe the behavior of these domain patterns, we first account for the orientation of all mesogens in the RVE. We derive the orientation order tensor (S) by averaging over all mesogen orientations (u)

$$S = \left\langle \left\langle u \otimes u - \frac{1}{3}I \right\rangle_{\text{mesogens in one domain}} \right\rangle_{\text{domains in RVE}} = Q \left\langle n \otimes n - \frac{1}{3}I \right\rangle_{\text{domains in RVE}}, \quad (2.4)$$

$$S' = \begin{pmatrix} S_m & 0 & 0 \\ 0 & \frac{1}{2}(-S_m + X) & 0 \\ 0 & 0 & -\frac{1}{2}(S_m + X) \end{pmatrix}, \quad (2.5)$$

where n is the director of each domain, Q is a scalar measuring mesogen alignments in one domain, and S' is S rewritten in the principal frame. From (2.5), S_m measures the degree of alignment of all mesogens along the principal direction in an RVE, while X measures the degree of planarity of all mesogens in the RVE.

We then consider a spontaneous deformation metric associated with the domain patterns in the RVE. Drawing inspiration from the monodomain-scale in the microscale, the authors in [1] postulated that the collection of domains at the mesoscale results in a spontaneous deformation G (assuming incompressibility),

$$G = PG_0P^T, \quad G_0 = \begin{bmatrix} \Lambda^2 & 0 & 0 \\ 0 & \Delta^2/\Lambda^2 & 0 \\ 0 & 0 & 1/\Delta^2 \end{bmatrix}, \quad (2.6)$$

with $\Lambda \geq \Delta/\Lambda \geq \Delta$, $P \in \text{SO}(3)$, corresponding to reorientation of domain patterns. We note that the G -tensor here is analogous to the microscopic, modomain-scale l -tensor in (2.1). This spontaneous deformation G is related to the domain pattern description above, where Λ (principal stretch) corresponds to S_m , and Δ (principal areal stretch) corresponds to $S_m + X$. We obtain the upper limit of this spontaneous deformation to be the state in which all the domains align in one direction, forming a monodomain. From the classical theory of monodomain LCE, the monodomain limit is attained when $\Lambda \leq r^{1/3}$ and $\Delta \leq r^{1/6}$. It follows that Λ and Δ are limited to the set $\mathcal{A}_G$

$$(\Lambda, \Delta) \in \mathcal{A}_G = \{(\Lambda, \Delta) : \Delta \leq r^{1/6}, \Delta \leq \Lambda^2, \Delta \geq \sqrt{\Lambda}\}. \quad (2.7)$$

Finally, the free energy is postulated to be of the form (limited to the isothermal case)

$$\mathcal{F}(F^T G^{-1} F) = \mathcal{F}_e(F^T G^{-1} F) + \mathcal{F}_r(\Lambda, \Delta), \quad (2.8)$$

where F is the deformation gradient of the domain pattern with respect to the isotropic state. The elastic energy $\mathcal{F}_e$ is taken with respect to the spontaneous deformation state, which is the ground state, and $\mathcal{F}_r$ captures any non-idealities. Picking a generalized Mooney-Rivlin energy as the elastic energy,

$$\mathcal{F}_e(F^T G^{-1} F) = \Phi(I_1(F^T G^{-1} F), I_2(F^T G^{-1} F)) \quad (2.9)$$

$$= \sum_{i=1}^I C_i(I_1^{n_i} - 3^{n_i}) + \sum_{j=1}^J D_j(I_2^{m_j} - 3^{m_j}). \quad (2.10)$$

The above Mooney-Rivlin energy is not quasi-convex. This lack of quasi-convexity leads to the formation of fine-scale microstructure, which arises as a means to minimize the energy. To account for such microstructure formations, relaxation of the free energy is computed by Agostiniani and Desimone [33]. In our model, the relaxed energy (W_{AD}) was proven to be achieved if P, Λ, Δ can evolve instantaneously to minimize free energy (2.8)

$$W_{AD} = \min_{\substack{P \in \text{SO}(3) \\ (\Lambda, \Delta) \in \mathcal{A}_G}} \mathcal{F}(F^T P G_0^{-1} P^T F). \quad (2.11)$$

Assuming instantaneous orientation of the domain patterns,

$$\min_{P \in \text{SO}(3)} \mathcal{F}(F^T P G_0^{-1} P^T F) = \Phi\left(\frac{\lambda^2}{\Lambda^2} + \frac{\delta^2 \Lambda^2}{\lambda^2 \Delta^2} + \frac{\Delta^2}{\delta^2}, \frac{\Lambda^2}{\lambda^2} + \frac{\lambda^2 \Delta^2}{\delta^2 \Lambda^2} + \frac{\delta^2}{\Lambda^2}\right), \quad (2.12)$$

where λ and δ are the principal stretch and principal areal stretch of the deformation gradient, respectively. The energy density per unit reference volume is then postulated to be of the form

$$W(F, \Lambda, \Delta) = \Phi(F, \Lambda, \Delta) + W_r, \quad \text{with} \quad W_r(\Delta) = C \frac{\Delta - 1}{(r^{1/6} - \Delta)^k}. \quad (2.13)$$

The residual energy, W_r above, takes into consideration the domain incompatibility as the monodomain state is approached. The evolution of the internal variables (Λ, Δ) is obtained by assuming a quadratic form of the dissipation potential,

$$D(\dot{\Lambda}, \dot{\Delta}) = \frac{\alpha_\Lambda}{2} |\dot{\Lambda}|^2 + \frac{\alpha_\Delta}{2} |\dot{\Delta}|^2, \quad (2.14)$$

which satisfies the dissipation inequality and is thermodynamically consistent. Stationarity of the energy and dissipation potential leads to the evolution law for the state or internal variables,

$$\alpha_\Lambda \dot{\Lambda} = -\frac{\partial W}{\partial \Lambda}, \quad \alpha_\Delta \dot{\Delta} = -\frac{\partial W}{\partial \Delta}. \quad (2.15)$$

The model parameters C_1 or μ , C , k , α_Δ can be fitted against experiments. Further, in rate-independent, quasi-static tests, one often assumes that $\alpha_\Lambda = \alpha_\Delta/100$ due to fast rotation of the domain patterns in plane rather than out of plane. This constitutive model shows excellent agreement with experiments on I-PLCE under complex, quasi-static multiaxial loading conditions [20], Figure 2.2 (adapted from [1]).

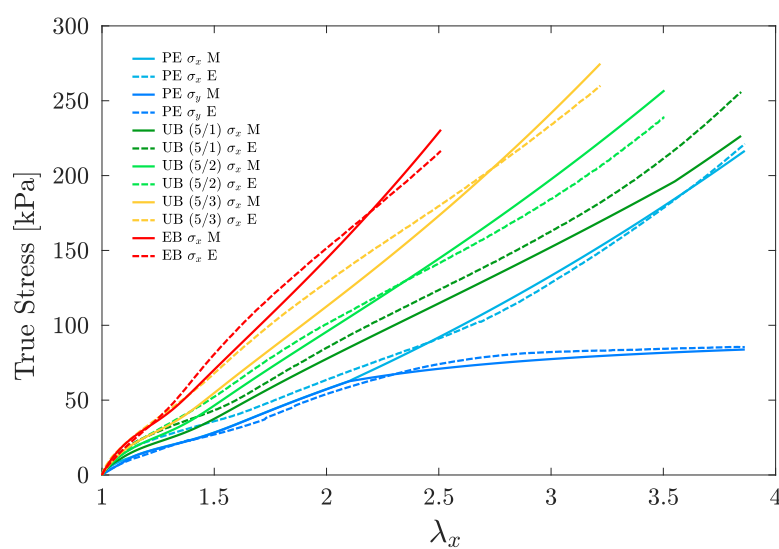


Figure 2.2: Comparison between the model (solid curves) and experimental observation (dashed curves) under quasi-static, multiaxial loading conditions. PE refers to planar extension, UB is unequal biaxial, EB is equal biaxial loading. Adapted from [1].

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Chapter 3

A NEW DROP WEIGHT TENSILE TESTING SYSTEM FOR SOFT MATTER AT INTERMEDIATE STRAIN RATES

J. C. Nieto-Fuentes, A. Wihardja, P. Stovall, T. Wilson, K. Bhattacharya, and D. Rittel, “A new drop weight tensile testing system for soft matter at intermediate strain rates”, *European Journal of Mechanics-A/Solids* **111**, 105507 (2025).

Contributions: A.W participated in developing experimental methodology, performed the experiments, produced and processed and analyzed the experimental data, developed and performed the simulation methodology, and wrote the manuscript.

Abstract

This paper presents a novel and versatile tensile testing system based on the drop weight technique, specifically designed for materials that can undergo significant tensile deformation, such as elastomers. The core apparatus comprises of a hanging slender bar, from which a steel sleeve (referred to as the striker) is released under controlled conditions. Accelerated in free fall, the striker impacts a stationary plate, initially held in place by a mechanical detent. The specimen, secured by a gripping system between the hanging bar and the stationary support, undergoes controlled stretching at a nearly constant velocity upon the release of the detent triggered by the striker’s impact. Full-field strain measurement is obtained using a high-speed camera in conjunction with digital image correlation. Additionally, strategically located piezoresistive force sensors enable real-time force measurements. By achieving strain rates ranging from 100 s^{-1} to 500 s^{-1} , this system addresses a notable gap in the literature concerning intermediate strain rate testing for soft materials.

3.1 Introduction

Tensile testing is a fundamental method used to evaluate the mechanical properties of materials, offering insights into strength, ductility, or elastic properties. It serves as a critical tool for researchers, engineers, and industries seeking to understand,

improve, and ensure the performance and reliability of materials in various applications. The most common experimental device to characterize the tensile mechanical behavior of materials is the universal testing machine, either hydraulically or electromechanically driven. These apparatuses have proven for many years to be the workhorse of quasi-static experimentation, specifically in the range of strain rates from 10^{-4} s^{-1} up to 1 s^{-1} . At higher strain rates, ($\geq 10^3 \text{ s}^{-1}$ or “dynamic”), the Split Hopkinson Pressure Bar (SHPB), or Kolsky bar, modified for tension [1–4] or variations of the expanding ring testing technique [5–9] are extensively used to test a whole range of metallic or composite materials under tensile loading.

These techniques leave a gap in the realm of tensile testing at intermediate strain rates, from say ten to a few hundred per second. Quasistatic universal testing machines can not exceed tens per second, or do so at the expense of significant system vibrations, potentially affecting the load cell measurements (system ringing) thereby compromising the reliability of the test. Similarly, traditional Hopkinson bar systems cannot reach rates below 10^3 s^{-1} . This constraint arises from the duration of the loading pulse in a typical SHPB experiment, which is dependent on the length of the striker (specifically the pulse is twice the elastic wave travel time through the bar). One might consider increasing the loading time, but it necessitates a proportional increase in the length of the bars. This has been accomplished in a few facilities, like the ELSA Hoplab [10] where a 200-m long Hopkinson bar can generate pulses up to 40 ms; however, this is not possible in a typical laboratory environment. An alternative to using the traditional three-bar Kolsky apparatus is direct impact testing techniques. Gilat and Matarka [11] employed a hydraulic actuator and a 40-meter-long transmitter bar to perform direct compression impact tests on epoxy samples. Full-field strain measurements were obtained using 3D Digital Image Correlation (3D-DIC), and force signals were captured by a single strain gage placed near the specimen-bar interface. They reported smooth signals without oscillations, with pulse durations of up to 4 ms. The same apparatus was adapted for tensile testing of stainless steel 316L, reaching strain rates of approximately 200 s^{-1} over a duration of about 2 ms [12].

The SHPB also poses a number of challenges for soft materials such as elastomers: the large deformations these materials experience under tensile loading, the ability to reach stress equilibrium necessary to obtain the material response [13, 14], the detection of weak signals to measure forces due to impedance mismatch [15], or the application of a near constant strain rate during the test. A number of works in the

literature have overcome these difficulties and successfully tested soft materials. For example, Nie *et al.* [3] tested EPDM rubber as a model material using a modified Split Hopkinson Tension Bar (SHTB), where cylindrical hollow bars were used instead of solid cross-section bars to minimize inertia effects, and force equilibrium was monitored by means of quartz crystal force transducers. Indeed, piezoelectric force transducers are widely used to increase the sensitivity of SHPB experiments when testing low-impedance and low-strength materials [16]. Another, more recent example is provided by Jakkula *et al.* [17], who used a 40-meter-long bar in combination with a heavy striker to generate extended pulses (16 ms) straining low-impedance materials at intermediate strain rates. The test results were compared to those from a *dynamic* servo-hydraulic machine and a setup where a load cell was directly connected to the bar end, demonstrating that the direct impact setup produced cleaner results with reduced inherent noise and system ringing. Chen *et al.* [18] used a Kolsky bar with a very long striker to test soft materials under compression. The specimen was deformed by summing multiple pulses, each generated as the stress wave traversed the length of the incident bar. Although pulse overlap was not ideal, it did not significantly affect force measurements, as piezoelectric force transducers were used instead of strain gages. Upadhyay *et al.* [19] modified the traditional SHPB by incorporating a custom-designed fixture that transforms the initial compressive wave in the incident bar into tensile loading on the specimen, using an innovative gripping method for the specimen to prevent slipping. However, despite these advancements, testing soft materials remains a challenge for SHPB-based techniques.

Other techniques for conducting tensile testing at intermediate strain rates include combining a drop tower with a Kolsky bar, as in the apparatus developed by Song *et al.* [20]. This setup is able to generate an impact pulse with a duration suitable for testing metallic materials, with a pulse duration of approximately 1.3 ms without wave overlap, though it is too short for testing soft materials. Perogamvros *et al.* [21] adapted a conventional drop tower machine into an apparatus consisting of one rigid and one movable frame. This setup was specifically designed for testing large metallic samples, where the system's high inertia would not be a drawback. However, when testing soft materials like elastomers, the heavy weight of the movable frame becomes a setback. Furthermore, micro-tensile [22] and micro-compressive [23] bench-top devices have also been developed, that use electro-mechanical actuators as the driving force. These setups have demonstrated the capability to test small-scale biological and soft materials at the intermediate range of strain rates. Readers

seeking a comprehensive review of experimental techniques for studying the high-rate mechanics of polymers should refer to [24].

In this work, we present a new and versatile, yet simple, tensile testing system based on the drop weight technique, capable of testing materials that experience large deformations under tension (soft materials like elastomers) at strain rates in the range $100 \text{ s}^{-1} - 500 \text{ s}^{-1}$. The device consists of a slender circular steel bar, on which a piece of steel tube (sleeve) is allowed to slide by gravity in free fall. The specimen is clamped between one end of the bar and at a standing plate which is being held by mechanical detents. Upon impact of the steel sleeve on the standing plate, the standing plate is released, causing the specimen to stretch uniaxially. The transient events and full-field strain measurements are recorded by means of a high-speed camera coupled with digital image correlation. Piezoresistive force sensors located on top of the bar allow for real-time force data acquisition. We present the system in Section 3.2, and demonstrate it with natural rubber in Section 3.3.

3.2 Experimental Method

Tensile drop tower design

The design of the tensile drop tower apparatus is based in the drop weight technique, a method commonly used for conducting compression tests at intermediate strain rates. In this study, we have adapted the same principle of a gravity-driven falling weight to conduct impact tests under tensile loading conditions. The apparatus can be basically divided into three parts: the top ensemble, the striker (driving force mechanism), and the bottom ensemble. The entirety of the setup is depicted in Figure 3.1, with details in Figure 3.2.

The top ensemble of the tensile drop tower, shown in Figure 3.2(a) consists of a stainless-steel slender bar with 31.75 mm of external diameter and a length of 1.8 m, suspended vertically from a steel plate. This plate is affixed to the laboratory ceiling via a series of rods, creating a 18 cm clearance between the plate and the ceiling. A precisely machined aperture in the plate allows the steel bar to pass through freely, enabling unimpeded vertical motion. To restrict vertical displacement (u_{yy}) during the experiment, an open swivel joint is affixed to the end of the bar. On the opposite end of the stainless-steel bar, a gripping system comprised of two plates with knurled surfaces secures one side of the dog-bone specimen in place. Knurling of the grip plates surfaces is essential to prevent slippage of the specimen's shoulder during the experiment. Similarly, the other end of the specimen is clamped by a comparable grip

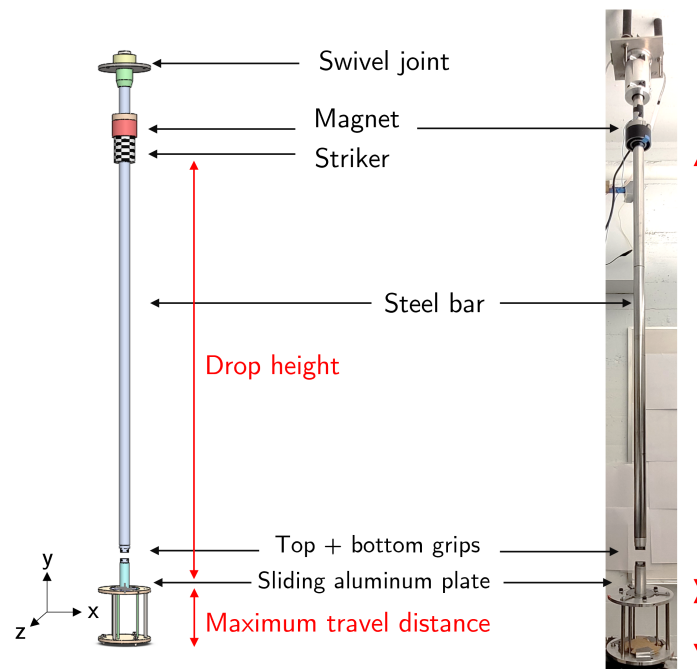


Figure 3.1: Schematic drawing (left) and image (right) of the drop weight tensile testing system. Note that the striker is not shown in the right image.

system, situated on the lower base (bottom ensemble) of the drop tower apparatus. To further minimize slippage of the sample, additional measures could be implemented, such as increasing the size of the sample's shoulders, enlarging the contact area with the grips, or inserting sandpaper between the specimen's shoulder and the grip plates to increase friction. Another potential solution is to glue the shoulders of the sample between aluminum plates, which can then be attached to the grips.

As depicted in Figure 3.2(b), the bottom ensemble of the drop tower apparatus primarily comprises the following components: two aluminum plates (identified as items number 1 and 2) interconnected by three slender bars, a sliding aluminum plate (item 3) equipped with a restraint mechanism, an extension bar (item 4) housing the specimen grip system (item 5), and a crash pad (item 6) designed to absorb the impact from the striker. Item 1 can be readily affixed to either the laboratory floor or a robust bench, depending on the specific length requirements of the drop tower apparatus. In the case of this study, the aluminum plate is securely attached to a heavy-duty bench to ensure stability and reliability during experimentation. The aluminum plate designated as item 2 serves to secure the sliding plate (item 3) in position through the use of three ball-nose spring plungers. These locking mechanisms

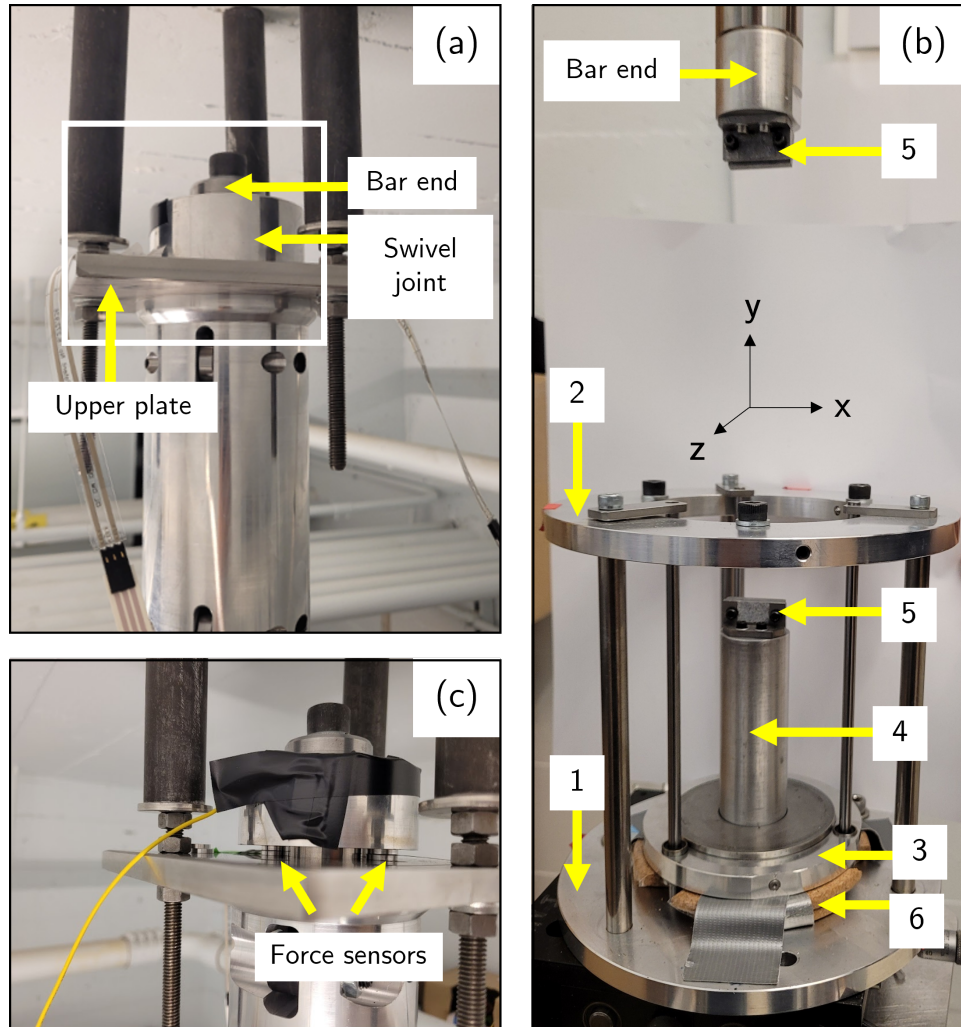


Figure 3.2: Details of the drop tower apparatus. (a) Top ensemble. (b) Bottom ensemble. (c) Force sensors placed in the top assembly of the drop tower. Note that the sliding plate (item 3) in this figure is in the lower position, and not being held by the mechanical detents located in the top aluminum plate (item 2).

facilitate a stable alignment of the sliding plate before impact. Moreover, the detent mechanism's quick release, triggered by the impact force of the striker, minimizes kinetic energy loss, thereby optimizing the efficiency of the testing process. It is important to note that the release time of the detent mechanism does not play a critical role in the data acquisition process. Instead, the force signal recording is triggered when the stress wave, traveling through the bar, reaches the force sensors positioned at the top of the setup - see Figure 3.2(c) and Section 3.2 for more details. Smooth motion of the sliding plate after-impact is facilitated by three linear shafts, in conjunction with linear bearings. This configuration ensures a seamless movement, contributing to the uniaxial stretch of the specimen during the test. The

extension bar (item 4), anchored to the sliding plate at one end and supporting the specimen's grip system at the other, measures 90 mm in length, mirroring the length of the striker to prevent obstruction of the specimen from the camera's field of view during testing. Maintaining coaxial alignment between the extension bar (that is, the bottom ensemble) and the hanging vertical bar (top ensemble) is crucial to minimize out-of-plane displacements of the specimen during testing. This coaxial alignment prior to the test is achieved using a transparent plastic tube with an internal diameter equal to the external diameter of the bar (32 mm). The extension bar from the bottom ensemble is pulled down and then inserted into the plastic tube for alignment. Once the alignment is complete, the tube is removed. Additionally, capturing a static image with the camera aids in reducing misalignment along the Y and Z axes.

The driving force for the experiment is provided by a steel sleeve, referred to as the striker, which features an internal diameter of 32 mm (coinciding to that of the bar, with a specific tolerance) and a thickness of 16 mm. The striker is securely held in place by a ring-shaped electromagnet, adjustable along the length of the hanging steel bar to meet the desired impact velocity for experimental needs. The release mechanism for the striker involves a normally-closed push button switch, offering a safe and controlled launch mechanism for the experiment. The sleeve has an internal Teflon (PTFE) bushing to facilitate a smooth sliding along the bar. The use of PTFE minimizes friction, allowing for effortless motion of the sleeve during the striker acceleration stage.

Full field strain characterization

The experiments are captured using a high-speed full-field imaging technique. To achieve this, a Photron FASTCAM NOVA S12 high-speed camera equipped with a 100 mm Tokina AT-X Pro lens is synchronized with the striker release mechanism, enabling the collection of 5437 frames at 10000 frames per second with a resolution of 1024×1024 pixels. Throughout the impact duration, a continuous light source (MultiLed LT) is used. The high-speed imaging setup is complemented by in situ 2D Digital Image Correlation (DIC), allowing for comprehensive analysis of displacement fields within the specimen. 2D DIC software (Vic2D, Correlated Solutions, Columbia, SC) was used to determine the full-field displacement of the specimen and the bottom plate. A black speckle pattern over a white background was used for correlation with a subset size of approximately 21×21 pixels and a step size of 1 pixel. The same parameters were used to determine the vertical velocity (v_{yy}) of the bottom plate, with a random speckle pattern applied to the bottom clamp

holding the specimen.

Noise floor strain in DIC measurement is quantified by taking a series of ten still images of a control sample. Any displacement measured from these images via DIC is indicative of noise inherent to the imaging system, introducing error in the DIC measurement. Under no deformation, the mean and standard deviation values of the full-field vertical displacement (u_{yy}) and strain (ϵ_{yy}) are shown in Table 3.1. The highest recorded displacement was 1.3×10^{-4} mm, with a corresponding strain of 0.004%. Across a selected set of control images, the intended zero strain was measured as being on the order of 10^{-5} . False measurements arising from the imaging setup were determined to be negligible.

Noise in DIC measurements	
Strain (ϵ_{yy}), mean	4×10^{-5}
Strain (ϵ_{yy}), standard deviation	6×10^{-5}
Vertical displacement (u_{yy}), mean	0.00009 mm
Vertical displacement (u_{yy}), SD	0.00008 mm

Table 3.1: Quantified DIC uncertainties from ten still images of the control sample.

Force measurement

Whereas strain gages are the most popular means to measure strains (quasi-static or even transient) in bars, their use is limited in the present setup by the fact that due to the pulse duration (several milliseconds), the “traditional” incident and reflected signals are due to overlap considerably, therefore precluding the straightforward determination of the applied load. To overcome this limitation, we opted for thin-film piezoresistive force (pressure) sensors in our design. These sensors offer rapid response times and high sensitivity, making them well-suited for capturing transient events typical of the intermediate strain rates encountered in drop tower testing. Specifically, we employed two Tekscan FlexiForceTMA201 sensors, capable of measuring up (but not really limited) to 445 N in the standard range.

To ensure accurate force measurements, the sensors underwent conditioning and calibration procedures at room temperature before installation in the drop tower apparatus. Calibration involved the use of an MTS servohydraulic machine (Model 358.10) to apply loads within the expected force range, with the process repeated multiple times. The resulting calibration curve, shown in Figure 3.3, provides the relationship between the sensors’ electrical output and actual force units. It is important to note that the sensor’s sensing area measures 9.53 mm in diameter.

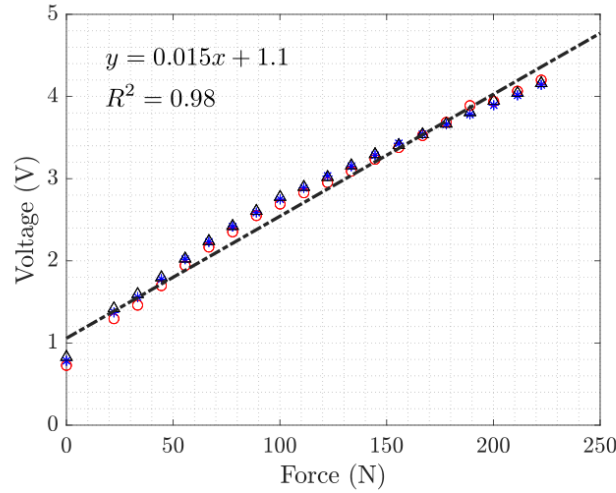


Figure 3.3: Calibration curve of the FlexiForce sensors used to measure force in drop tower experiments.

To guarantee that the load is solely supported by this sensing area, the sensor is sandwiched between two thin aluminum discs of a diameter slightly smaller than the sensing area. This configuration ensures that the force applied is accurately detected by the sensor without interference from surrounding areas. We would like to point out that a dynamic calibration of the force sensors can also be performed, as demonstrated in the work of Richler and Rittel [13]. In their study, they used a Split Hopkinson Pressure Bar setup, placing a force sensor between the incident and transmitter bars. The force sensor's readings were compared to the forces calculated using strain gage measurements, with a good correlation observed for frequencies up to 150 kHz. In our case, given the strain rates considered and the fast response time of our force sensors (approximately $5 \mu\text{s}$), we determined that a dynamic calibration was not necessary.

After calibration, the force sensors are positioned within the drop tower apparatus. Specifically, we have placed two diametrically opposed sensors, each sandwiched between the swivel joint on one end of the bar and the upper supporting plate, within the top assembly of the drop tower. A detailed view of this arrangement is provided in Figure 3.2(c). After the striker is launched, once the stress wave reaches the force sensors, diagnostics are triggered at 200 MHz using a Siglent SDS 1202X-E digital oscilloscope to start recording force data.

Noise-floor oscillations are present in the force measurements, introducing some uncertainty across all our experiments. We attribute these oscillations to several

factors, particularly the natural frequency of the suspended 1.8-meter stainless-steel bar. It is well known that the natural frequency of the system is a critical factor in high-rate testing [25]. However, we chose not to include any damping mechanism to reduce these oscillations, as doing so could potentially slow the rising rate of the stress pulse, thereby diminishing the dynamic nature of the experiment. Additionally, electrical interference in the laboratory may further amplify these oscillations in the force measurements. To quantify the uncertainty, we measured the magnitude of the oscillations under no-load conditions. On average, force oscillations of 7 N were recorded, and these oscillations persisted throughout the entire testing duration.

3.3 Demonstration

Drop tower tests were carried out on six different samples of natural rubber (McMaster-Carr, 87145k411). Each sample is cut to be 30 mm $\times$ 10 mm and 1.5 mm in thickness. In each test, the striker was dropped from a height of 0.75 m above the specimen, that is clamped between the sliding plate and the stainless-steel bar, resulting in an impact velocity $v_{\text{impact}} = \sqrt{2gh} \approx 3.8$ m/s.

Figure 3.4 collects the observations of one particular test. Snapshots of the deformation, and the resulting strain field across the specimen as captured by DIC, are shown in Figure 3.4(a). There is no noticeable cracking or removal of the speckles during DIC, and a good correlation is maintained throughout the duration of impact, despite the large amount of strain produced in the specimen. Further, the strain field is uniform across the sample, and such uniformity is present across all samples tested. There is a significant extension of the specimen, but there is also some lateral (u_{xx}) motion of the bottom clamp due to the small tolerance between the striker and the sliding plate that holds the specimen. By adding a speckle pattern to the bottom clamp and using DIC, the velocity and displacement history of the bottom clamp is obtained; this is shown in Figure 3.4(b). The vertical velocity v_{yy} of 3.7 m/s at impact is consistent with the estimated nominal impact velocity. There is an initial short lateral displacement u_{xx} (1.5 mm at its peak), but this rapidly diminishes (after about 5 ms), being negligible compared to the vertical displacement u_{yy} . We obtain the strain in the sample from the DIC observations of the specimen, and the strain history is shown in Figure 3.4(c). Notice that the strain rate (the slope of Figure 3.4(c)) is relatively uniform up to approximately 25 ms. Finally, the average force exerted on the specimen is obtained via force sensors, as shown in Figure 3.4(d). Due to noise-floor already present in the force measurements, we filter our force

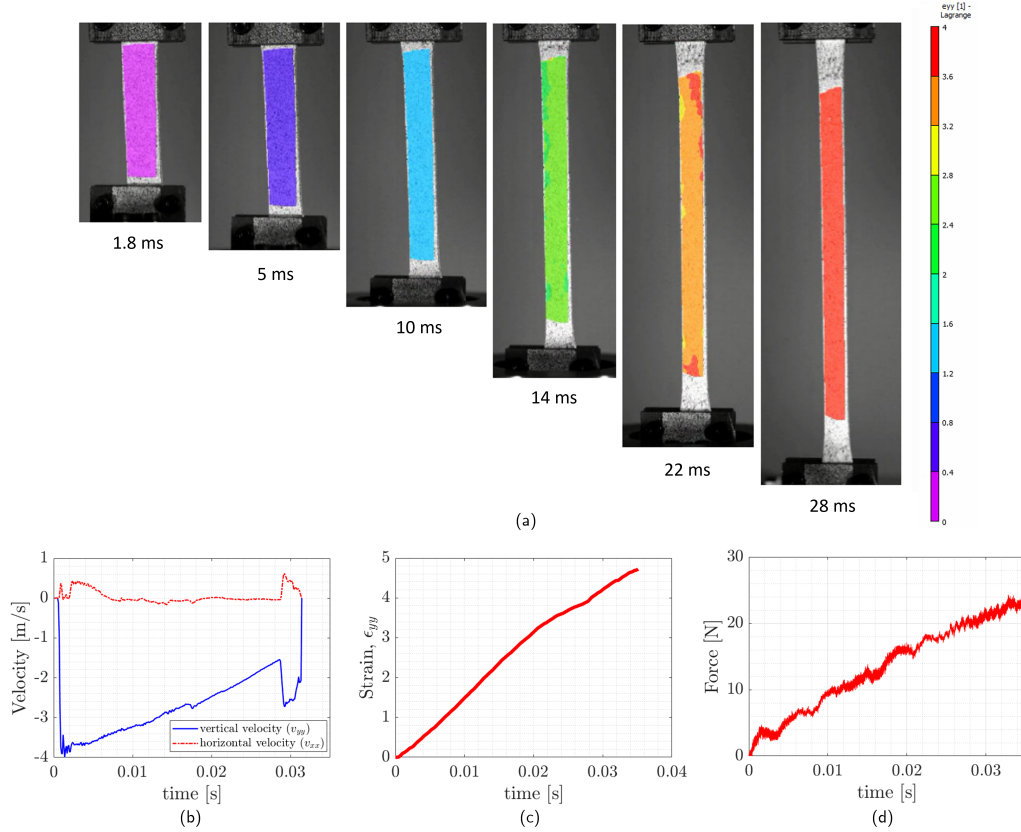


Figure 3.4: Representative experimental observations. Top: (a) Deformation of the specimen and the evolution of the strain fields as captured by DIC at different times. Note that the strain is uniform across the sample. Bottom: (b) Vertical (v_{yy}) and horizontal (v_{xx}) velocity of the sliding plate, obtained from DIC analysis. (c) Strain in time, obtained from DIC analysis. (d) Force in time, obtained from force sensors.

measurements with a moving filter which uses a window size that averages high frequency oscillations. Specifically, the time increment between each filter is 1×10^{-5} s and the window size is 2×10^{-5} s.

We now discuss the average behavior of all six natural rubber specimens tested. The resulting strain rates in all six samples are shown in Figure 3.5(a), where the green envelope represents the spread of strain rates across all samples. Across all six samples, the nominal average strain rate achieved is 155 s^{-1} . As depicted, the specimens begin deforming at a relatively constant strain rate for approximately 20 ms. The experimental rise time is typically around 0.8 ms, during which a strain of 0.08 is accumulated. Following this, the strain rate stabilizes at an average of 155 s^{-1} from 2.5 ms to 20 ms. The maximum standard deviation of the strain rate across all experiments is 16 s^{-1} . After 20 ms, there is a gradual drop in the strain

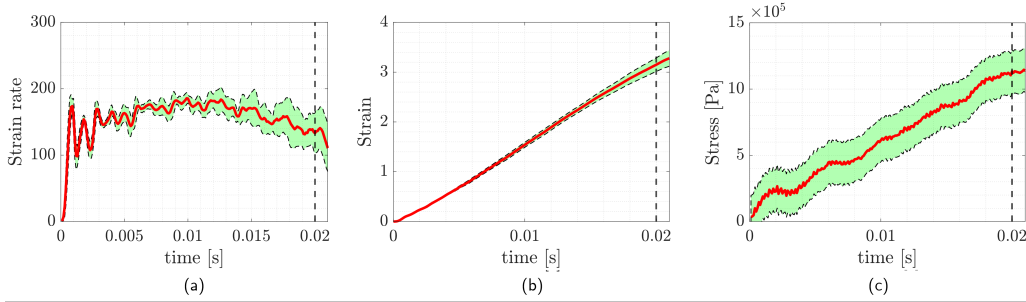


Figure 3.5: Drop tower results across six samples. (a) Average nominal strain rate in time. The green envelope represents the spread of strain rates across all six specimens. (b) Nominal strain in time. The green envelope represents the spread of strain values across all six specimens. (c) Piola-Kirchhoff stress in time. The upper and lower bounds of stress are calculated from uncertainties in force measurements, as detailed in Chapter 3.2, with a maximum standard deviation of 0.16 MPa.

rate, which DIC analysis attributes to slipping near the clamped region. This drop is followed by a sudden increase in strain rate, caused by a second impact of the striker, as confirmed by DIC. The second impact occurs when the striker bounces off the sliding plate after the initial impact and falls back, striking the plate again. However, in post-processing, we discard any data once a significant drop in strain rate or a second impact is observed, whichever comes first. Specifically, data after 20 ms is omitted - see the dashed lines in the three plots of Figure 3.5. The strain evolution over time is shown in Figure 3.5(b), where, once again, the envelope represents the spread across all six samples. On average, the strain increases up to nearly $\epsilon_{yy} = 3$ within 20 ms. It has a maximum spread of 0.2 at 3.5 strain across all six specimen tested. Variations in loading conditions and initial impact velocity contribute to the observed differences in strain rates and strain across the specimens. Figure 3.5(c) shows the nominal (Piola-Kirchhoff or engineering) stress history, which increases up to 1.2 MPa in 20 ms. The primary source of uncertainty in the stress response arises from noise-floor oscillations in the force measurements, as discussed in Section 3.2. The maximum standard deviation of the stress measurements is 0.16 MPa, and this uncertainty is illustrated by the envelope in Figure 3.5(c).

To verify our technique, we supplement our experimental observations with finite element simulations. We implement a Neo-Hookean constitutive model through deal.II finite element library [26]. We begin with the quasi-incompressible Neo-Hookean free energy of the form

$$W = \frac{\mu}{2} \left[J^{-2/3} (\lambda_1^2 + \lambda_2^2 + \lambda_3^2) - 3 \right] + \frac{k}{2} (J - 1)^2, \quad (3.1)$$

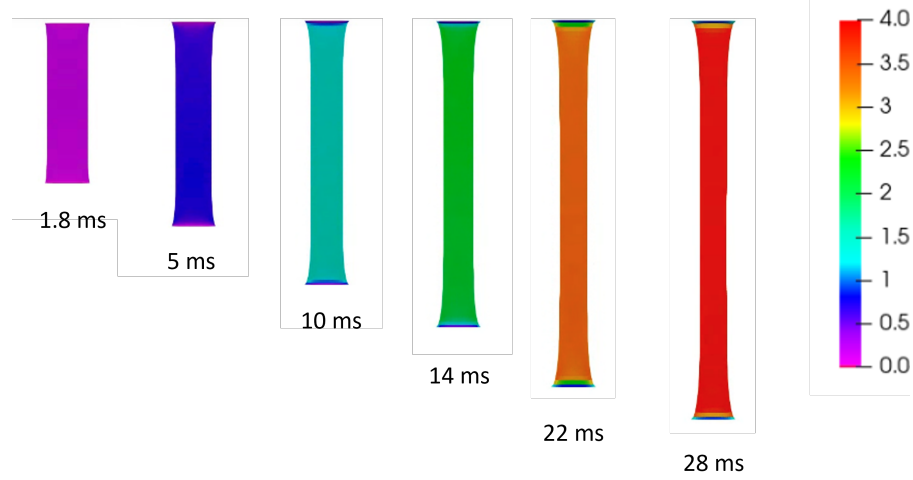


Figure 3.6: Contour plots of the simulated nominal strain in the longitudinal direction of the sample (ϵ_{yy}) at different simulation times.

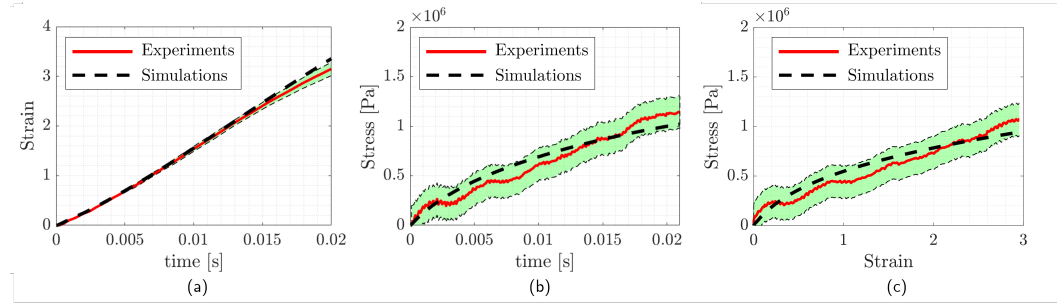


Figure 3.7: Comparison of the numerical simulation results against the drop tower experimental tests. (a) Strain in time. (b) PK stress in time. (c) Stress-strain in time. The green envelopes represent experimental uncertainty.

where $\lambda_1, \lambda_2, \lambda_3$ are the eigenvalues of the deformation gradient tensor, F , and $\lambda_1 \geq \lambda_2 \geq \lambda_3$. As usual, J is the determinant of F . We also add a viscous contribution to the stress so that the Piola-Kirchhoff stress

$$S = \frac{\partial W}{\partial \nabla u} + J [\nu_1 \text{Tr}(D) + \nu_2 \text{dev}(D)] F^{-T} \quad (3.2)$$

where D is the rate of deformation, and ν_1 and ν_2 are the volumetric and deviatoric viscosity respectively. The parameters used in our simulations are listed in Table 3.2, and an impact velocity of 3.8 m/s is used. Further details can be found in Appendix 3.A.

Figure 3.6 shows snapshots of the simulated deformation and strain fields. Note that the strain fields are uniform, as observed in the experimental tests. Comparing the simulated and the experimental deformation fields, we note that there is slipping in the experimental tests near the grips. Hence, the velocity of the sliding plate

Parameters for numerical simulations	
Shear modulus μ	0.2×10^6 Pa
Poisson's ratio ν	0.49
Density ρ	1000 kg/m ³
Bulk viscosity, ν_1	150 Pa · s
Deviatoric viscosity, ν_2	20 Pa · s

Table 3.2: Parameters used for the numerical simulations of the drop tower experiments on natural rubber specimens.

(used as boundary condition in our simulations) is higher than the actual velocity experienced by the specimen in our experiments. This slip, which is present across all samples tested, results in a higher perceived strain in the simulations (Figure 3.7(a)), especially closer to 20 ms. Nevertheless, there is a good agreement between experimental and simulated values. The observed experimental stress evolution and stress-strain curve are also consistent with the simulated values (see Figure 3.7(b) and (c)).

3.4 Conclusion

We have described a novel and versatile tensile testing system based on the drop weight technique, specifically designed for materials that can undergo significant tensile deformation, such as elastomers. The core apparatus comprises of a hanging slender bar, from which a steel sleeve (referred to as the striker) is released under controlled conditions. Accelerated in free fall, the striker impacts a stationary plate, initially held in place by mechanical detents. The specimen, secured by a gripping system between the hanging bar and the stationary support, undergoes controlled stretching at a nearly constant velocity upon the release of the detent triggered by the striker's impact. Full-field strain measurement is obtained using a high-speed camera in conjunction with digital image correlation. Additionally, strategically located piezoresistive force sensors enable real-time force measurements. By achieving strain rates ranging from 100 s^{-1} to 500 s^{-1} , this system addresses a notable gap in the literature concerning intermediate strain rate testing for soft materials.

This test system fills an important gap between universal testing machines that are typically limited to strain rates of 1 s^{-1} and below, and split-Hopkinson (Kolsky) pressure bars that are typically limited to strains of 10^3 s^{-1} and above. We have demonstrated the system with experiments in natural rubber with strain rates of 10^2 s^{-1} and strains of 3. In current and future work, we use the testing system to study the rate and history dependent properties of soft materials, including liquid crystal

elastomers and biological materials, as well as architected materials.

Acknowledgement

We gratefully acknowledge the financial support of the National Science Foundation (DMS:2009289) and the Army Research Office (W911NF22-2-0120). J.C. Nieto-Fuentes acknowledges support from the CONEX-Plus programme funded by Universidad Carlos III de Madrid and the European Union's Horizon 2020 research and innovation programme, under the Marie Skłodowska-Curie grant agreement No. 801538.

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APPENDIX TO CHAPTER 3

3.A Computational approach

We implement this constitutive model to a dynamic setting through the incremental action integral

$$\mathcal{A} = \int_{t_1}^{t_2} \left[\left(\mathcal{E} - \int_{\Omega} f \cdot u \, d\Omega - \int_{\partial_t \Omega} t \cdot u \, dS \right) - \int_{\Omega} \rho \frac{\dot{u}^2}{2} \, d\Omega \right] dt, \quad (3.3)$$

where f and t are the body force and surface traction, respectively, and $\partial_t \Omega$ is the traction boundary. Here, the energy $\mathcal{E}$ is composed of our stored elastic energy density,

$$\mathcal{E} = \int_{\Omega} W(\nabla u, \Lambda, \Delta) \, d\Omega. \quad (3.4)$$

Stationarity of the action integral ($\delta \mathcal{A} = 0$) allows us to obtain equation of motion

$$0 = \int_{\Omega} \left(-\frac{\partial W}{\partial \nabla u} \cdot \nabla \varphi + f \cdot \varphi - \rho \ddot{u} \cdot \varphi \right) d\Omega + \int_{\partial_t \Omega} t \cdot \varphi \, dS \quad \forall \varphi \in \mathcal{U}, \quad (3.5)$$

with $\mathcal{U} = \{u : u \in H^1(\Omega \times [0, T]), u = u_*(t) \text{ on } \partial_u \Omega\}$.

We consider discretization of the displacement field with bilinear quadrilateral, Lagrange finite element of degree 1. Thus, $u(x) = \sum_i u_i N_i(x)$, where u_i are the unknown expansion coefficients and $N_i(x)$ are the finite element shape functions, i.e., linear with compact support. The system is then solved by solving for displacement fields explicitly. We discretize equation (3.5) in the finite element space with central difference scheme in time, where we take $\ddot{u} = \frac{u^{n+1} - 2u^n + u^{n-1}}{\delta t^2}$. The resulting linear equation to be solved is thus $M_{ij} u_j^{n+1} = F_i^n$, with

$$M_{ij} = \int_{\Omega} \frac{\rho}{\Delta t^2} N_i(x) N_j(x) \, d\Omega, \quad (3.6)$$

$$F_i^n = \int_{\Omega} -\sigma(\nabla u^n, \Lambda^{n+1}, \Delta^{n+1}, u_*^{n+1}) \cdot \nabla N_i(x) + \rho \left(\frac{2u_j^n - u_j^{n-1}}{\Delta t^2} \right) \cdot N_i(x) \, d\Omega. \quad (3.7)$$

The integrals above are computed using Gauss quadrature formula.

Chapter 4

HIGH STRAIN RATE BEHAVIOR OF LIQUID CRYSTAL ELASTOMERS

A. Wihardja, J. C. Nieto-Fuentes, D. Rittel, and K. Bhattacharya, “High strain rate behavior of liquid crystal elastomers”, *Journal of the Mechanics and Physics of Solids*, 106501 (2026).

Contributions: A.W developed the experimental methodology, performed the experiments, produced and processed and analyzed the experimental data, developed model, performed model simulation, and wrote the manuscript.

Abstract

Liquid crystal elastomers are rubbery solids that couple liquid crystalline order and deformation. This coupling leads to properties that are attractive for a number of applications in soft robotics and energy absorption. This paper is motivated by the latter application, and provides a systematic experimental study of a particular class of liquid crystal elastomers – the isotropic genesis polydomain liquid crystal elastomers – over a wide range of strain rates in tension and compression. An important aspect of this study is a novel tensile drop-tower that enables tensile strain rates of 100 s^{-1} that are important to application but previously inaccessible. The paper also extends a recently proposed constitutive model to the high strain rate regime, and shows that it can be fit to describe the observed behavior across the spectrum of examined behavior.

4.1 Introduction

Liquid crystal elastomers (LCEs) are cross-linked elastomers in which stiff nematic mesogens are incorporated into the polymer chains [1, 2]. The cross-linking is light enough to enable the mesogens to undergo order-disorder transitions. In nematic elastomers, the mesogens are randomly aligned in a disordered phase or isotropic phase above a transition temperature, but align around a particular direction (director) in the nematic phase below the transition temperature. Since these mesogens

are incorporated in the polymer chains, the isotropic to nematic transition leads to significant spontaneous or residual elongation along the director. The transition can also be triggered by other stimuli, including solvents, electric field, magnetic field, and light, depending on the chemistry. This interplay between temperature, nematic order, deformation, and stimuli makes LCE a promising material for actuation [3–5], mechanical dissipation [6–8], and energy absorption [8, 9].

Various types of LCEs exist depending on their chemistry and synthesis history. There are both main-chain [10, 11] and side-chain [12] LCEs. Further, they can be synthesized in both the monodomain configuration (MLCE) [13], where all mesogens are aligned uniformly during synthesis, or in the polydomain configuration (PLCE), where the mesogens form domains each having its own local orientation during synthesis. Since PLCEs can be made without requiring any alignment, they are scalable and easy to manufacture, making them good engineering materials [11]. There are two main types of PLCEs: isotropic-genesis PLCEs (I-PLCEs) and nematic-genesis PLCEs (N-PLCEs). I-PLCEs are made via cross-linking in the high-symmetry isotropic phase in solution before removing the solvent to trigger the phase transition to the nematic phase. I-PLCEs typically have a large number of randomly oriented domains in the nematic phase, and the material retains its overall isotropy. N-PLCEs are cross-linked in the nematic phase. Importantly, this different cross-linking state gives rise to dissimilar mechanical behavior, especially concerning the soft behavior [14, 15].

A unique feature of LCEs is their soft behavior, where they can undergo large stretch under low, constant stress [1, 13, 14, 16, 17]. While initially discovered in MLCEs [13], the soft behavior is also observed in I-PLCEs [14, 16, 17] as a result of a polydomain to monodomain transition [15–18]. Indeed, this soft behavior can manifest itself in rather complex ways, including an in-plane liquid-like behavior, i.e., they are unable to sustain shear stresses in the plane [19].

This soft behavior is known to be extremely sensitive to the strain rate [16, 20–22]. This indicates that the polydomain to monodomain transition, and mesogen reorientation more broadly, is rate-dependent. This is supported by the fact that these materials have a large loss tangent over a wide range of temperature and frequencies (75–110°C, 1–100 Hz) [6, 7].

The combination of soft behavior and large mechanical damping makes these materials extremely attractive for energy absorption and mechanical protection applications. For example, Jeon *et al.* [9] showed that I-PLCE-based architected material

displays extremely large energy absorption density (5MJ/m^3) at a high overall strain rate of 600 s^{-1} . This has motivated the study of the rate-dependent behavior of I-PLCEs. These include small amplitude vibration characterization using a Dynamic Mechanical Analysis, quasi-static conditions in the range of $10^{-4} - 10^{-1}\text{ s}^{-1}$ using universal testing machines, and compression tests at very high strain rates of 10^3 s^{-1} using a split Hopkinson pressure bar (SHPB) or Kolsky bar [8]. Unfortunately, this leaves a critical gap at strain rates of 10^2 s^{-1} , especially in tension, where much of the interesting physics of impact mitigation and soft behavior occurs.

This paper addresses this critical gap in thiol-acrylate-based main chain I-PLCEs. We conduct quasistatic tests in both tension and compression using a universal testing machine, dynamic tension tests at strain rates of 10^2 s^{-1} using a newly developed tensile testing apparatus [23], and dynamic compression tests at strain rates of strain rates of 10^3 s^{-1} using a split Hopkinson pressure bar. Together, they provide a comprehensive description of the strain-rate behavior of the I-PLCEs.

We also develop a macroscopic model of this complex strain-rate-dependent soft behavior by extending the model of Lee *et al.* [24] to include rate-dependence, viscoelasticity and compressibility. I-PLCEs have multiscale nature as has been discussed elsewhere [14, 15, 18, 24]. We are interested in the macroscopic or engineering scale where a typical representative volume consists of a large number of domains. Each domain has a relatively uniform director, i.e., the nematic mesogens are aligned almost uniformly about a particular direction or director. The mechanical behavior of the elastomer at the domain scale is described by the celebrated Bladon-Warner-Terentjev theory [25]. This theory is characterized by an anisotropy parameter r that describes the ratio of the spontaneous or residual stretch in the direction of the director to that perpendicular to the director. This theory is non-convex and leads to domains in monodomain LCEs, and the relaxation of this theory has been computed by Desimone and Dolzmann [26]. In I-PLCEs, one has domains due to relaxation, but also due to synthesis, and thus one needs a combination of relaxation and homogenization. This is studied with bounds in Biggins *et al.* [15] and with numerical simulation in Zhou and Bhattacharya [18]. Importantly, the domain patterns as well as the orientation of the directors within the domain, and consequently the spontaneous deformation, can change when the I-PLCE is subject to stress. These results are the basis of the macroscopic constitutive model of Lee *et al.* [24]. The key idea of this theory is that the spontaneous deformation of I-PLCEs is characterized by two internal variables Λ and Δ , and the behavior is characterized

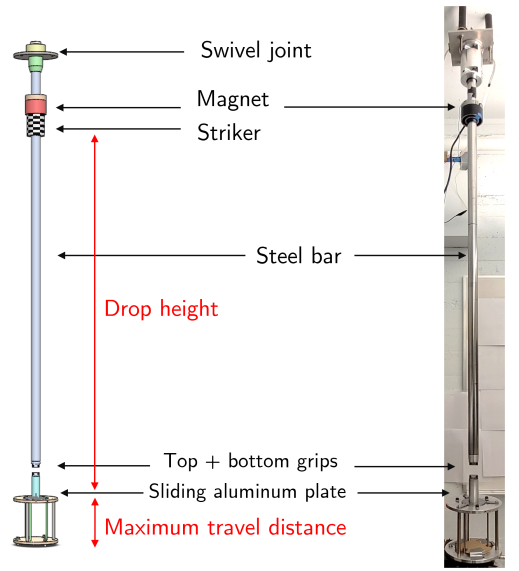


Figure 4.1: Schematic drawing (left) and image (right) of the drop weight tensile testing system. Note that the striker is not shown in the right image.

by the evolution of these internal variables. This theory is rich enough to address monodomain and polydomain LCEs. The strain rate response of LCEs depends on both the kinetics of evolution and the viscoelasticity of the polymer network. This has been modeled by Wang *et al.* [27] focusing on monodomain LCEs, building on the viscoelastic framework of Reese and Govindjee [28]. In this work, we extend the model of Lee *et al.* [24] to include network viscoelasticity. We show that all our experimental observations can be fit to a single set of parameters in this model, and that the model provides further insight into the polydomain-monodomain transition at various rates.

4.2 Experimental Methods

Liquid Crystal Elastomers Synthesis

We study main chain, thiol-acrylate-based, isotropic-genesis polydomain LCEs (I-PLCEs) fabricated using Michael addition click reaction [11] (also [29]).

Tension tests

Quasistatic tests, at a strain rate of $\dot{\epsilon} = 10^{-2} \text{ s}^{-1}$, are conducted using a universal testing machine, MTS Model 358.10 with an axial load capacity of 13.3 kN.

Intermediate strain rate tests are conducted with a custom-made drop tower setup shown in Figure 4.1, and are discussed in detail in Nieto-Fuentes *et al.* [23]. Briefly,

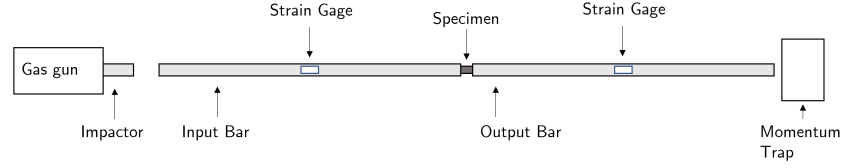


Figure 4.2: Schematic drawing of the split Hopkinson Pressure Bar (SHPB) setup consisting of impactor, input bar, and output bar.

the setup consists of a stainless steel striker, which is guided down by a stainless steel bar. At the bottom of this bar is a clamp that secures one edge of the I-PLCE specimen. The other edge of the specimen is clamped to the bottom clamp, which is connected to a sliding aluminum plate capable of traveling vertically up and down. When the striker is released, it travels through the guiding rod until it hits the bottom clamp, pushing it downward and applying a tensile force on the specimen. The impact velocity and strain rate of the material can be controlled by adjusting the drop height of the striker.

A series of images is taken immediately following impact, using a high-speed camera (Photron Fastcam NOVA S12 camera and Tokina AT-X PRO lens, 100F 2.8D) at 10,000 frames per second with a resolution of 1024 by 1024 pixels under a continuous light source. The images are used to obtain the displacement field within the specimen and the displacement of the bottom clamp using 2D digital image correlation (DIC) with the Correlated Solutions Vic2D software. The subset size is 21×21 pixels and the step size is 1 pixel. The Green-Lagrange strain tensor ($E = \frac{1}{2}(F^T F - I)$ where F is the deformation gradient) is calculated using a strain filter of 5. The forces exerted by the specimen during impact are measured using two force sensors (Tekscan FlexiForce™ A201 of 445 N limit) at the top of the setup. Very high frequency oscillations in the force sensors are filtered by averaging over a window of 5 microseconds that is much smaller than the phenomena of interest [23]. After the striker is launched, once the stress wave reaches the force sensors, diagnostics are triggered at 200 MHz, and a Siglent SDS 1202X-E digital oscilloscope records the force data.

All tension tests are carried out with a specimen shown in Figure 4.5(a).

Compression tests

Quasistatic compression tests are conducted on I-PLCE pads with diameter of 10 mm and thickness of 3 mm, at a strain rate of $\dot{\epsilon} = 10^{-2} \text{ s}^{-1}$, using a universal testing machine, MTS Model 358.10.

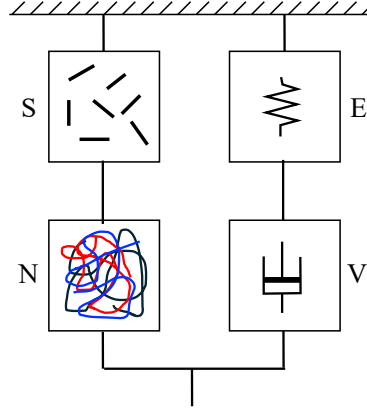


Figure 4.3: Rheological model of LCE.

High strain rate compression tests are conducted on I-PLCE pads of diameter 6.7 mm and thickness 3 mm using a split-Hopkinson pressure bar (SHPB) detailed in [30] and shown schematically in Figure 4.2. The setup consists of an impactor, an incident bar, and an output bar. Strain gages (Omega SGD-2D/350-LY1) are placed in the middle of the incident and output bars, and the specimen is placed between the input and output bars. Petroleum jelly is used between the specimen and the bars to minimize the friction between the specimen and the bars. The strain gages are connected to a signal conditioning amplifier (Vishay 2310B), which connects to the digital oscilloscope (2.5 GHz Tektronix DPO 3014). A Maraging 350 steel (AMS 6515) is used for the impactor (20/30 cm), incident bar (1.24 m), and output bar (1.24 m). The impactor, launched into the system using a gas gun, hits one end of the input bar, giving rise to a compressive stress wave that travels through the input bar to the specimen. Once this stress wave reaches the incident strain gage, diagnostics are triggered, and the oscilloscope records the strain gage voltage data. A Photron Fastcam NOVA S12 camera with a 100 mm Tokina AT-X Pro lens with a continuous light source is once again used to capture the images of the specimen during impact at 200,000 frames per second with a resolution of 256×128 pixels.

4.3 Constitutive Model of I-PLCE

Kinematics

We consider a specimen of I-PLCE occupying a region Ω in its natural reference configuration at a temperature above the isotropic-nematic transition temperature T_{ni} . An I-PLCE has a multiscale structure (see Figure 1 of Lee *et al.* [24]), and we seek a model to describe the behavior at the macroscopic or engineering scale.

Thus, each material point corresponds to a representative volume that potentially consists of a large number of domains as described in Lee *et al.* [24]. We denote the deformation as $y : \Omega \rightarrow \mathbb{R}^3$, the deformation gradient as F , and the temperature by T .

We model the I-PLCE using the rheological model shown schematically in Figure 4.3 following [28]. The left branch describes the mechanics of the nematic domains and the entropic elasticity of the rubber chain, and largely follows the treatment of Lee *et al.* [24]. The right branch adds viscoelasticity [28]. We introduce a spontaneous stretch F^s , a network elastic deformation F^n , a viscous deformation F^v and an elastic deformation F^e that follow

$$F = F^s F^n = F^e F^v. \quad (4.1)$$

The spontaneous stretch F^s reflects the macroscopically stress-free deformation due to the presence of a domain pattern at the material point. It is a unimodular, positive-definite, symmetric tensor. Following Lee *et al.*, we use a representation in terms of its eigenvalues and orientation:

$$F^s = F^s(\Lambda, \Delta, Q) = G^{1/2}, \quad G = QG_0Q^T, \quad \text{where} \quad G_0 = \begin{bmatrix} \Lambda^2 & 0 & 0 \\ 0 & \Delta^2/\Lambda^2 & 0 \\ 0 & 0 & 1/\Delta^2 \end{bmatrix} \quad (4.2)$$

in an orthonormal basis fixed in the material frame. Note that we parametrize the two eigenvalues of the spontaneous stretch with parameters Λ and Δ and the orientation with Q . As described in Lee *et al.*, these are related to the domain pattern. Specifically, the internal variable Λ describes the degree of nematic order, while the internal variable Δ describes the degree of planar order. They are subject to the constraints,

$$(\Lambda, \Delta) \in \mathcal{A}_G = \{(\Lambda, \Delta) : \Delta \leq r^{1/6}, \Delta \leq \Lambda^2, \Delta \geq \sqrt{\Lambda}\}. \quad (4.3)$$

The values of the internal variables for some special states are shown in Table 4.1. For example, if the material point were a monodomain, G is the anisotropy tensor of Bladon, Warner and Terentjev [25]: $G = I_n = r^{-1/3}(I + (r - 1)n \otimes n)$ where $n = Qe_1$ is the orientation of the director. Q denotes the orientation of spontaneous stretch, and maps a frame in the reference configuration to a frame in the current configuration.

For future use, we denote the spatial velocity gradient as $L = \dot{F}F^{-1}$, and its viscous contribution as $L^v = \dot{F}^v(F^v)^{-1}$. It follows that

$$L = \dot{F}^e(F^e)^{-1} + F^e \dot{F}^v(F^v)^{-1}(F^e)^{-1}. \quad (4.4)$$

Table 4.1: The internal variables and spontaneous stretch.

Domain alignment	$\{\Lambda, \Delta\}$	$G_0^{1/2}$
Random or isotropic	1, 1	$\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$
Fully aligned	$\{r^{1/3}, r^{1/6}\}$	$\begin{pmatrix} r^{1/3} & 0 & 0 \\ 0 & r^{-1/6} & 0 \\ 0 & 0 & r^{-1/6} \end{pmatrix}$
Random in-plane or equi-biaxial	$\{r^{1/12}, r^{1/6}\}$	$\begin{pmatrix} r^{1/12} & 0 & 0 \\ 0 & r^{1/12} & 0 \\ 0 & 0 & r^{-1/6} \end{pmatrix}$

Balance Laws

The balance laws are standard,

$$\rho \ddot{\mathbf{y}} = \nabla \cdot \mathbf{P} + \mathbf{b} \quad (\text{linear momentum}), \quad (4.5)$$

$$\mathbf{F} \mathbf{P}^T = \mathbf{P} \mathbf{F}^T \quad (\text{angular momentum}), \quad (4.6)$$

$$\dot{U} - s + \nabla \cdot \mathbf{h} - \mathbf{P} \cdot \dot{\mathbf{F}} = 0 \quad (\text{energy}), \quad (4.7)$$

$$\dot{\eta} - \frac{s}{T} + \nabla \cdot \left(\frac{\mathbf{h}}{T} \right) \geq 0 \quad (\text{Clausius-Duhem inequality}) \quad (4.8)$$

in local Lagrangian form where ρ is the density, $\mathbf{P}$ the Piola-Kirchhoff stress, $\mathbf{b}$ the body force, U the internal energy density, s the heat source, $\mathbf{h}$ the heat flux and η the entropy density (note that all densities are per unit reference volume). We introduce a free energy density $W = U - T\eta$, and use the balance of energy in the Clausius-Duhem inequality to rewrite it as

$$\mathbf{P} \cdot \dot{\mathbf{F}} - \dot{W} - \eta \dot{T} - \frac{1}{T} \mathbf{h} \cdot \nabla T \geq 0. \quad (4.9)$$

Constitutive framework

We postulate that

$$\begin{cases} W = \hat{W}(F, F^e, \Lambda, \Delta, Q, T), \\ \mathbf{P} = \hat{\mathbf{P}}^e(F, F^e, \Lambda, \Delta, Q, T), \\ \eta = \hat{\eta}(F, F^e, \Lambda, \Delta, Q, T), \\ \mathbf{h} = \hat{\mathbf{h}}(\nabla T, T). \end{cases} \quad (4.10)$$

Substituting it into the inequality (4.9) yields

$$\begin{aligned} & \left(\hat{P}^e(F, F^e, \Lambda, \Delta, Q, T) - \frac{\partial \hat{W}}{\partial F} - \frac{\partial \hat{W}}{\partial F^e} (F^v)^{-T} \right) \cdot \dot{F} + \frac{\partial \hat{W}}{\partial F^e} \cdot F^e L^v \\ & - \frac{\partial \hat{W}}{\partial \Lambda} \dot{\Lambda} - \frac{\partial \hat{W}}{\partial \Delta} \dot{\Delta} - \frac{\partial \hat{W}}{\partial Q} \dot{Q} - \left(\frac{\partial \hat{W}}{\partial T} + \eta \right) \dot{T} - \frac{1}{T} h \cdot \nabla T \geq 0. \end{aligned} \quad (4.11)$$

We expect that this inequality holds for all processes consistent with the balance of momenta and energy. Therefore, arguing as in Coleman and Noll [31], we conclude

$$\hat{P}^e = \frac{\partial \hat{W}}{\partial F} + \frac{\partial \hat{W}}{\partial F^e} (F^v)^{-T}, \quad \eta = -\frac{\partial \hat{W}}{\partial T}, \quad h \cdot \nabla T \leq 0. \quad (4.12)$$

The inequality now reduces to

$$(F^e)^T \frac{\partial \hat{W}}{\partial F^e} \cdot L^v - \frac{\partial \hat{W}}{\partial \Lambda} \dot{\Lambda} - \frac{\partial \hat{W}}{\partial \Delta} \dot{\Delta} - \frac{\partial \hat{W}}{\partial Q} \dot{Q} \geq 0. \quad (4.13)$$

We note that each term is a product of an energetic quantity and a rate quantity, and therefore use it to identify the driving forces

$$\begin{cases} d_v = (F^e)^T \frac{\partial \hat{W}}{\partial F^e} & \text{for } L^v, \\ d_\Lambda = -\frac{\partial \hat{W}}{\partial \Lambda} & \text{for } \dot{\Lambda}, \\ d_\Delta = -\frac{\partial \hat{W}}{\partial \Delta} & \text{for } \dot{\Delta}, \\ d_Q = \text{skew} \left(-\frac{\partial \hat{W}}{\partial Q} Q^T \right) & \text{for } A, \text{ where } \dot{Q} = AQ. \end{cases} \quad (4.14)$$

that determine the evolution of L^v , state variables Λ , Δ and microstructure orientation Q respectively. Note that $Q \in SO(3)$, and hence $\dot{Q} = AQ$ for some A skew, and it is natural to only look at the skew part of the final term. We postulate

$$\begin{cases} L^v = \hat{L}^v(d_v), \\ \dot{\Lambda} = \hat{k}_\Lambda(d_\Lambda), \\ \dot{\Delta} = \hat{k}_\Delta(d_\Delta), \\ \dot{Q} = \hat{A}(d_Q). \end{cases} \quad (4.15)$$

In summary, the constitutive relations are specified as $\hat{W}, \hat{L}^v, \hat{k}_\Lambda, \hat{k}_\Delta, \hat{A}$ with the remaining quantities obtained from (4.12).

Before we describe specific constitutive relations, we note that the energy balance may be written as

$$c\dot{T} = s - \nabla \cdot h + d_\Lambda \dot{\Lambda} + d_\Delta \dot{\Delta} + d_Q \dot{Q} + d_v \cdot L^v. \quad (4.16)$$

where

$$c = -\frac{\partial^2 W}{\partial T^2} T \quad (4.17)$$

is the specific heat.

Specific constitutive relations

We begin with the energy, and assume

$$\hat{W}(F, F^e, \Lambda, \Delta, Q, T) = \hat{W}^n(F, \Lambda, \Delta, Q, T) + \hat{W}^e(F^e, T) + W^t(T), \quad (4.18)$$

where W^n describes the nematic elasticity (analogous to the equilibrium elasticity in viscoelasticity), W^e the network elasticity that contributes to the viscoelasticity (analogous to the non equilibrium elasticity), and W^t is the purely thermal energy. We further assume that

$$\hat{W}^n(F, \Lambda, \Delta, Q, T) = \hat{W}^{ne}(F^T Q G_0^{-1}(\Lambda, \Delta) Q^T F, T) + \hat{W}^{nn}(\Lambda, \Delta, T). \quad (4.19)$$

$\hat{W}^{ne}$ accounts for the entropic elasticity of the polymer chains averaged over the domains, and W^{nn} is the stored energy that results when the domains are forced out of their as-synthesized isotropic state.

Note that W^{ne} can depend on the orientation Q of the domain pattern, and can be anisotropic for a given domain pattern. However, I-PLCE is synthesized in the isotropic state, and the overall behavior of the material is consequently isotropic. So, we assume that the material forms a domain pattern in the most energetically favorable orientation. It follows that $d_Q = 0$, and we can minimize out Q from the energy and replace $\hat{W}^{ne}$ with

$$\tilde{W}^{ne}(F, \Lambda, \Delta, T) = \min_Q \hat{W}^{ne}(F, \Lambda, \Delta, Q, T). \quad (4.20)$$

If $W^{ne}(C, T)$ is isotropic in C (i.e., $W^{ne}(R^T C R, T) = W^{ne}(C, T) \forall R \in SO(3)$), then this minimum is attained when the principal directions of F coincide with the eigendirections of $Q G_0^{-1}(\Lambda, \Delta) Q^T$. Consequently, $\tilde{W}^{ne}$ is isotropic and we take

$$\tilde{W}^{ne}(F, \Lambda, \Delta, T) = \frac{\mu_1}{2} \left(J^{-2/3} \left(\frac{\lambda_1^2}{\Lambda^2} + \frac{\lambda_2^2 \Lambda^2}{\Delta^2} + \lambda_3^2 \Delta^2 \right) - 3 \right) + \frac{\kappa_1}{2} \left(\frac{J^2 - 1}{2} - \ln J \right), \quad (4.21)$$

where $\lambda_1 \geq \lambda_2 \geq \lambda_3$ are the ordered principal values of F and $J = \det F = \lambda_1 \lambda_2 \lambda_3$, by extending the incompressible formulation of Lee *et al.* [24] to the compressible case. We also assume

$$\hat{W}^{nn}(\Lambda, \Delta, T) = C \frac{\Delta - 1}{(r^{1/6} - \Delta)^k}, \quad (4.22)$$

$$\hat{W}^e = \frac{\mu_2}{2} \left(J_e^{-2/3} \text{tr}(F^{eT} F^e) - 3 \right) + \frac{\kappa_2}{2} \left(\frac{J_e^2 - 1}{2} - \ln J_e \right) \quad (4.23)$$

and

$$\hat{W}^t = -cT \ln T. \quad (4.24)$$

Turning now to the kinetics or evolution (4.15), we assume linear kinetics for the state variables:

$$\dot{\Lambda} = \alpha_\Lambda d_\Lambda, \quad \dot{\Delta} = \alpha_\Delta d_\Delta \quad (4.25)$$

for kinetic coefficients $\alpha_\Lambda, \alpha_\Delta$. Finally, note that the postulated $\hat{W}^e$ is frame-indifferent (following standard practice in viscoelasticity), and therefore d_v is symmetric. Consequently, we can replace L^v with its symmetric part or strain rate tensor D^v in (4.13), and consequently (4.15)₁. We follow the practice in viscoelasticity and rewrite the first member of (4.15) as $d_v = \hat{d}_v(D^v)$. We also decompose the strain rate into hydrostatic and deviatoric parts, and assume a nonlinear relation for the deviatoric evolution and linear relation for the hydrostatic viscosity:

$$d_v = \nu_h(\text{tr } D^v)I + \nu_d(||D^{v,\text{dev}}||)D^{v,\text{dev}} \quad (4.26)$$

where I is the identity, $D^{v,\text{dev}} = D^v - \frac{1}{3}(\text{tr } D^v)I$ is the deviatoric part of D^v , ν_h is the volumetric viscosity, $\nu_d(\dot{\gamma})$ is the rate-dependent viscosity (it is shear hardening if it is increasing and shear weakening if it is decreasing) that satisfies $\nu_d(0) = \nu_0$, $\nu_d \geq 0$. We specifically take ν_d to be in the Cross form [32]

$$\nu_d(\dot{\gamma}) = \nu_\infty + \frac{\nu_0 - \nu_\infty}{1 + (\dot{\gamma}/\dot{\gamma}_0)^m} \quad (4.27)$$

for constants ν_0 (creeping viscosity), ν_∞ (high rate viscosity), $\dot{\gamma}_0$ (reference shear rate) and m (viscosity exponent).

4.4 Results and Discussion

Tension

Unaligned, isotropic-genesis, polydomain liquid crystal elastomers (I-PLCEs) are tested at three nominal strain rates of 0.01 s^{-1} , 100 s^{-1} , and 300 s^{-1} . The samples used in both 100 s^{-1} and 250 s^{-1} tests come from two different batches, but we have verified that the batch-to-batch variation is small compared to the rate effects.

Quasistatic tests Figure 4.4 shows the stress-strain behavior of the I-PLCE at a strain rate of $\dot{\epsilon} = 10^{-2} \text{ s}^{-1}$. The noise and the apparent strain increase when the

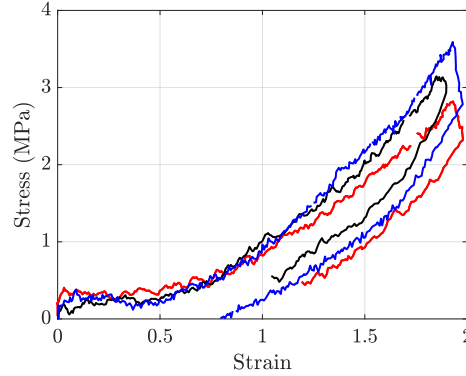


Figure 4.4: Behavior of I-PLCE in tension at quasistatic strain rate of $\dot{\epsilon} = 10^{-2} \text{ s}^{-1}$.

stress is reversed reflect the signal to noise ratio of a 13.3kN load capacity universal testing machine on a soft material. Still, the loading behavior is consistent with many observations in the literature. We observe the characteristic soft behavior with a stress plateau of about 0.3 MPa.

Tests at 100 s^{-1} We obtain a striker velocity of 3.4 m/s with a drop height of 0.7 m, and this leads to a nominal strain rate of 100 s^{-1} as we see later. Typical results are shown in Figure 4.5. The specimen before (top) and after (bottom) the test is shown in Figure 4.5(a)¹. Note that the test proceeds until specimen failure, which typically occurs at the grip. While this indicates that failure is influenced by local stress concentrations at the clamping region, the stress-strain response of large strains up to failure is reproducible and representative of the bulk material behavior. A typical snapshot superposed with the DIC strain field is shown in Figure 4.5(b), while a typical observation of the Green-Lagrange strain (averaged at mid gage over an area of 60 mm^2) and Piola-Kirchhoff engineering stress as a function of time is shown in Figures 4.5(c) and (d) respectively. We observe oscillations in the stress response with a frequency of about 4 milliseconds. These oscillations are present across all our experiments, and as discussed extensively in our previous work [23], are artifacts created by the interaction between longitudinal and flexural waves in the steel bar and load sensors. So we disregard these oscillations as measurement artifacts and do not consider them in our models in the future section. Keeping with our approach of displaying the data as obtained, we do not filter or process them, but simply do not consider them in our models.

¹The image of the failed specimen in Figure 4.5(a) was taken shortly after failure, and remains unchanged at room temperature for months.

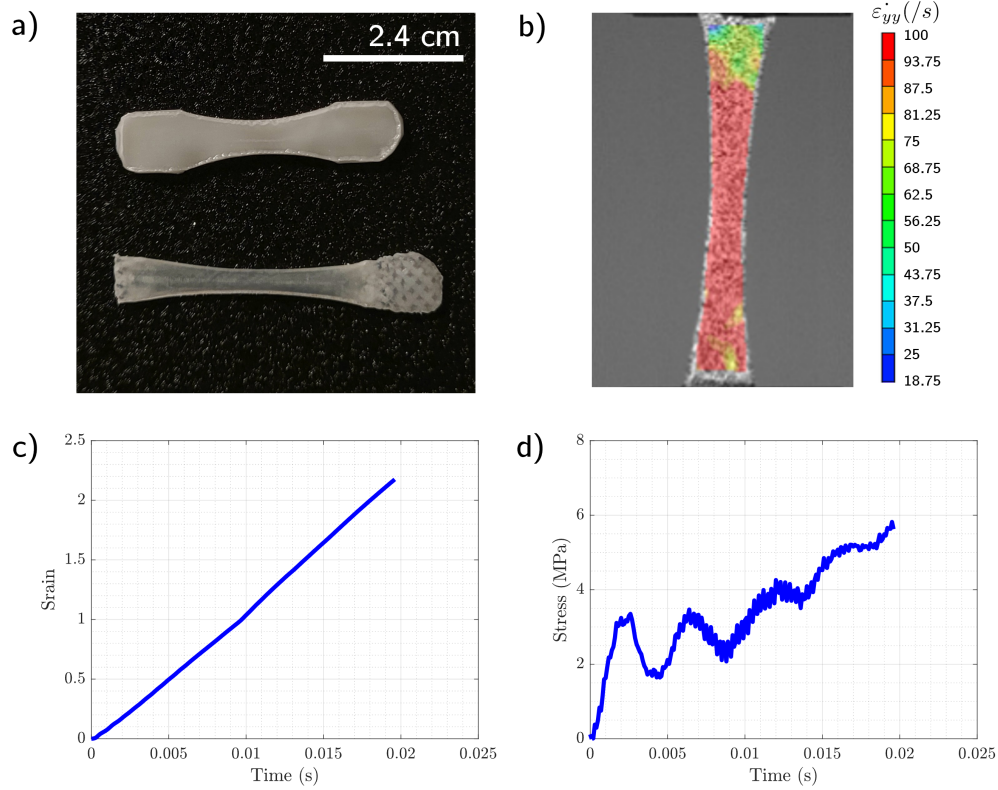


Figure 4.5: Typical results of a test at 3.4 m/s striker velocity or 100 s^{-1} nominal strain rate. (a) Specimen before and after the test. (b) Snapshot of the specimen during the test superposed with the DIC strain rate in the stretching direction. (c) Green-Lagrange strain as a function of time. Strain is computed as the average value obtained at mid gage over an area of 60 mm^2 . (d) Piola-Kirchhoff or engineering stress as a function of time.

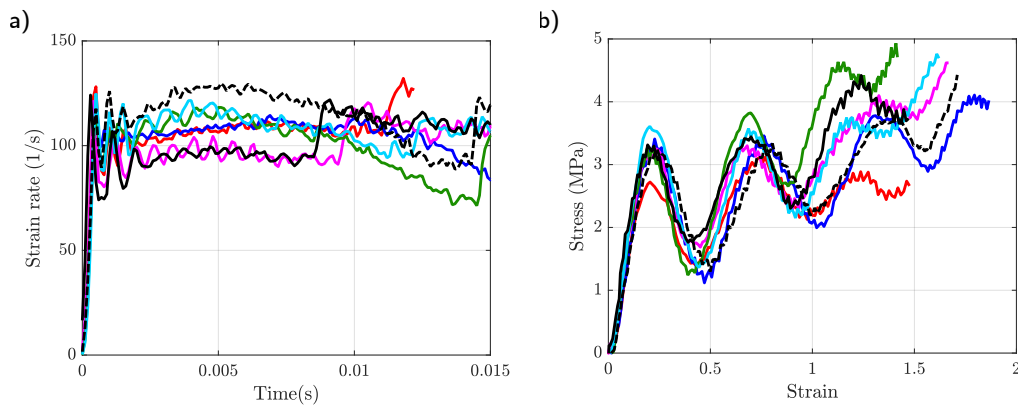


Figure 4.6: The observed response of the I-PLCE at a nominal strain rate of 100 s^{-1} . (a) Strain rate evolution in time and (b) Stress and strain response.

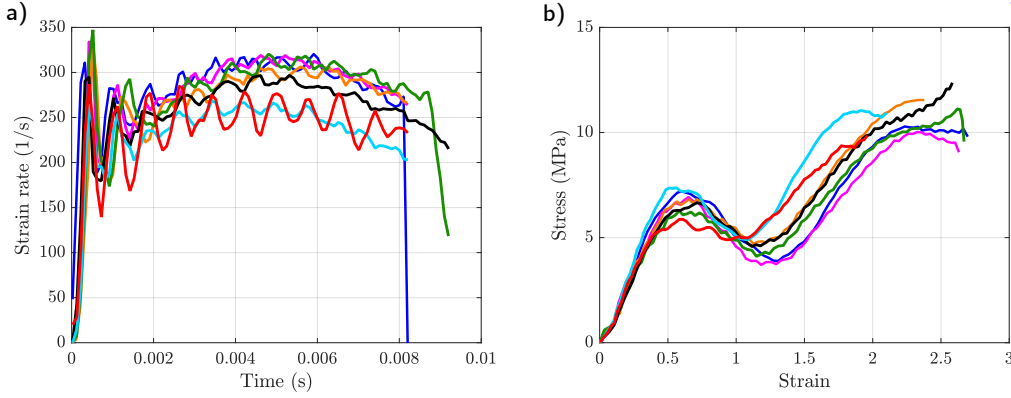


Figure 4.7: The observed response of the I-PLCE at a nominal strain rate of 250-300 s^{-1} . (a) Strain rate evolution in time and (b) Stress and strain response.

The strain rate as a function of time, as well as the stress-strain behavior, over a number of tests is shown in Figures 4.6(a) and (b). We observe that the strain rate is approximately constant within $\pm 10\%$ at a value of 100 s^{-1} . Therefore, we regard these tests as tests at a nominal strain rate of 100 s^{-1} . In general, the stress-strain response at this rate is much stiffer than that in the quasi-static regime shown in Figure 4.4. For example, the peak stress at 100 s^{-1} reaches 3 MPa, compared to 0.3 MPa at 0.01 s^{-1} .

The soft behavior corresponding to the polydomain to monodomain transition is not immediately evident in Figure 4.6: the response is initially elastic but then transitions to a flow-type behavior (with the oscillations discussed earlier). However, we observe that the specimen changes from opaque to translucent during the tests (see Figure 4.5(a)), showing that there is indeed alignment of the domains or mesogens towards the loading direction. So we conclude that the soft behavior is present, but masked by the rate effects, in the macroscopic stress-strain measurements.

Finally, the figures show that there is some variability in the observations due to variability across specimens. To be quantitative, we examine the standard deviation across the response of the seven specimens at 12 milliseconds. We find an average strain of 1.4 with 0.065 standard deviation, average strain rate of 105 s^{-1} with 12 s^{-1} standard deviation and average stress of 3.8 MPa with 0.6 MPa standard deviation. We also note that we do not observe a clear correlation between higher nominal strain rates and higher stress amongst these tests: compare the two tests marked in black in the figure: the test indicated in a solid line has a lower strain rate, but a higher stress value.

Tests at 300 s^{-1} We obtain a striker velocity of 5.4 m/s with a drop height of 1.6 m. The strain rate as a function of time, as well as the stress-strain behavior, over a number of tests is shown in Figure 4.7(a) and (b). We observe that the strain rate is not constant over time (in contrast to the previous set of tests at a striker velocity of 3.4 m/s). Instead they start off at around 250 s^{-1} and reach a peak of about 300 s^{-1} before falling back to around 250 s^{-1} . We label these tests with the peak strain rate of 300 s^{-1} .

We observe that we have an initial elastic behavior until about a stress of 7 MPa which is significantly higher than the 3 MPa observed in tests with striker velocity 3.4 m/s, and the 0.3 MPa in quasistatic tests. We have a flow-like behavior after the initial elastic behavior with a hardening rate that is higher than in the previous tests with striker velocity 3.4 m/s. We do not see any identifiable soft behavior in the stress-strain curve. We also observe that the specimen shows little change in transparency, unlike the tests at 100 s^{-1} . There is some variability due to variations in batches and tests: at 7 milliseconds, we find an average strain of 2 with 0.3 standard deviation and average stress of 9.6 MPa with 0.7 MPa standard deviation across seven specimens.

Discussion Comparing across all experiments, we conclude that the stress response is strain rate dependent, with the peak stress increasing with increasing strain rate. These are consistent with the finding of [22] and [21] who tested LCEs at much small rates (10^{-4} s^{-1} to 1 s^{-1}). We also conclude that the plateau behavior of the polydomain-to-monodomain (PMT) regime, as well as the onset of this regime, are affected by the strain rate. The limited softness at high strain rates can be attributed to the timescale associated with domain reorientation, again consistent with previous observations at lower rates [20–22, 33].

Compression

The behavior of I-PLCE pads (4.2) in compression is studied at three different strain rates of 0.01 s^{-1} , 8000 s^{-1} , and 19500 s^{-1} .

Quasistatic tests To avoid buckling under compression, we test LCE pads with a 10 mm diameter and 3 mm thickness. The quasistatic response of I-PLCE pads at a strain rate of 10^{-2} s^{-1} is shown in Figure 4.8. The stress increases nonlinearly with strain, reaching up to 35 MPa at a strain of 0.55. We do not see any soft behavior,

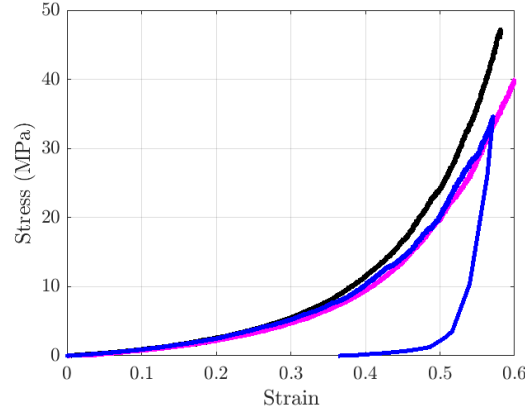


Figure 4.8: Behavior of LCE pads in compression at quasistatic strain rate of $\dot{\epsilon} = 10^{-2} \text{ s}^{-1}$.

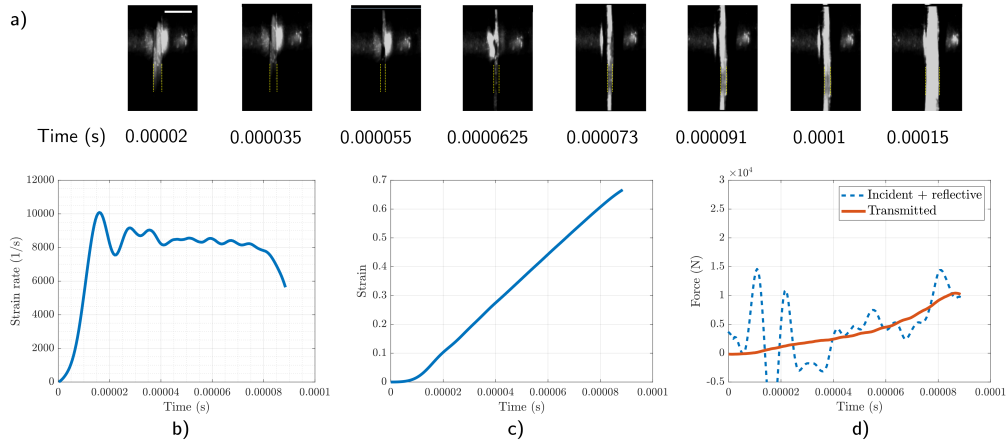


Figure 4.9: Typical results of a test at a nominal strain rate of 8000 s^{-1} . (a) Snapshots showing the specimen being compressed between the incident and the transmitted bar (scale bar is 3 mm). (b) Axial strain as a function of time. (c) Strain rate as a function of time. (d) Force acting on the specimen due to the incident and reflected pulse, as well as the transmitted pulse, as a function of time.

consistent with the observations of Saed *et al.* [8]. Note that unloading was only performed in the test marked with blue solid line.

Tests at 8000 s^{-1} We test LCE pads with a 6.7 mm diameter and 3 mm thickness. Typical results of a test performed at a nominal strain rate of 8000 s^{-1} are shown in Figure 4.9. Figure 4.9(a) shows a series of snapshots of the specimen being compressed between the incident and the transmitted bar of the SHPB apparatus. We observe that the specimen is compressed as the wave arrives and until the incident bar separates from the specimen. There are, in fact, several impacts (see

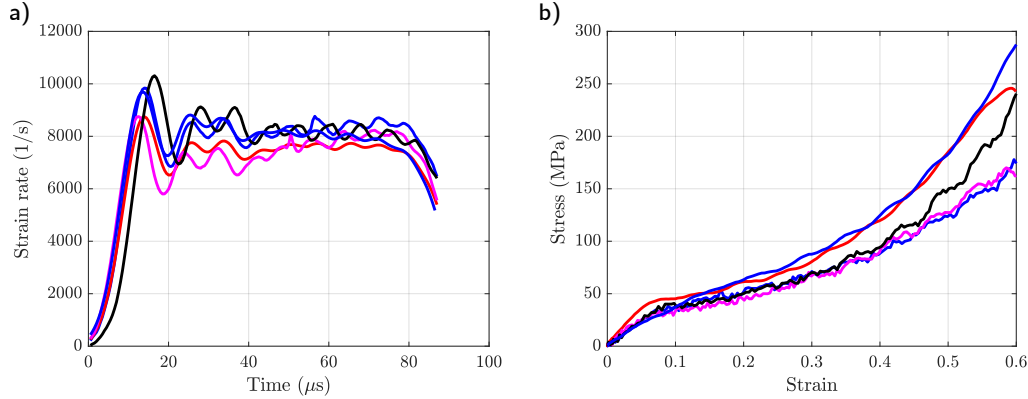


Figure 4.10: The observed response of the I-PLCE in compression at a nominal strain rate of 8000 s^{-1} . (a) Strain rate evolution in time and (b) Stress and strain response.

video in Supplementary Materials) due to the reverberations of the incident bar, and the material begins to disintegrate (fragment).

We only consider the measurements from the time between the arrival of the incident wave and the first separation of the incident bar. Figure 4.9(b) shows the axial strain inferred from the measurements in the incident and transmitted bars as a function of time. Figure 4.9(c) shows the forces on the specimen at the specimen-bar interface (input and output bar) as a function of time. We see that, after an initial transient, the two forces are balanced, indicating that the specimen is in dynamic equilibrium.

The strain rate as a function of time, as well as the stress-strain behavior, over a number of tests is shown in Figure 4.10(a) and (b). We observe that the strain rate is almost constant during the test. We also observe that the stress increases in a nonlinear fashion to reach approximately 100 MPa at a strain of 0.5, and these values are significantly higher than those in the quasistatic test. We again do not observe any evident soft behavior.

There is again some variability in response across specimens. At 21.5 milliseconds, the average strain is 0.1 with 0.013 standard deviation and the average stress is 40 MPa with 4.5 MPa standard deviation, while at 42 microseconds, the average strain is 0.25 with 0.03 standard deviation and the average stress is 77 MPa with 13 MPa standard deviation across five specimens.

Tests at 19500 s^{-1} We test LCE pads with a 6.7 mm diameter and 0.6 mm thickness. The tests are repeated with a higher striker velocity. As before, there are several impacts, and the samples disintegrate. Indeed, the specimen disintegrates into

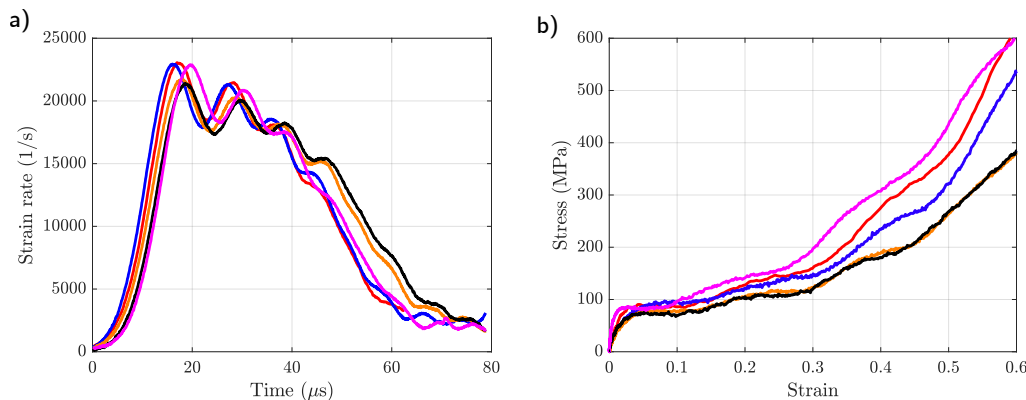


Figure 4.11: The observed response of the I-PLCE in compression at a nominal strain rate of 19500 s^{-1} . (a) Strain rate evolution in time and (b) Stress and strain response.

small charred fragments, and this is accompanied by a strong smell characteristic of toluene. We deduce that the generation of heat during repeated impact, as well as the friction between the specimen and the bars ignite the highly flammable toluene (autoignition temperature of pure toluene is 536°C [34]).

We only use data from before the first separation. The strain rate as a function of time and the stress-strain behavior, over a number of tests, are shown in Figure 4.11(a) and (b) respectively. We observe that the strain rate is not constant over time (in contrast to the previous set of tests at 8000 s^{-1}). The stress is even higher than that at 8000 s^{-1} , reaching 175 MPa at 0.5 strain across five specimens.

The stress-strain curve shows some apparent plateau behavior at small strains. This could be interpreted as soft behavior. However, recall from the test at 8000 s^{-1} in Figure 4.9(b), that the specimen is not at equilibrium in the early part of the test. This transient dynamic effect is larger at tests at $19,500 \text{ s}^{-1}$. Therefore, the observed behavior at small strains is likely due to a dynamic transient effect rather than soft behavior.

As before, there is variability across specimens. At 16 milliseconds, the average strain is 0.1 with 0.03 standard deviation, and the average stress is 93 MPa with 26 MPa standard deviation while at 23 microseconds, the average strain is 0.25 with 0.03 standard deviation, and average stress is 146 MPa with 40 MPa standard deviation.

Discussion We do not find evidence of soft behavior in our tests, including the quasi-static test, consistent with previous observations [8]. Further, since the spec-

imens disintegrated on impact, it is not possible to visually examine the opacity change in the specimen that can indicate any domain reorientation. Furthermore, we recall that the soft behavior in compression is expected to be significantly smaller than in tension: in the ideal case, we expect the plateau strain to be $1 - r^{1/12}$ in compression as compared to $r^{1/3} - 1$ in tension, where $r > 1$ is the anisotropy parameter. This can be further reduced due to lateral confinement.

The contact conditions between the specimens and the bars can affect the results significantly. We applied petroleum jelly to reduce the friction. Still, the geometry of our specimens (the thickness of the specimen is significantly smaller than its diameter) and the tackiness of LCEs make it inevitable that there is some slip and friction. The images shown in Figure 4.9 suggest the presence of slip, but they do not provide sufficient resolution to quantify the amount of slip. The friction contributes to lateral confinement, as does inertia in the pad geometry.

The friction, the geometry, the dissipation in the material due to domain evolution, and viscosity all lead to a temperature increase. We can obtain an order of magnitude estimate of the temperature increase by considering the energy deposited during impact (the area below the stress-strain curve) and assuming adiabatic conditions and negligible elastic response so that all of the work is converted to heat. Approximating the stress-strain behavior to be linear, we get that

$$\Delta T = \frac{1}{2c} \sigma_{\max} \epsilon_{\max} \quad (4.28)$$

where c is the specific heat per unit volume, $\sigma_{\max}$ the peak stress and $\epsilon_{\max}$ the highest strain. The specific heat of LCEs is $1.57 \text{ J/g}^\circ\text{C}$ and density is 1.25 g/cm^3 , and so $c = 1.93 \times 10^6 \text{ Pa/}^\circ\text{C}$ ($\text{J/m}^3^\circ\text{C}$). Taking the peak stress to be 200 MPa and 500 MPa for 8000 and 19500 s^{-1} cases respectively, and a maximum strain of 0.6, gives us a temperature rise of 30.3°C and 75.8°C for the 8000 and 19500 s^{-1} cases. This is, of course, an overestimation, since we assume that all work done is converted to heat. Alternatively, we can use equation (4.16): using the parameters in Table 4.2 under the assumption of uniaxial compression, we obtain a temperature increase of 26 and 40 $^\circ\text{C}$ temperature rise in the 8000 and 19500 s^{-1} cases. These, by themselves, are not high enough to lead to the self-ignition of toluene. However, these ignore friction that would add to the temperature increase. Further, there are multiple impacts, fragmentation of the specimen, and possible hot spots due to heterogeneity or collapse of a microvoid. In fact, a single hot spot reaching the autoignition temperature of toluene could be sufficient to initiate the whole reaction.

Parameter	Value
Shear modulus (equilibrium), μ_1	4.5 MPa
Shear modulus (nonequilibrium), μ_2	1.8 MPa
Bulk modulus (equilibrium), κ_1	0.025 GPa
Bulk modulus (nonequilibrium), κ_2	1.3 GPa
LCE anisotropy parameter, r	6.0
Hardening coefficient, C	100 Pa
Hardening exponent k	2
Kinetic Δ coefficient, α_Δ	$5.0 \times 10^3 (\text{Pa} \cdot \text{s})^{-1}$
Kinetic Λ coefficient, α_Λ	$5.0 \times 10^1 (\text{Pa} \cdot \text{s})^{-1}$
Hydrostatic Viscosity, ν_h	$1.0 \times 10^5 \text{ Pa} \cdot \text{s}$
Deviatoric Viscosity, ν_d :	
Creeping viscosity, ν_0	$1.0 \times 10^5 \text{ Pa} \cdot \text{s}$
High rate viscosity, ν_∞	$3.0 \times 10^6 \text{ Pa} \cdot \text{s}$
Reference shear rate, $\dot{\gamma}_0$	$3.71 \times 10^2 \text{ s}^{-1}$
Viscosity exponent, m	2.9
The deviatoric viscosity takes the following values at the various uniaxial (normal) strain rates	
$\dot{\epsilon} = 10^{-2} \text{ s}^{-1}$,	$1.0 \times 10^5 \text{ Pa} \cdot \text{s}$
$\dot{\epsilon} = 100 \text{ s}^{-1}$	$1.35 \times 10^5 \text{ Pa} \cdot \text{s}$
$\dot{\epsilon} = 300 \text{ s}^{-1}$	$7.7 \times 10^5 \text{ Pa} \cdot \text{s}$
$\dot{\epsilon} = 8000 \text{ s}^{-1}, 19500 \text{ s}^{-1}$	$3.0 \times 10^6 \text{ Pa} \cdot \text{s}$

Table 4.2: Parameters used for fitting with tensile and compression experiments.

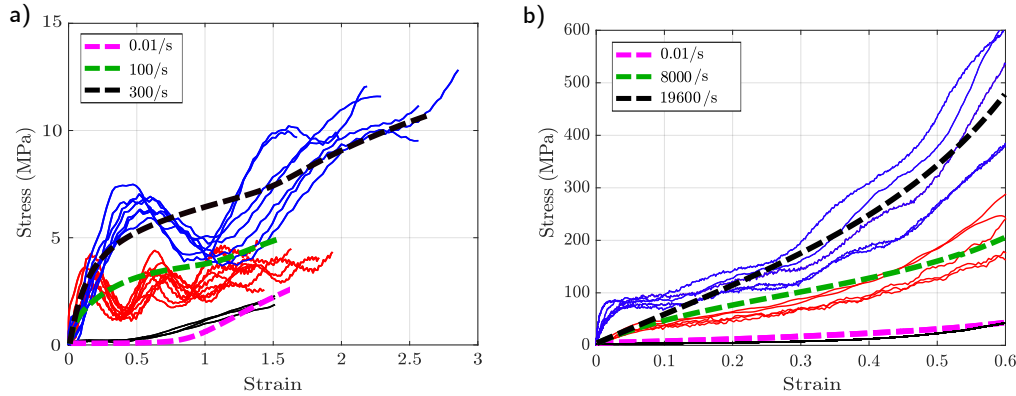


Figure 4.12: Fit of the constitutive model presented in Section 4.3 to the experimental data in (a) tension and (b) compression. Parameters are given in Table 4.2.

Finally, it is possible to have bar-to-bar contact in the thin 0.6 mm specimens used for the 19500 s^{-1} case.

Comparison between model and experiments

Fitting the constitutive model We examine the ability of the macroscopic model outlined in Section 4.3 to describe the experiments. We use a material point calculation assuming homogeneous deformation to model each test. We neglect inertia. We enforce the constraint (4.3) using a predictor-corrector approach with nearest-point projection.

We model the tensile tests by assuming a state of uniaxial stress and isothermal conditions in light of the geometry. Specifically, we assume that the deformation gradient is the form $F = \text{diag}(\lambda_1, \lambda_2, \lambda_2)$, and that the Piola-Kirchhoff stress is $P = \text{diag}(\sigma, 0, 0)$ in an orthonormal basis where the first direction is aligned with the length of the specimen. This ignores the effect of the grips, but we believe that uniaxial stress is reasonable given the length of the specimen and our DIC observations. Further, given the small thickness and large area, we believe that isothermal is a reasonable assumption.

We model the compressive tests by assuming a state of uniaxial strain in light of the small thickness of the specimen as well as the contact with the grips. As discussed above, there is some slip between the specimen and the bar. Therefore, a full model should take this as well as friction into account. We also ignore the transient and inertial effects. We could consider these following full field inversion approaches, but this is beyond the scope of the paper. So, we have to either choose a state of uniaxial stress or uniaxial strain to proceed. We chose the latter. Specifically we assume that $F = \text{diag}(\lambda, 1, 1)$. We also assume isothermal conditions, and this is a significant assumption due to the temperature increase discussed above.

We fit the constitutive constants progressively. We fit the anisotropy ratio, the equilibrium shear modulus, the hardening coefficient and exponent, and kinetic coefficients from quasi-static tension. We then use the intermediate strain rate experiments to fit the non-equilibrium shear modulus and creeping viscosity. Finally, we fit the high rate viscosity, the hydrostatic viscosity, and bulk moduli from the compression tests. One could use emerging regression and optimization techniques, but we have not done so. The resulting parameters are given in Table 4.2, and the comparison between the experimental observations and model results with the chosen parameters are shown in Figure 4.12. We emphasize that we use the same set of parameters in the comparison in Figure 4.12. We note that the agreement is very good.

We have a high degree of confidence in the modeling of the quasistatic and dynamic tension tests, and thus the ability of the model to describe processes up to 10^2 s^{-1}

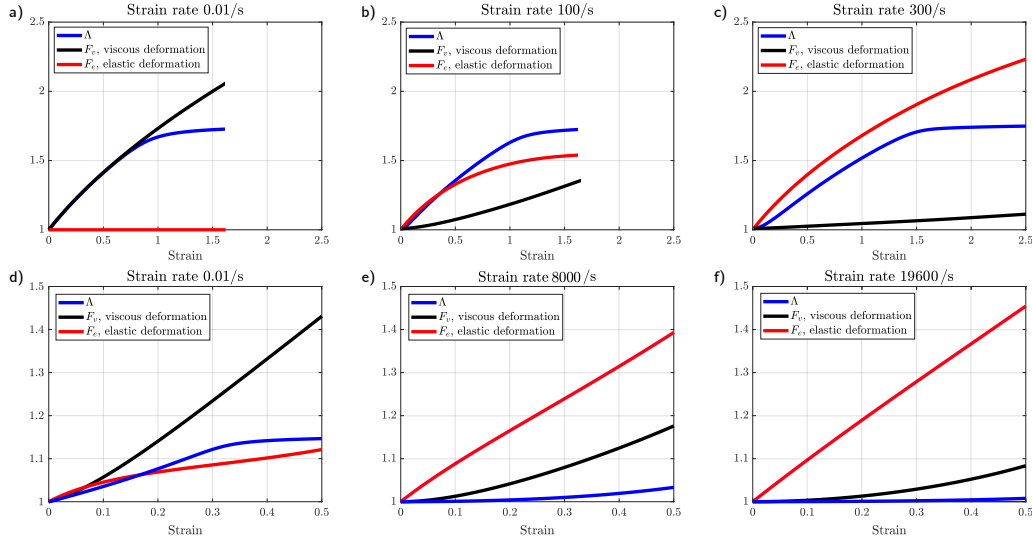


Figure 4.13: The evolution of various internal variables in the model in tension (a-c) and compression (d-f). Parameters are given in Table 4.2.

strain rates. We have less confidence in the compressive due to transients, contact, fragmentation, and temperature increase in the dynamic compression tests, and thus the ability of the model to describe processes in the 10^3 s^{-1} strain rates. Still, it is notable that we are able to describe the entire range of tests and conditions with one single set of model parameters.

Suppression of mesogen and domain reorientation at high rates We finally use the model to gather further insight into the behavior of the materials. We do so by examining the evolution of the internal variable Λ as well as the uniaxial elastic and viscous deformations (largest principal values of F^e and F^v , respectively). This is shown in Figure 4.13(a-c) for the tensile tests. At very small strain rates, we see that the internal variable Λ evolves till close to the value of 1.82 (corresponding to $r^{1/3}$ for $r = 6$) till an imposed strain of about 0.8. We see very little elastic strain. This means that much of the deformation is a result of the mesogen and domain reorientation, resulting in the soft behavior. At higher strain rates, we observe that the evolution of the Λ is retarded while the elastic strain increases rapidly. This indicates that the viscosity masks the mesogen and domain reorientation as observed in the experiments. In compression, the ability of the I-PLCE to undergo domain rotation is already reduced since it changes from isotropic to equi-biaxed. Further, the strain rates are significantly higher, and so viscosity suppresses it further. Figure 4.13(d-f) shows the corresponding evolution in compression, and the response is largely viscous with very little mesogen reorientation.

4.5 Conclusion

In this study, we probe the behavior of isotropic-genesis polydomain liquid crystal elastomers (I-PLCEs) over a wide range of strain rates: 10^{-2} to $3 \times 10^2 \text{ s}^{-1}$ in tension and 10^{-2} to $19.5 \times 10^3 \text{ s}^{-1}$ in compression. Tensile drop tower tests reveal a clear rate-dependent stiffening in the stress-strain response and a limitation of the classical soft elasticity at high rates. Compression split-Hopkinson pressure bar tests, similarly, show a rate-dependent stiffening and limited soft-behavior of I-PLCE pads. These observations indicate that at high strain rates, away from equilibrium, the rate-dependency of the entropic network relaxation is not trivial. We propose a thermodynamically-consistent model that incorporates both the nematic-order evolution as well as polymer viscosity, showing good agreement across all studied strain-rate regimes. Our model captures the coupling of mesogen kinetics and viscoelastic flow across various strain rates as demonstrated by its ability to fit the whole range of tests with a single set of parameters. Therefore, it can serve as a predictive framework for designing I-PLCEs for various applications.

Acknowledgement

We gratefully acknowledge the support of the National Science Foundation (2009289) and the Army Research Office (W911NF-22-1-0269).

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CHARACTERIZATION OF THE SOFT BEHAVIOR OF NEMATIC ELASTOMERS OVER A RANGE OF TEMPERATURE AND STRAIN RATES

A. Kutsyy, A. Wihardja, V. Lee, and K. Bhattacharya, “Characterization of the soft behavior of nematic elastomers over a range of temperature and strain rates”, *European Journal of Mechanics-A/Solids*, 106035 (2026).

Contributions: A.W. participated in the conception of the project, development and validation of the experimental methodology, data analysis and validation, and model analysis. A.W. guided the undergraduate first author throughout the experimental, analytical, and writing stages of the project, and contributed to the writing and revision of the manuscript.

Abstract

Nematic elastomers are a particular class of liquid crystal elastomers (LCEs) that exhibit both liquid-crystalline order and rubber (entropic) elasticity. This combination makes them stimuli-responsive soft materials with a number of unusual thermo-mechanical properties. They have been proposed for various applications, including soft robotics, enhanced adhesion, and impact resistance. This paper presents a new experimental setup and a comprehensive dataset characterizing the soft behavior of nematic elastomers over a range of temperatures and strain rates. We also fit the results to a recently developed model of nematic elastomers [1].

5.1 Introduction

Liquid crystal elastomers (LCEs) are a class of stimuli-responsive soft materials that exhibit both liquid crystalline order and rubber (entropic) elasticity. They are composed of lightly cross-linked polymer chains that have nematic mesogens incorporated into their main or side chains [2]. They can display nematic, smectic, and cholesteric ordering depending on the temperature, material, and crosslinking. In this work, we consider nematic elastomers: at high temperatures, mesogens are

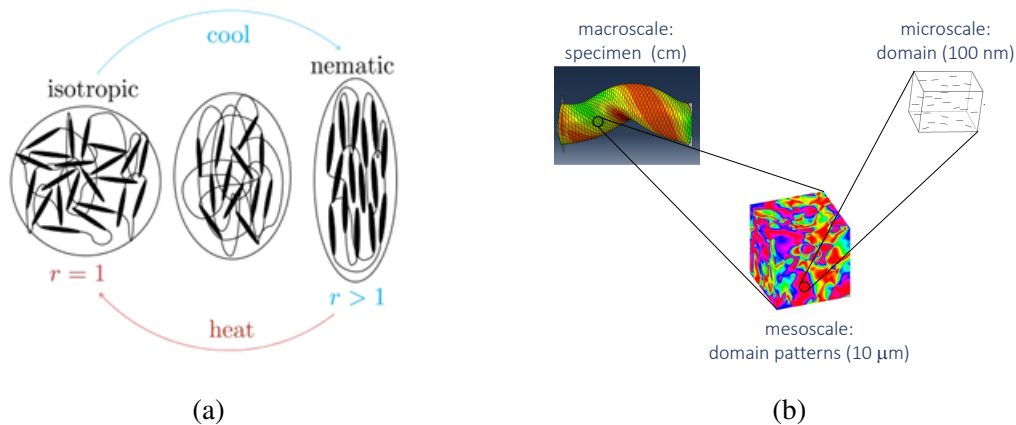


Figure 5.1: (a) The temperature-driven isotropic to nematic transition and the resulting deformation in nematic elastomers. (b) The multiscale nature of I-PLCEs; a typical specimen consists of numerous domains of varying nematic orientation [1].

randomly aligned due to entropy, but undergo an isotropic to nematic transition on cooling, and display nematic ordering where the mesogens are aligned in one direction, as shown in Figure 5.1(a). In particular, we consider isotropic-genesis polydomain nematic LCEs (I-PLCEs) that are synthesized in the isotropic state, but then cooled to a polydomain state where different domains show different nematic ordering directions as shown in Figure 5.1(b).

The combination of entropic elasticity and nematic ordering leads to unusual thermo-mechanical properties in LCEs, and specifically in I-PLCEs. They can also be activated by chemical, optical, electrical, and magnetic stimuli by incorporating appropriate stimuli-responsive molecules that couple to the nematic ordering. Therefore, they have been proposed for a variety of applications, including artificial muscles, soft robotics, drug delivery, stretchable membranes, and damping (see [3–6] for reviews).

An important attribute of I-PLCEs is the soft elastic behavior: the material stretches at almost zero stress, resulting in a soft plateau region in the stress-strain curve [7–10] as shown in Figure 5.2. This is attributed to the polydomain-monodomain transition (PMT): the material, as synthesized, is polydomain, but the mesogens rotate and the domain patterns change until all the domains and mesogens are aligned with the principal stress. This PMT-induced soft behavior results in reduced wrinkling in thin sheets [11], high energy absorption in impact [12], surface instabilities [13], in-plane liquid-like phenomena [14], anomalous behavior in contact [15], and

enhanced adhesion [16–18]. The complex behavior of I-PLCEs has been described in a constitutive law by Lee *et al.* [1], which has been used in finite element analysis to study in-plane liquid-like behavior [1], contact [15], adhesion [18], amongst others.

There has been considerable effort to characterize the thermo-mechanical behavior and properties of polydomain LCEs, both I-PLCEs synthesized in the isotropic state and N-PLCEs synthesized in the nematic state. The behavior of N-PLCEs at various strain rates at room temperature, and across temperatures ranging from room temperature to the transition temperature at a low strain rate is examined in Azoug *et al.* [19]. In this work, however, the soft-elasticity behavior in N-PLCEs is limited by the polymer network’s entropic constraints. Still, they exhibit a PMT regime, which is found to depend on both strain rate and temperature. I-PLCEs with low cross-link density have been studied in Linares *et al.* [20], demonstrating the characteristic soft-elasticity region at different strain rates and mesogen alignment order parameters. However, temperature effects are not probed. This work found that the plateau stress and soft-elasticity regime vary strongly with strain rate, over the range 10^{-2} to 10 s^{-1} . In both of these studies, however, the cross-link density is kept fixed. Recently, Traugutt *et al.* [21] examined both I-PLCEs and N-PLCEs across multiple cross-link densities, showing that the PMT regime and soft behavior in both types are significantly affected by cross-link density. All of the above work concerns main-chain PLCEs synthesized via the Michael addition click reaction as described in [22]. Other studies by Terentjev and collaborators [23, 24] have explored side-chain polydomain nematic elastomers and their temperature dependence from room temperature to the phase transition temperature at a low strain rate of 10^{-4} s^{-1} [23] and strain-rate dependence at low rates (10^{-4} to 10^{-7} s^{-1}) [23, 24].

While these works have provided significant insight and have motivated comprehensive constitutive models [1, 25, 26], the increasing engineering applications [3–6] and the complexity of these constitutive models call for a comprehensive dataset over a range of temperatures, strain rates, and cross-link density. This motivates the current work. We describe a new experimental setup and a comprehensive dataset characterizing the soft behavior of isotropic-genesis polydomain nematic elastomers over a range of temperatures and strain rates. We also fit the results to a recently developed model of nematic elastomers [1].

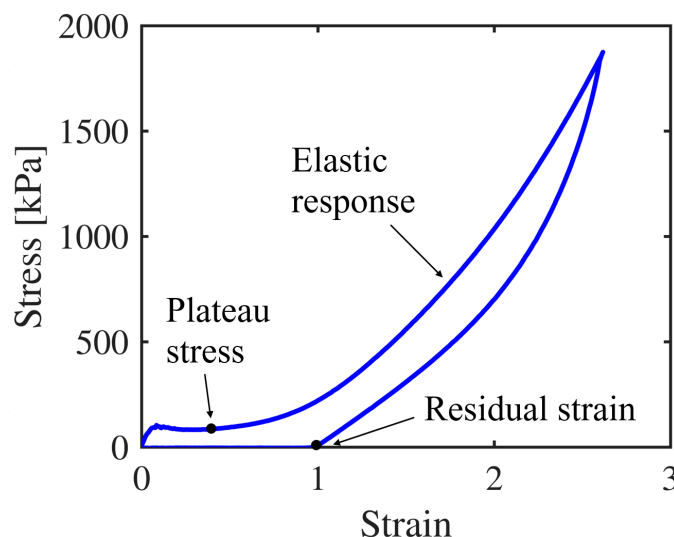


Figure 5.2: Stress-strain curve demonstrating LCE soft behavior. Plateau stress, elastic response, and residual strain are labeled.

5.2 Experimental methods

Experimental setup

The experimental setup used to perform tensile tests at various strain rates, temperatures, and cross-linker densities is shown in Figure 5.3. It consists of a heated chamber with a window, a linear translation stage, an imaging system, various controllers, and a data acquisition system.

The chamber (see Figure 5.3c) is insulated on the sides and equipped with heaters and resistance temperature detector (RTD) sensors. The temperature of the chamber is controlled using an Omega CS8DPT Benchtop controller. The maximum deviation over all tests is 8°C, and occurred during the slowest strain rate trial (10^{-3} s^{-1}) at 90°C; typical deviations are much smaller.

The chamber contains a stationary bottom clamp. The specimen is held in the chamber between the stationary bottom clamp and a top clamp that is attached to a pull-rod that emerges from the chamber. The pull-rod is attached to a Physik Instrumente linear stage and controller, and an Omega LC101-50 load cell.

The specimen is illuminated and imaged through the window using a Photron FASTCAM NOVA S12 high-speed camera equipped with a 100 mm Tokina AT-X Pro lens at 2 frames per second and a resolution of 1024 by 1024 pixels for the duration of the test. The images are analyzed using 2D digital image correlation (DIC) (Vic2D, Correlated Solutions, Columbia, SC) with a typical 25 pixel subset size and step size of 1 to obtain the displacement field. The Green-Lagrange strain

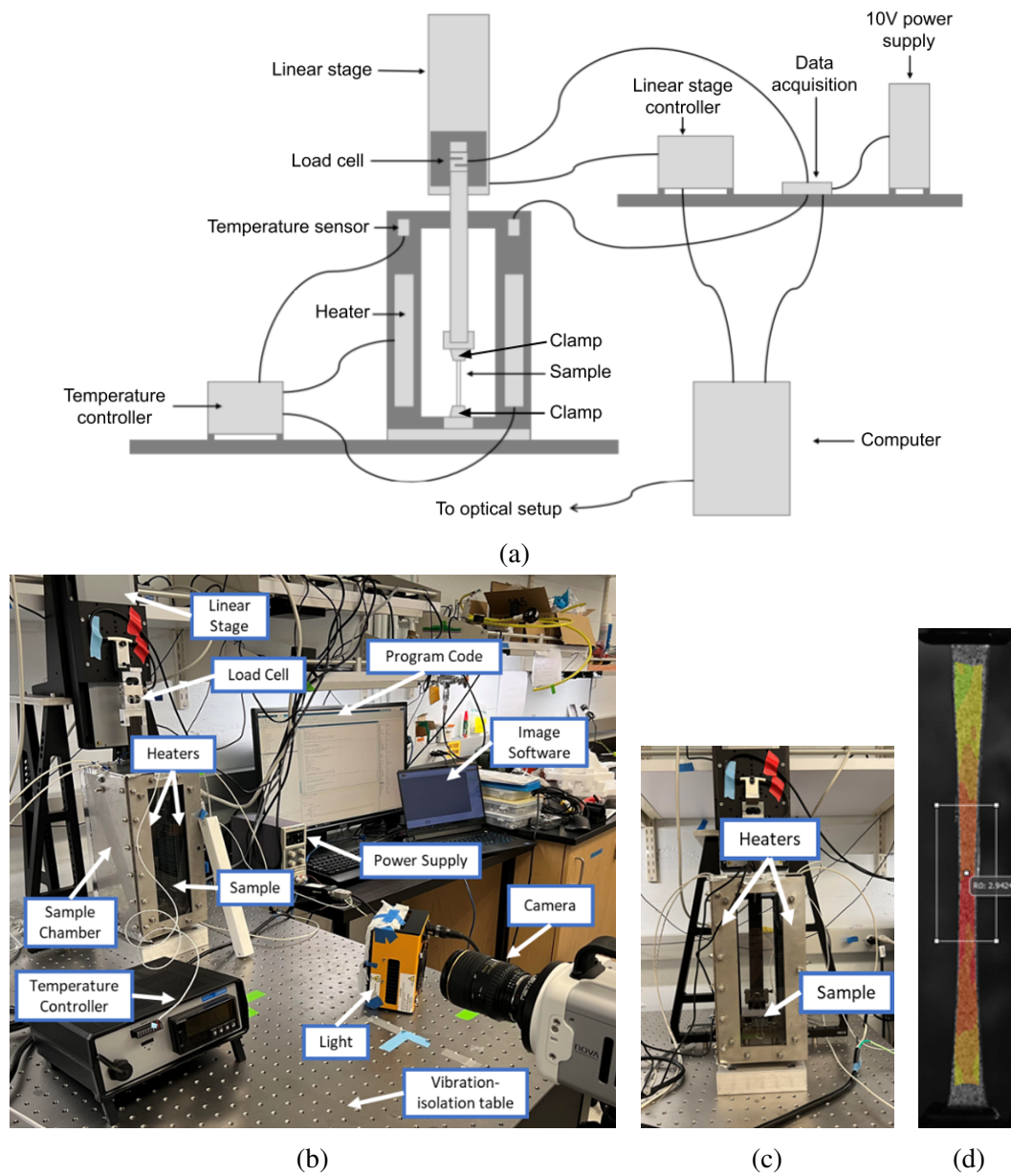


Figure 5.3: The experimental setup and typical result. (a) Schematic diagram, (b) the overall setup (c) the heated chamber, and (d) a typical DIC result.

tensor ($E = \frac{1}{2}(F^T F - I)$ where F is the deformation gradient) is calculated with DIC. A typical strain map is shown in Figure 5.3d. We observe that the deformation is largely uniform across the specimen with no stripes or other almost periodic oscillations. There is some non-uniform strain near the grips due to the shape of the specimen and the effect of grips. We find good correlation for the 50 mol% specimens, but not the 25 mol% specimens, due to the large strains in the latter. Therefore, we use the imaged strain in the gage section for the 50 mol% specimens, but calculated strain from the clamp-to-clamp distance in the 25 mol% specimens. The stress and strain values are correlated using their first respective non-zero value.

Liquid Crystal Elastomer Synthesis

We study polydomain, isotropic-genesis LCEs (I-PLCEs) fabricated using a Michael addition click reaction for main-chain LCEs according to Saed *et al.* [27]. We synthesize I-PLCEs with 50 mol% and 25 mol% cross-linker density (indicating the molar ratio of thiol functional groups between PETMP and EDDET). We focus on these higher concentrations to minimize the network viscosity and focus on the soft behavior due to domain reorientation, consistent with previous studies [14, 21, 27]. Others have used lower densities to probe network viscosity [19], or used second stage crosslinking [20]. Toluene is used to dissolve the di-acrylate mesogens (RM 257) at 80°C until a clear solution emerges, and then the spacer (2,2-(ethylenedioxy) diethanethiol) and crosslinker (pentaerythritol tetrakis) are added. The catalyst solution (dipropylamine) diluted in Toluene is then added to the mixture. The solution is mixed and vacuumed to remove any air bubbles. The resulting solution is poured into molds and cured at room temperature before being heated in an oven at 80°C under a 71.1 kPa vacuum for 12 hours to complete the polymerization at the isotropic phase. Samples are cooled, unmolded, and then heated briefly to remove any stretch. All tests are performed within 2-14 hours after samples are fully cured. The length of the tested specimens is 0.05 m, with a thickness of 0.0015 m. The width of the specimen varies from 0.012 m to 0.005 m in the gauge section. We study the resulting I-PLCE from this method with differential scanning calorimetry (DSC) and recover a nematic to isotropic phase transition temperature (T_{ni}) of 87 °C.

5.3 Experimental observations

We probe the behavior of I-PLCEs at four strain rates (5×10^{-2} , 10^{-2} , 5×10^{-3} , 10^{-3} s⁻¹), three temperatures (26°C or room, 55°C or intermediate, 90°C or high), and two

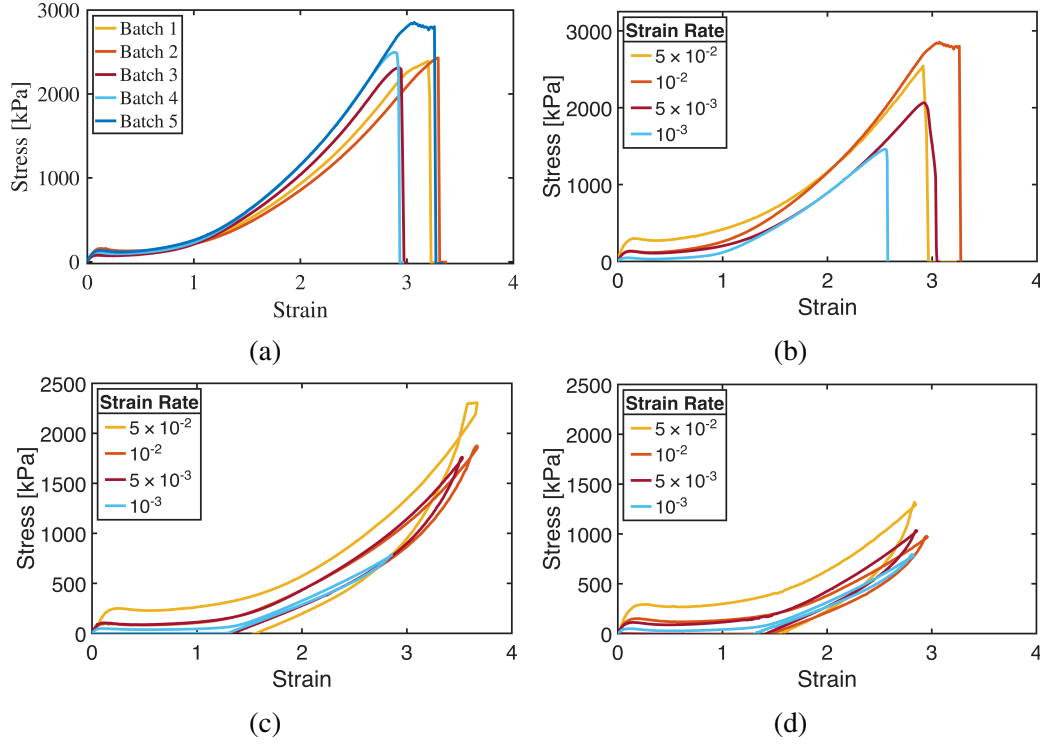


Figure 5.4: Room temperature tensile tests for 50 mol% cross-linker density. (a) Test to failure for a number of batches at the same 10^{-2} loading rate, and (b) for various loading rates. (c) Load-unload tests for the first batch tested to a maximum strain of 3.5, and (d) load-unload tests for the second batch tested to a maximum strain of 3.0.

Table 5.1: Maximum nominal (clamp-to-clamp) strains for load-unload curves.

Strain Rate	$5 \times 10^{-2} \text{ s}^{-1}$	10^{-2} s^{-1}	$5 \times 10^{-3} \text{ s}^{-1}$	10^{-3} s^{-1}
RT or 26°C	2.625, 2.031	2.625, 2.031	2.625, 2.031	2.031
55°C	0.861	0.861	0.625	0.625
90°C	0.48	0.345	0.345	0.345

cross-linker densities (50 mol%, 25 mol%). The nematic to isotropic transformation temperature T_{ni} for the specimen is measured to be 87°C using differential scanning calorimetry. Therefore, two of the temperatures are below the nematic-isotropic transition temperature T_{ni} , and one is above it. Unless otherwise noted, all samples in each figure are from the same synthesis batch, all strains are Lagrangian strains, and all stresses are nominal stresses.

Tests on specimens with cross-linker density 50 mol%

Tensile tests at room temperature (26°C) We first perform tensile test to failure, and the results are shown in Figures 5.4a and 5.4b. We observe in Figure 5.4a that the batch to batch variation is small across trials with the same temperature (RT) and strain rate (10^{-2} s^{-1}). Figure 5.4b shows the rate-dependence, and provides the maximum strain that captures the general behavior without risking failure. We then conduct load-unload tests for strain up to the maximum values, and the results are shown in Figures 5.4c and 5.4d. These are performed for two batches of specimens; each batch is tested at four strain rates, and the two batches are subjected to different maximum stretches, listed in Table 5.1. The overall behavior is consistent across the two batches.

The observed response clearly shows the well-known soft behavior of I-PLCEs [9] marked by a notable plateau in the stress-strain response, and a change in opacity of the specimen. This is a result of the polydomain-monodomain transition (PMT): nematic directors are randomly oriented in the as fabricated polydomain specimens but align along the loading direction, thereby accommodating a larger strain with no increase in stress [8–10]. This change also manifests as a change of opacity: the sample is opaque due to light scattering of the random directors, but becomes transparent as they align. The PMT also manifests as a residual strain, approximately 1.5 across all strain rates and both batches.

We observe small loops at the end of loading and beginning of unloading in some of our tests. These are artifacts caused by two reasons. First, we measure forces using a load cell and strain from raw images processed with DIC. A synchronization error between the two channels of information can lead to these loops. Second, we observe that the specimens with loops develop small out-of-plane motion during the early part of the unloading. There are a few possible reasons: grip effects, some slip at the grips, some temperature gradients at the grips, and the kinetics of reverse transformation. Since we use 2D-DIC to measure displacements and strains, any out-of-plane motion can lead to artifacts in the correlation. In particular, the out-of-plane motion would lead to a smaller apparent strain and manifest as loops.

The PMT and the resulting stress plateau depend on the strain rate: at a strain rate of $5 \times 10^{-2} \text{ s}^{-1}$, the stress plateau is around 235 kPa and the soft behavior extends to approximately 0.9 strain, while at $1 \times 10^{-3} \text{ s}^{-1}$, the stress plateau is around 35 kPa and the soft behavior extends to around 1.2 strain. The lower strain rates offer greater time for the domains and directors, as well as the underlying elastomers, to relax to their preferred orientations [28, 29].

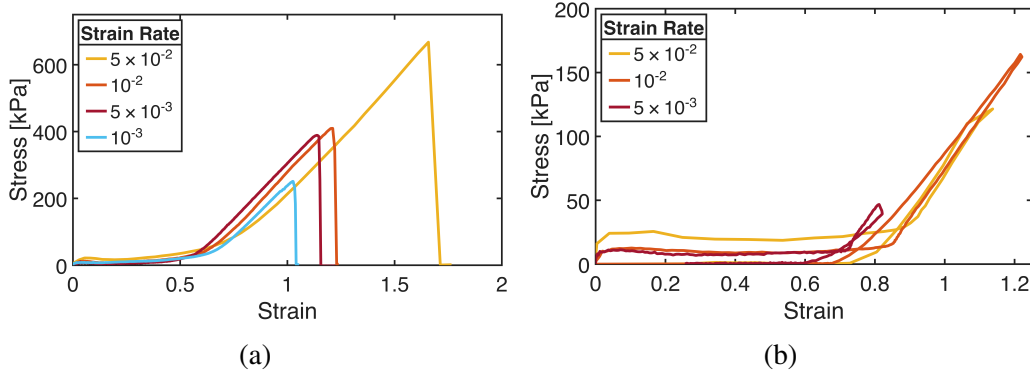


Figure 5.5: 55°C tensile tests for 50 mol% cross-linker density at various strain rates. (a) Tests to failure, and (b) load-unload tests.

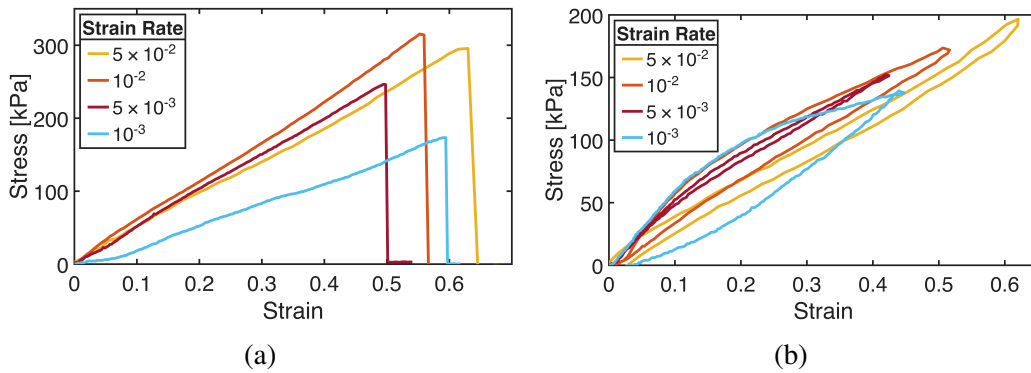


Figure 5.6: 90°C tensile tests for 50 mol% cross-linker density at various strain rates. (a) Test to failure, (b) load-unload tests.

We observe elastic behavior with hardening beyond the plateau as expected for elastomers. It is hard to discern any strain rate effects in our observations.

Tensile tests at 55°C The results of the tests at 55°C are shown in Figure 5.5. The plateau stress increases with strain rate, as seen before, although it is nearly identical between the 10^{-2} s⁻¹ and 5×10^{-3} s⁻¹ strain rates. The extent of the plateau regime is approximately 0.8 strain, and the residual strain is approximately 0.7. As in the case with room temperature tests, we attributed the loop seen in our measurements due to misalignment in synchronization of the stress and strain readings, which were recorded on different systems and synchronized using their first respective non-zero value. Additionally, it may be due to slight out-of-plane deformations, especially near the grips due to temperature gradient.

Tensile tests at 90°C The results of the tests at 90°C are shown in Figure 5.6. We do not observe any stress plateau, and there is negligible residual strain. Since the

test temperature is above T_{ni} of 87 °C, the specimen is in the isotropic state and we do not expect any soft behavior. There are also no noticeable strain rate effects.

Comparison across temperatures We revisit the results above by looking at the effect of temperature at the various strain rates in Figure 5.7. While the tests are conducted with different batches at different temperatures, the batch to batch variation as shown in Figure 5.4a is small compared to the temperature variation (also see Appendix 5.B).

The key aspects of the temperature dependence at a strain rate of $5 \times 10^{-2} \text{ s}^{-1}$ are summarized in Figure 5.8. We observe that the stress and strain at failure decrease with increasing temperature. This suggests that mesogen reorientation provides some toughening as suggested by [14, 24, 30]. We also observe that the plateau and residual strain decrease with increasing temperature, consistent with the decrease of nematic order with increasing temperature. The plateau stress similarly decreases, suggesting that with increasing temperature, the network elasticity likely overshadowed the liquid crystal behavior, resulting in a softer response overall as temperature increases. This finding is consistent with [19, 20, 23], where it was also suggested that the network behavior is more dominant than the mesogen ordering at high temperature due to increased anisotropy, following the proposal of [28].

Cross-linker density 25 mol%

We perform similar tests at room temperature on I-PLCEs with a lower cross-linker density of 25 mol%. These samples show significantly softer behavior, displayed in Figure 5.9. DIC was not performed due to the high strains that result in large distortions of the speckles, and the data shown are based on the data output by the linear stage. The results are qualitatively similar to those described earlier for the 50 mol% specimens, though these specimens are much softer and elongate to much greater strains before failure (for example, 7 vs 3.5 strain at a strain rate of $5 \times 10^{-2} \text{ s}^{-1}$). Figure 5.9c compares the loading curves at 25 mol % and 50 mol % across all strain rates, showing that the difference in crosslinking density significantly impacts plateau stress and strain relative to loading rate. Additionally, the end of the soft behavior or plateau regime is less obvious at lower cross-linker density, as the curve gradually increases rather than having a defined transition region. These results are consistent with previous studies, in which I-PLCEs with various cross-link densities (15 mol%, 25 mol%, 45 mol%) were studied, and similar trends were observed [21]. Since the PMT regime is present in both cross-link densities, we conclude that the

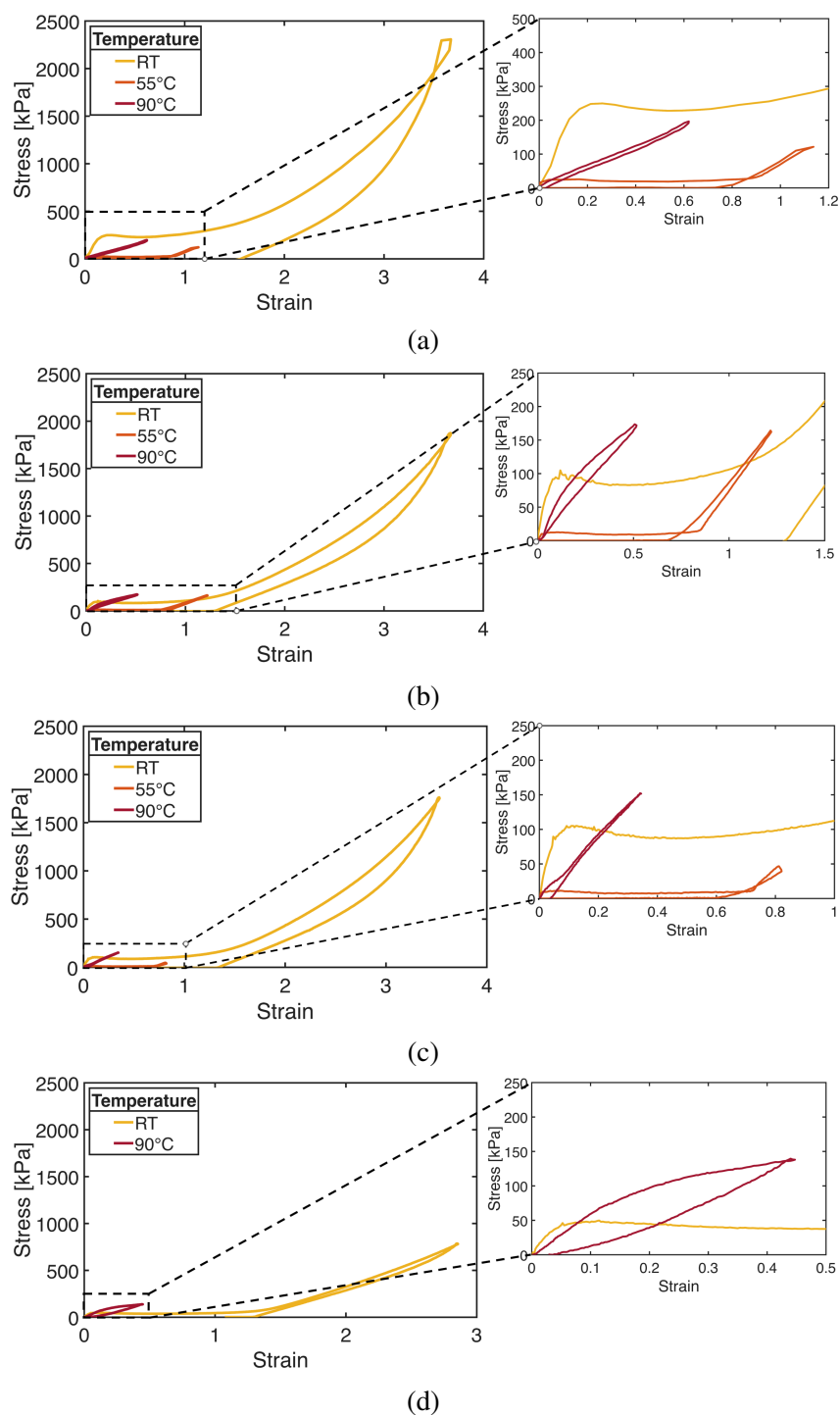


Figure 5.7: Comparison across temperatures for trials with a strain rate of (a) $5 \times 10^{-2} \text{ s}^{-1}$, (b) 10^{-2} s^{-1} , (c) $5 \times 10^{-3} \text{ s}^{-1}$, (d) 10^{-3} s^{-1} .

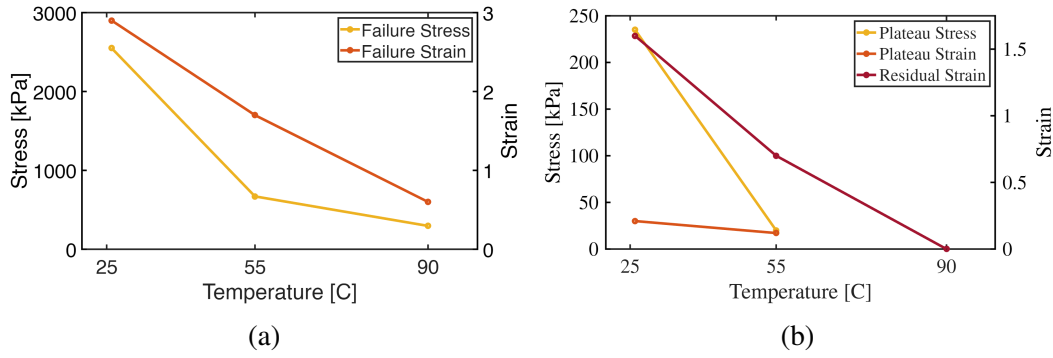


Figure 5.8: Summary of temperature dependence at $5 \times 10^{-2} \text{ s}^{-1}$ nominal strain rate. (a) Failure and (b) Soft behavior. Note that data points above the phase transition temperature are not included, since PMT regime does not exist.

trends across strain rates and temperatures we studied at higher cross-link densities should remain consistent at low cross-link densities.

5.4 Fit to a macroscale constitutive model

We now use the results to validate a model proposed in Lee *et al.* [1]. Briefly, the model incorporates the evolution of microstructure (mesogens) through the internal variable framework of continuum mechanics. These internal variables encode the statistics of the mesogens at the microscopic scale and they capture the microstructure dynamics of the mesogens and domains from a coarse-grained perspective at the macroscopic scale. The model has successfully captured the behavior of I-PLCEs under multi-axial loading conditions observed experimentally in biaxial tests [14], Hertzian contact [16, 17], and adhesion [18] in the quasi-static setting. In the dynamic setting, it has been extended to incorporate network viscosity and has successfully captured the behavior of I-PLCEs under impact loading [31].

To capture microstructure dynamics in the macroscopic scale, the model introduces two internal variables (Λ, Δ) that describe the domain patterns: Λ serves as a measure of the degree of alignment of all mesogens along the principal stretch direction in any representative volume element (RVE) while Δ serves as a measure of the degree of planarity of all mesogens in the RVE. The model assumes isotropy and incompressibility, and thus the deformation is characterized by the principal stretch λ and the product of the two largest principal stretches δ . The stored energy density is postulated to be

$$W = \frac{\mu}{2} \left(\frac{\lambda^2}{\Lambda^2} + \frac{\delta^2 \Lambda^2}{\lambda^2 \Delta^2} + \frac{\Delta^2}{\delta^2} - 3 \right) + C \frac{\Delta - 1}{(r^{1/6} - \Delta)^k} \quad (5.1)$$

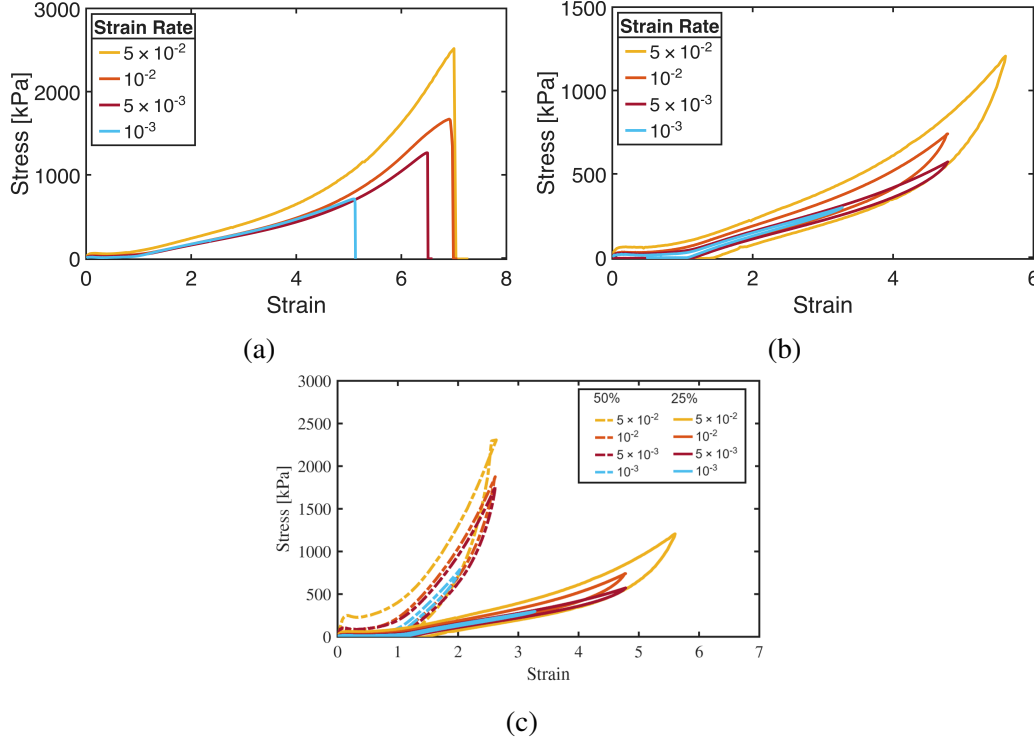


Figure 5.9: Room temperature tensile for 25 mol% cross-linker density at various strain rates and comparison. (a) Test to failure, (b) load-unload tests, and (c) comparison between the 25 mol% and 50 mol% cross-linker densities showing all four loading rates

where r is the so-called anisotropy parameter that describes the degree of nematic order, μ is the shear modulus, and C is a hardening coefficient, and k is a hardening exponent. The first term in the energy is the elastic energy of the polymer network accounting for the spontaneous deformation of the domains, while the second term describes the memory of the domains and their energetic cost to align, as well as the saturation of the internal variables at perfect alignment state. In particular, this contribution is small near the isotropic case but increases as domain patterns evolves to be fully ordered in the plane, due to incompatibility from neighboring domains. Given the energy form above, the Piola-Kirchhoff stress is

$$S = -pI + \frac{\partial W}{\partial F} \quad (5.2)$$

where p is the constitutively indeterminate hydrostatic pressure. The evolution laws of the internal variables are taken to be linearly dependent on their thermodynamic driving forces, hence

$$\dot{\Lambda} = -\alpha_{\Lambda} \frac{\partial W}{\partial \Lambda}, \quad \dot{\Delta} = -\alpha_{\Delta} \frac{\partial W}{\partial \Delta}. \quad (5.3)$$

The coefficients α_Δ and α_Λ determine the rate of change of the mesogen orientation. Larger values correspond to faster kinetics. Further details are found in [1].

We note that the model focuses on domain reorientation, and therefore ignores network viscosity. This is consistent with our experiments with high crosslink density. Still, the model is rate-dependent through these evolution equations 5.3. We refer the reader to [31] for the extension of this model to include network viscosity and high strain rates.

We also note that the model is isothermal, though the parameters depend on temperature. Specifically, we anticipate the anisotropy parameter r that describes nematic order to decrease with increasing temperature, reaching 1 at T_{ni} , the kinetic coefficients $\alpha_\Delta, \alpha_\Lambda$ that control the reorientation of the mesogens to increase with increasing temperature, and the hardening parameter that is associated with nonideality C to decrease with increasing temperature.

The loading portion of one stress-strain curve from each temperature and strain rate pair at 50 mol% cross-linker density is used to fit the model. The resulting parameters are shown in Table 5.2, and the fits in Figure 5.10. Note that all parameters are held fixed across strain rates at each temperature. Given that much of the elastic and nematic behavior is entropic in nature, it is natural to expect temperature dependence. We observe in Figure 5.10 that the model describes the behavior extremely well.

We emphasize that we only use the loading portion of the experiments to fit the model. So the unloading behavior serves as an independent test, and shows good agreement. We observe some offset in the unloading strains, and there are a few reasons. Recall that some of tests had loops during the loading to unloading transition. Further, we observed occasional cracks near the grips at large stretch and also some slippage at the grips. Finally, our tests go to very large strains, well beyond the Gaussian chain assumption of the elasticity in our model. We believe that these are the reasons for the off-set in the experimental observations.

5.5 Conclusion

In this study, we have used a new tensile-testing experimental platform to probe the soft-elasticity behavior in isotropic-genesis polydomain liquid crystal elastomers (I-PLCEs) over a wide range of temperatures (room temperature to beyond the phase-transition temperature) and across various strain rates (10^{-3} to $5 \times 10^{-2} \text{ s}^{-1}$) and cross-linker densities (25% and 50%). Below the nematic-isotropic phase

Table 5.2: Temperature dependent parameters for each model curve shown in Figure 5.10.

Parameter	Room Temperature	55°C	90°C
r	10	5	1
μ (kPa)	2000	2000	90
k	2	2	2
$\alpha_{\Delta}(\text{Pa} \cdot \text{s})^{-1}$	6×10^4	4.5×10^3	1
$\alpha_{\Lambda}(\text{Pa} \cdot \text{s})^{-1}$	$\alpha_{\Delta}/100$	$\alpha_{\Delta}/100$	$\alpha_{\Delta}/100$
C (Pa)	0.4	0.09	0.01

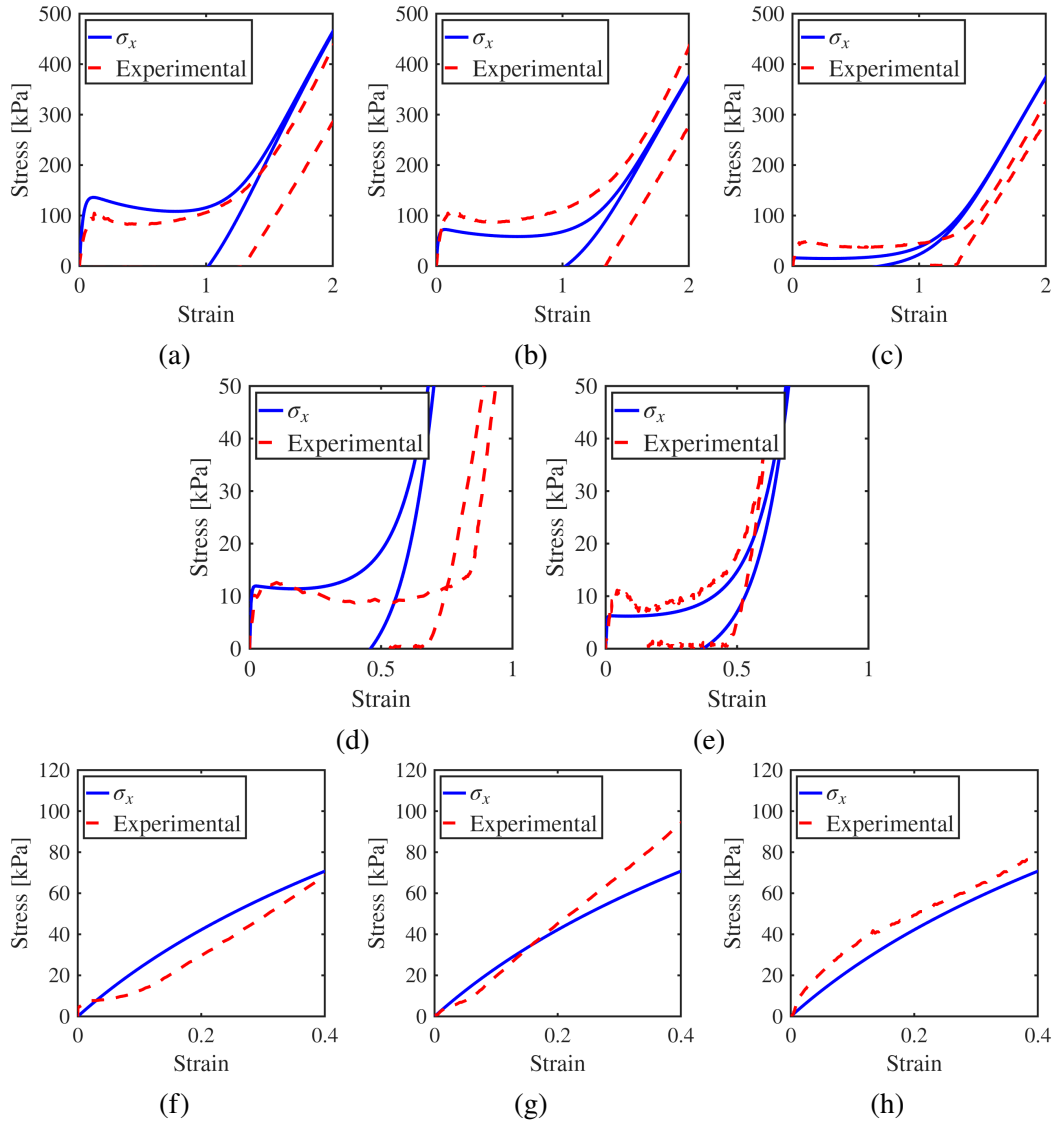


Figure 5.10: Experimental data compared to the model, for each parameter set, listed as temperature, strain rate: (a) RT, 10^{-2} s^{-1} , (b) RT, $5 \times 10^{-3} \text{ s}^{-1}$, (c) RT, 10^{-3} s^{-1} , (d) 55°C, 10^{-2} s^{-1} , (e) 55°C, $5 \times 10^{-3} \text{ s}^{-1}$, (f) 90°C, 10^{-2} s^{-1} , (g) 90°C, $5 \times 10^{-3} \text{ s}^{-1}$, and (h) 90°C, 10^{-3} s^{-1} .

transition, all specimens exhibited the characteristic stress plateau of I-PLCEs, with both plateau stress and strain and residual strain decreasing as temperature increased and strain rate decreased. Above this transition temperature, the plateau regime vanished, and there was no noticeable strain-rate-dependent response of the polymer network. Moreover, reducing the cross-linker density resulted in softer behavior and higher failure strains. We further fit a continuum model that incorporates the microstructural kinetics of the domain patterns to our experiments, showing excellent agreement across all studied temperatures and strain-rate regimes using a single temperature-dependent parameter set. Our experimental results elucidate the coupled effects of temperature, strain rate, and network density on the soft-elastic behavior of I-PLCEs. Furthermore, our model is shown to capture these coupled effects and can serve as a predictive framework for designing I-PLCEs for various applications.

We conclude by noting that while we have probed the material at various temperatures, this work is limited to isothermal applications. It would be of interest to develop a complete thermo-mechanical model that combines systematic calorimetry with mechanical tests at more temperatures. This remains a task for the future.

Conflict of interests

The authors declare that they have no conflict of interests.

Acknowledgement

The authors gratefully acknowledge the financial support of the US National Science Foundation (DMS-2009289) and the US Army Research Office (W911NF-22-1-0269).

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APPENDIX TO CHAPTER 5

5.A Experimental Setup Details

Various parts used in the experimental setup, organized by subsystem, are:

- Custom-made chamber assembly
 - Chamber components (McMaster 1658A12, 8983K128, 8983K118, 2313N23)
 - Various hardware (nuts, bolts, washers, etc.)
 - Various adapters (McMaster 8489K44, 4416T47)
 - Windows and sealing (McMaster 8476K999, 92320A662)
 - Stationary bottom clamp and moving clamp on pullrod (McMaster 12755T82)
 - Moving pullrod (McMaster 8489K46)
- Heating
 - Two heaters (Omega OTF-102/120V)
 - Two RTD air temperature sensors (Omega RTD-805-B)
 - Temperature controller (Omega CS8DPT)
 - 3M High-Temperature Flue Tape
- Extension
 - Linear stage (Physik Instrumente M-531.EC)
 - Linear stage controller (Physik Instrumente C-863.11)
 - Linear stage z-axis mounting bracket (Physik Instrumente M-592.10)
 - Suspension system (ThorLabs VB01B)
- Load
 - Load cell (Omega LC101-50)
 - External 10V power supply
- Optics
 - Vibration-isolation table (ThorLabs T46J)

Table A1: Average stress and standard deviation at various strains across batches at room temperature at a loading rate of 10^{-2} s^{-1} .

Strain	σ_{avg} (kPa)	σ_{std} (kPa)
0.25	104.02	24.66
0.5	137.67	53.64
1	228.26	17.97
1.5	540.90	65.05
2	1021.74	134.44
2.5	1689.20	236.41

- Continuous light source
- Camera (NOVA-S128M)
- Photron FASTCAM Viewer software
- Data acquisition
 - Lab computer (Lenovo ThinkStation P330)
 - DAQ (Omega INET-600)
 - MATLAB software
 - VIC-2D software

Since tests are performed at various temperatures up to 90°C , the pullrod and one of the adapters were made out of ceramic to prevent the load cell from overheating. Binder clips were used to clamp the samples because they are self-tightening, and the inside of the clips were sanded to increase friction between the sample and the clip.

5.B Batch and temperature variation

The average stress and standard variation across various batches tested at room temperature at a loading rate of 10^{-2} s^{-1} is given in Table A1. The mean stress at various temperatures tested at a loading rate of 10^{-2} s^{-1} is given in Table A2.

Table A2: Direct comparison of stress values at specific strains across temperatures. All data is taken from trials with a strain rate of 10^{-2} s^{-1} .

Strain	σ_{avg} at RT (kPa)	σ_{avg} at 55°C (kPa)	σ_{avg} at 90°C (kPa)
0.25	104.02	10.67	86.40
0.5	137.67	8.98	166.20
1	228.26	74.81	

Part II

Data-driven Methods on Model Discovery and Optimal Experiments

*Chapter 6***BACKGROUND: INVERSE METHODS TO DISCOVER
MATERIAL MODEL**

Predicting the behavior of all engineering structures and systems requires material constitutive models that close the governing balance laws. These balance laws capture universal principles of physics that must be satisfied independent of the material. Meanwhile, the constitutive model encodes the material-specific relation between kinematic quantities, such as deformation or strain, and dynamic quantities, such as stress. Therefore, the fidelity of any mechanical simulation and subsequent engineering predictions depends critically on the accuracy of the constitutive model used to describe the material response.

In practice, these constitutive models are calibrated from experimental data to emulate real materials used in applications. This is inherently an inverse problem: to discover a material law, one measures experimental quantities such as boundary displacements, applied forces, and images of the deforming specimen, and then infers the form or the parameters (or both) of the constitutive model that best explains these measured data. However, the quantities in the constitutive model are not directly measurable, i.e., stresses and strains cannot be directly measured. Thus, to infer a material model, one has to reconstruct stresses and strains from the directly measured data as part of the inversion procedure.

In this chapter, we introduce and review the inverse problem of inferring constitutive models from experimental data. We review the currently established practice and inversion methods to obtain material models and discuss limitations associated with the current methods. We then turn to describe a technique called the adjoint method, used in our gradient-based PDE-constraint optimization problems throughout this thesis (Chapters 7,8). We draw comparison between the adjoint method and a closely related, well-established method called the virtual fields method (VFM). We close this background on inverse problems by discussing the integration of the Bayesian perspective to solve inverse problems, which we leverage in Chapter 8 as a quantitative framework to design optimal experiments.

6.1 Constitutive Relations via Direct Inversion

Traditionally, the inverse problem to infer constitutive relations is addressed by designing experiments that produce simple, nearly homogeneous deformation states. Examples include uniaxial tension, compression, shear, or biaxial testing [1–7]. In all these procedures, the specimen geometry and boundary conditions are chosen so that the strain field is approximately uniform within the tested sample. Under this assumption, boundary force measurements can be directly converted into stresses, while imposed boundary displacements can be directly converted into strains. The experiment then yields a single stress-strain data point, which can be used to calibrate a particular constitutive model form.

This approach is extremely robust and successful because of its ability to directly relate measured data to the unknown quantities in the model. However, each experiment is specifically designed to produce a simple deformation mode; many repeated experiments are needed to sufficiently probe the material response to inform the unknown constitutive model. This is a low-throughput, time-consuming process. Another, more severe, limitation is that the resulting models are calibrated primarily under homogeneous deformation states, which is often insufficient to predict complex loading conditions and heterogeneous deformation often encountered in real engineering applications.

6.2 Constitutive Relations via Indirect Inversion with Full-Field Measurements

Advances in digital imaging, optics, and electronics have enabled us to collect full-field data during experiments. In particular, one can obtain full-field images of the tested specimen as it deforms throughout the entire experiment. This creates the potential of leveraging complex, nonuniform deformation states of the specimen to generate more informative measured data that better inform the model inference for complex materials. However, to leverage this information, one has to solve an indirect inverse problem and reconstruct internal states, stresses, and strains as part of the inversion procedure. This is a challenging inverse problem, and current established approaches are discussed in the subsequent subsections.

Current methods to infer displacement from images

Current approaches to solve the material parameter identification problem often divide the inversion into two smaller inverse problems. The first inversion is obtaining displacement from images, followed by the second inversion of obtaining material

parameters from the inferred displacements. The first step is typically performed using image-based approaches, such as speckle interferometry, the grid method, and digital image correlation (DIC), with DIC being the most common choice. DIC is a well-known optical experimental technique to measure displacements in solids and structures [8–10]. In doing DIC, we decorate the surface of the sample with a speckle pattern. We then take a series of images of the decorated surface undergoing deformation and compare them using image-tracking methods to infer the deformation. This approach is popular due to the availability of CCD cameras and many robust commercial [11] and open source software [12].

The problem formulation of DIC is seen in Figure 6.1. We consider a body occupying

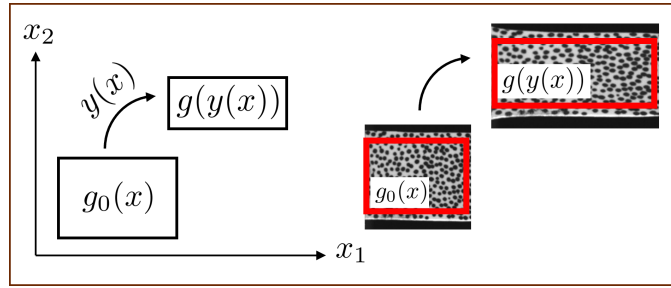


Figure 6.1: Image matching problem of Digital Image Correlation (DIC).

the region $\Omega \subset \mathbb{R}^n$ in the natural reference configuration. The body is undergoing deformation $y : \Omega \rightarrow \mathbb{R}^n$. We denote x as the reference or undeformed position of a particle in Ω and denote $y(x)$ as its current position. After decorating the sample, we have a speckle pattern with grayscale value $g_0(x)$ in the reference domain and the corresponding grayscale value $g(y(x))$ in the current configuration. Supposing the deformation convects with the grayscale, we have

$$g_0(x) = g(y(x)). \quad (6.1)$$

The DIC problem is then in finding the deformation field $y(x)$ satisfying the (6.1). This is posed as an optimization problem of minimizing the speckle mismatch

$$\int_{\Omega} |g_0(x) - g(y(x))|^2 d\Omega \quad (6.2)$$

over all possible deformation solution field, $y : \Omega \rightarrow \mathbb{R}^n$. This is an optimal transport problem, where it is well-known that a solution may not exist or minimizing sequences may not converge. In general, the image matching of the speckles is an ill-posed problem unless additional regularization is introduced in the solution field. Two approaches are common, resulting in the so-called local and global DIC

methods. The local approach [8, 9, 11] involves selecting a grid of points $\{\bar{x}^i\}$ and then assuming that the deformation is piecewise affine within a sub-domain (often called a subset in the local DIC method) around these points

$$y(x) = \sum_i (y^i + F^i(x - \bar{x}^i)) \chi_i(x), \quad (6.3)$$

where χ_i is the indicator function of the i^{th} subset around $\bar{x}^i$. The local DIC method performs correlation within a subset, independent of the neighboring subset. Thus, this local correlation can be performed in parallel, resulting in a fast, scalable determination of the displacement fields. However, since correlation is performed locally, the resulting displacement field may not be compatible, and strain fields are generally not continuous.

On the other hand, the global approach [10, 13] assumes that deformation is spanned by a set of chosen finite-dimensional basis $\{\psi_i\}$,

$$y(x) = \sum_i y^i \psi_i(x). \quad (6.4)$$

Unlike the local approach, the displacement ansatz here leads to a displacement solution that is globally compatible. However, this method often leads to an ill-conditioned system of equations, since the image gradients are often noisy, and the resulting stiffness matrix may be ill-conditioned. Therefore, various regularization methods must be employed to enable robust inference of the global displacement fields. This regularization is arbitrary and often problem-specific and requires specific domain knowledge [14].

Another approach that seeks to combine the advantages of local and global DIC is the Augmented Lagrangian Digital Image Correlation (ALDIC) approach [12, 15, 16]. ALDIC matches subsets locally as in local DIC methods, but enforces compatibility in the deformation field as a constraint. An auxiliary displacement field that is globally compatible is introduced, and this constraint is implemented with the augmented Lagrangian method (also known as the method of multipliers). It augments the objective in (6.1) with a linear constraint term as in the classical Lagrange multiplier method and a quadratic constraint term as in the penalty method, which enables numerical stability and ease of convergence. The augmented objective under the displacement ansatz is discretized as

$$\sum_i \int_{\Omega_i} |g_0(x) - g(F_i(x), u_i(x))|^2 + \frac{\beta}{2} |(D\hat{u})_i - F_i + W_i|^2 + \frac{\mu}{2} |\hat{u}_i - u_i + v_i|^2 d\Omega_i, \quad (6.5)$$

where $\hat{u}$ is the auxiliary displacement fields. For $\bar{x}^i$ the center of subset i , $F_i = \nabla \hat{u}(\bar{x}^i)$ is the local displacement gradient, $u_i = \hat{u}(\bar{x}^i)$ is the local displacement. D is the discrete gradient operator, β, μ are the Lagrange multipliers and penalty terms. This ALDIC method uses the alternating direction method of multipliers (ADMM) to solve for the local correlation and the auxiliary displacement fields.

All of the aforementioned DIC methods yield a solution, but there is no notion of convergence and hence no notion of error as the number of subsets or basis functions goes to infinity. In practice, this means that the inferred displacement solution depends on user-selected parameters such as subset size, interpolation order, number of basis functions considered, and other regularization choices. These choices are often arbitrary and experiment-dependent. Further, it is important to note that the quality of the inferred deformation gets extremely sensitive to these arbitrary regularization parameters in complex, nonuniform deformation fields: near holes, notches, boundaries, or regions with large strain gradients. Thus, DIC tends to lose correlation in these complex regimes. This is not ideal since these complex regions contain the most useful information to inform a constitutive model.

Current identification methods to obtain material parameters from inferred displacements

After obtaining displacement fields from images, one needs to solve the second inversion problem to obtain the appropriate model parameters. This analysis step is often done with identification methods. Several methods exist to infer material parameters based on full-field or boundary measurements, such as the Finite Element Updating Method (FEMU), Virtual Fields Method (VFM), Constitutive Equation Gap Method (CEGM), Equilibrium Gap Method (EGM), and Reciprocity Gap Method (RGM). There exist review papers describing these techniques and a comparative study of this method using experimental data from standard tests [17, 18]. The most popular methods are FEMU and VFM.

Finite Element Updating Method. In Finite Element Updating Method (FEMU) [19, 20], we seek to identify parameters by minimizing the difference between measured and predicted quantities (displacements or boundary forces). This method is flexible and works for many kinds of incomplete measured data. Typically, the predicted quantity is a result of solving a well-posed forward problem, and the minimizer is attained through an iterative procedure. This method has been demonstrated to recover parameters for orthotropic elasticity from uniaxial test measurements

[21, 22], and to recover hyperelastic material parameters [23]. One of the main drawbacks of FEMU, however, is that for N parameters, the forward problem must be solved $2N$ times (typically with finite element methods) at each optimization iteration to compute the sensitivity of the minimization objective with respect to the parameters. This is numerically expensive and easily becomes intractable with a large number of material parameters or for materials with history.

Constitutive Equation Gap Method. Another method is the Constitutive Equation Gap Method (CEGM). In CEGM, we seek to minimize the constitutive mismatch between a statically admissible stress field and a kinematically admissible displacement field. This method is variationally consistent [24] and ensures that equilibrium and compatibility conditions are satisfied. However, to ensure this, one introduces admissible stress and displacement fields and then must solve a nested, alternating optimization problem. The first optimization problem seeks to find admissible fields of stress and displacements from measurements, and afterwards, the second optimization finds the material parameters. Both are usually done via finite element methods. As with the FEMU, this method is flexible and can work for many kinds of incomplete measured data. However, it is costly due to the introduction of more unknowns in addition to the unknown material parameters. It also inherits all problems associated with nested optimization, such as error accumulation and sensitivity mismatch. Further, imposing equilibrium and compatibility on noisy measurements requires care, and the resulting parameters are thus sensitive to noise. A poor choice of interpolation space in the stress or displacement fields to impose admissibility will thus lead to errors.

Virtual Fields Method. A very popular method is the Virtual Fields Method (VFM) due to its fast computation. The VFM is based on the principle of virtual work, where both the full-field displacement measurement and the assumed constitutive law are used to infer the full-field stress. Under the exact constitutive law, the principle of virtual work is then satisfied for any admissible displacement virtual fields. This leads to an optimization problem to minimize the gap [25, 26] (assuming linear elastic, small-strain),

$$\int_{\Omega} \sigma(\epsilon(u^{\text{meas}}); P) \epsilon(v^*) d\Omega - \int_{\partial_t \Omega} \bar{t} v^* dA - \int_{\Omega} b v^* d\Omega = 0, \quad \forall v^* \in \mathcal{U}, \quad (6.6)$$

where v^* is any virtual fields in $\mathcal{U}$, the space of admissible displacement variations. Here, u^{meas} is the full-field displacement measurement, P is the unknown model parameters to be inferred, $\bar{t}$ is the imposed traction over the traction boundary $\partial_t \Omega$, and b is the body force.

The principle of virtual work must be satisfied over the entire body Ω , so full-field stress in the domain is needed. This means that to do VFM, one must have the strain field within the entire domain of the tested specimen. Thus, the samples tested are often under plane stress or strain to enable extension of full-field surface measurements over the entire domain under those plane assumptions. This is an issue in VFM, since the displacement full-field measurements from DIC often lose correlation near sample boundaries, near holes, and notches (again, due to the ill-posedness of the problem). Further, the input to (6.6) is the full-field strain, which is only obtained after differentiating those noisy full-field displacement fields, exacerbating any noise or correlation issues and creating significant uncertainties in the inferred parameters. It is important to note that all of these issues often restrict the use of VFM to simple geometries, which leads us back to having sparse data. This is a major drawback of VFM.

Another disadvantage of VFM is that the quality of the inversion is highly dependent on the chosen virtual fields. This choice is user-dependent and arbitrary, with some virtual fields being better than others, and poor choices can lead to ill-conditioning during inversion to obtain parameters. Choosing suitable virtual fields is much less straightforward in complex materials, such as those with history (e.g., elastic-plastic materials), where (6.6) now needs to be satisfied incrementally, at each loading step and one must choose virtual fields at each loading step, [27]. This can be intractable and require domain knowledge. Importantly, there is high sensitivity to noise in measurements for each loading, leading to accumulation of errors in this incremental approach. Therefore, VFM is not a robust approach to learning complex materials.

One advantage of VFM is that a finite element analysis is not needed, and the integration of measurement data directly in (6.6) results in a non-iterative method. However, there is an iterative aspect of this method, which comes from constructing suitable virtual fields. Due to the sensitivity of the results with respect to this choice and the noise of the full-field measurement, several methodologies have been developed to iteratively solve for the most optimal virtual fields that eliminate measurement noise, are sensitive to the parameters, and make the resulting linear equation from (6.6) well-conditioned [26, 28]. To address this issue, piecewise virtual fields have also been explored [29]. This gives rise to the Equilibrium Gap Method (EGM).

Reciprocity Gap Method. Lastly, another approach that is typically chosen if measurements are available on the boundary is the Reciprocity Gap Method (RGM).

This method is based on the Maxwell-Betti reciprocity theorem. Here, one first sets up a trial set of admissible elastic parameters and their corresponding adjoint state of stress and strain. Then, given known/measured boundary displacement and tractions, a minimization problem of the reciprocity gap between the real material and the adjoint state is solved [30]. This method is also limited to linear elastic materials. A linearized approach has been developed for nonlinear materials.

The performance comparison of the methods discussed above in experiments with standard specimens and tests has been done by Avril *et al.* in a review [17]. From their study, it is clear that the inverse problem solution is sensitive to error sources in the data, even with a large amount of data and full-field measurement. Among these methods, VFM particularly suffers from noisy displacement data and post-DIC processing. This is not ideal, especially since the DIC+VFM workflow is often the main choice of identification method for model parameter identification.

To summarize, the above two-step approach is a well-established pipeline to address the difficult problem of indirect inversion to obtain material models, and there exist various established methods and software options to address each step. Regardless, important to note that in this sequential approach, the first step (DIC) is performed independently of the governing physics. The displacement field is inferred by matching images, not by enforcing equilibrium, compatibility, and knowledge of the constitutive relation during the image inversion step. Any known physics and mechanical constraints enter only afterward, during the analysis step to obtain the parameter. This leads to an intermediate deformation field that may contain components that are optimal for image matching yet inconsistent with the physics of the deformation. In addition, physically meaningful information contained in the raw images may be lost when the images are compressed into an ansatz-dependent displacement field. After this DIC step, in the following step to infer parameters, most of the identification methods outlined above are highly sensitive to the measurement noise and errors in the displacement fields. To enable robust inversion, one needs reliable full-field displacement measurements, and thus one is often limited, again, to simple configurations, which leads to sparse data with significant uncertainties. Importantly, most of the identification methods above are only well-developed for a simple constitutive law, such as linear elastic materials. Extension to complex materials, materials with history, or materials with heterogeneity is not straightforward. These methods and thus the overall current workflow to learn material models suffer to learn the behavior of complex, multifunctional materials we are interested in.

Other variants of DIC and integrated approaches

Besides local and global DIC and other approaches described above, several variants have been developed to reduce the gap between image-based displacement measurement and model parameter identification. Here, we shall describe some of them.

Firstly, the global DIC method can be specialized to a finite-element-based decomposition, where the displacements are assumed to be described by bilinear quadrilateral shape functions (Q4) often used in finite element simulations (also called Q4-DIC) [31]. This identical description provides a natural interface with numerical modeling for the problem of the identification of material parameters. Following this idea, an integrated DIC approach (I-DIC) for material identification was proposed in [32], where the global DIC method is coupled with FEMU. The global displacement is represented by a set of global basis functions that match any arbitrary FE mesh and FE shape functions. Given that the measured displacement fields are FE-compatible, the FEMU step can be used to identify model parameters directly. The authors demonstrate this for the recovery of simple linear elastic materials.

Other approaches that integrate DIC and parameter identification were developed mainly for fracture problems to identify stress intensity factors. In [33], for example, an integrated approach where the images are directly used to infer stress intensity is proposed. The idea is to leverage *a priori* knowledge of the type of mechanical behavior in the solid. The displacement ansatz in (6.4) is enriched with singular fields in the local crack frame, which is directly related to the stress intensity factors. Thus, processing of the images immediately provides an estimate for all the stress intensity factors. It is important to note that this approach made use of the known closed-form ansatz of elastic crack fields. The performance of this integrated approach is comparable to that of an equivalent two-step approach [34]. Therefore, this integrated approach is accurate as long as one assumes the correct analytical form of the displacement solution for the particular problem.

In global DIC, the displacement field is continuous. An extended DIC (X-DIC) approach for analyzing discontinuous displacement has been proposed [35, 36]. To capture cracks or slip interfaces, the authors enrich the global displacement ansatz with additional functions. Now, the displacement ansatz in (6.4) is enriched with Heaviside-type functions to capture jumps across interfaces, as well as singular basis functions involving $\sqrt{r}$ -type behavior. However, here, the material parameters

are not directly recovered. This method is then extended in [37] to combine with finite element methods to extract the material parameters (stress intensity factors) from displacements that minimize both the image-matching functional and the equilibrium of the solid. The functional (6.2) can also be enriched with an additional term penalizing the equilibrium gap— each term has its own penalty weights to be tuned. This approach is called extended and integrated DIC (XI-DIC). It is important to note that here, the equilibrium is not imposed exactly; its satisfaction depends on the penalty weight assigned to the equilibrium term. Thus, XI-DIC lies between a purely kinematic DIC and a physics-constrained problem. The performance of these various approaches for the identification of material parameters (stress intensity factors) has been done in [38].

In summary, these methods aim to introduce existing mechanical and physics knowledge into the image-matching problem. One can choose to enrich the displacement basis functions depending on the available knowledge about the deformation in the solid. Another option is to enrich the image-matching functional to incorporate this knowledge. In all cases, the goal is to reduce the information loss in the image-based inversion to obtain a more robust parameter identification, but in most cases, this two-step workflow is still needed.

Other data-driven approaches

Various alternatives to the classical two-step approaches have recently been developed, which will be discussed briefly here. Note that all of the above methods generally assume that the material response can be described by a parametrized constitutive model, so that the model discovery reduces to a parameter identification problem. The work in Chapter 7 of this thesis bypasses the first step of the conventional pipeline by inferring model parameters directly from raw images.

One can also consider an alternative route that bypasses the second identification step. Instead of identifying an explicit constitutive law, one bypasses the constitutive model and uses material data directly to close the mechanics problem. Here, the forward problem is solved by finding the best match between a material data set in the phase space (e.g., obtained from full-field experimental measurements) and the constraint set of admissible stress-strain fields (including any known physics). This was introduced and demonstrated in [39, 40]. The corresponding variational analysis has been established in both linear elasticity [41] and finite elasticity [42] settings. Another related approach is the recently developed EUCLID framework

[43, 44]. This method involves constructing a library of candidate constitutive model forms to then perform model selection and parameter identification simultaneously. Unlike conventional parameter calibration, the model form is part of the inversion. However, we note that these methods often still rely on processed data (such as displacement fields first obtained from image-based methods) rather than raw data directly.

A different one-step variational approach was proposed in [45]. Here, one aims to recover the finite-elastic energy density form. The data considered is the direct experimental input of prescribed boundary displacements and measured total elastic energies computed via boundary work. The method then compares these measured energies with computed energies from the proposed material law, posed as an optimal-control problem. This approach is well-posed, though it is currently limited to elasticity in a quasi-static setting.

Overall, these approaches demonstrate the growing interest in moving beyond the classical two-step model discovery. In this thesis, we address current challenges from a different perspective by incorporating raw experimental images directly into the inverse problem while enforcing the governing physics exactly. This motivates the formulation of constitutive model discovery as a PDE-constrained optimization problem that considers raw images and forces directly.

6.3 Adjoint method for PDE-constraint optimization problems

To solve any optimization problem, a gradient-based approach is often used. Throughout the works in Chapter 7 for optimization of image-matching, and in Chapter 8 for topology optimization, we pose the problem of interest as a PDE-constrained optimization. The adjoint method is leveraged to obtain sensitivities of the optimization objective with respect to the unknown model parameters or the density field. These sensitivities are then used to update the estimates of those unknown fields. This method is used to circumvent the calculations of the sensitivity of the PDE solution with respect to the unknown parameters. Such calculations are difficult to perform, and results are difficult to store; hence, eliminating such terms from the overall sensitivity calculation is desired. We illustrate this below.

The physical phenomena (or PDE) governing our material can be written in a weak form. We concern ourselves with the equilibrium PDE throughout this work, which in weak form is

$$\int_{\Omega} \sigma(\nabla u; P) \nabla \lambda \, d\Omega = 0, \quad \forall \lambda \in \mathcal{U}, \quad (6.7)$$

where $\mathcal{U}$ is the space of admissible displacement variations,

$$\mathcal{U} := \{u \in W^{1,2}(\Omega), u = 0 \text{ on } \partial_u \Omega\}. \quad (6.8)$$

We write our objective in a general form $O(u, \sigma(\nabla u; P); P)$, and limit ourselves to displacement fields that already satisfy equilibrium (and any other known physics). Given that equilibrium is satisfied, we can augment our objective with the weak form of the equilibrium

$$O^* = O(u, \sigma(\nabla u; P); P) + \int_{\Omega} \sigma(\nabla u; P) \nabla \lambda \, d\Omega; \quad (6.9)$$

this augmented objective is equivalent to the objective $O(u, \sigma(\nabla u; P); P)$ for any $\lambda \in \mathcal{U}$. Now we want to obtain the sensitivity of the objective with respect to the unknown field. We perturb the unknown P in the direction δP , resulting in the corresponding displacement perturbation

$$u_{,P} \delta P. \quad (6.10)$$

Then, taking variations of the augmented objective with respect to P ,

$$O^*_{,P} \delta P = O_{,P} \delta P + O_{,u} [u_{,P} \delta P] + O_{,\sigma} [\sigma_{,P} \delta P + \sigma_{,\nabla u} \nabla u_{,P} \delta P] \quad (6.11)$$

$$+ \int_{\Omega} [\sigma_{,P} \delta P + \sigma_{,\nabla u} \nabla u_{,P} \delta P] \nabla \lambda \, d\Omega. \quad (6.12)$$

Note that $u_{,P} \delta P$ or $\nabla u_{,P} \delta P$, the sensitivity of the PDE solution with respect to the unknown parameters, is difficult to compute. So, we group those difficult terms and claim that the sensitivity of the objective with respect to the parameters can be written as

$$O^*_{,P} \delta P = O_{,P} \delta P + O_{,\sigma} \sigma_{,P} \delta P + \int_{\Omega} \sigma_{,P} \delta P \nabla \lambda \, d\Omega, \quad (6.13)$$

if the adjoint variable $\lambda \in \mathcal{U}$ satisfies the following adjoint equation

$$0 = O_{,u} \phi + O_{,\sigma} [\sigma_{,\nabla u} \nabla \phi] + \int_{\Omega} (\sigma_{,\nabla u} \nabla \phi) \nabla \lambda \, d\Omega \quad \forall \phi \in \mathcal{U}. \quad (6.14)$$

6.4 Adjoint method vs. VFM: Adjoint method picks the optimal virtual field

We now turn to emphasize the similarity and main differences between the adjoint method and VFM. The VFM infers unknown model parameters P given full-field displacement measurements u^{meas} (described in Section 6.2).

For the rest of this section, we assume, for simplicity's sake, no traction boundary and no body force. First, to see their similarity, we shall consider the optimization problem considered in VFM, i.e., displacement-matching to recover parameters, P :

$$O(u) = \frac{1}{2} \int_{\Omega} |u - u^{\text{meas}}|^2 \, d\Omega, \quad (6.15)$$

where u satisfies the weak equilibrium condition, i.e.,

$$0 = \int_{\Omega} \sigma(\nabla u; P) \nabla \lambda \, d\Omega. \quad (6.16)$$

We take variations of the augmented objective with respect to P ,

$$O^*_{,P} \delta P = \int_{\Omega} (u - u^{\text{meas}})_{u,P} \delta P \, d\Omega \quad (6.17)$$

$$+ \int_{\Omega} [\sigma(\nabla u; P)_{,P} \delta P + \sigma(\nabla u; P)_{,\nabla u} \nabla u_{,P} \delta P] \nabla \lambda \, d\Omega. \quad (6.18)$$

Following the approach above, we claim our sensitivity to be

$$\int_{\Omega} \frac{\partial \sigma(\nabla u; P)}{\partial P} \nabla \lambda \, d\Omega, \quad (6.19)$$

if and only if $\lambda \in \mathcal{U}$ satisfies the adjoint PDE

$$0 = \int_{\Omega} (u - u^{\text{meas}}) \phi + \frac{\partial \sigma(\nabla u; P)}{\partial \nabla u} \nabla \phi \, \nabla \lambda \, d\Omega \quad \forall \phi \in \mathcal{U}. \quad (6.20)$$

The sensitivity in (6.19) in the small strain setting is

$$\frac{\partial O}{\partial P} = \int_{\Omega} \frac{\partial \sigma(\varepsilon(u); P)}{\partial P} \varepsilon(\lambda) \, d\Omega, \quad (6.21)$$

note its similarity with the sensitivity $\partial r / \partial P$ in VFM methods:

$$\frac{\partial r}{\partial P} = \int_{\Omega} \frac{\partial \sigma(\varepsilon(u^{\text{meas}}); P)}{\partial P} \varepsilon(v_*) \, d\Omega, \quad (6.22)$$

It is straightforward to see that the adjoint method selects the most optimal virtual field (λ) based on the objective, while VFM relies on user-prescribed virtual fields (v_*). The choices of virtual fields in VFM may not be optimal, or a chosen virtual field may not be linearly independent of another choice, which affects the quality of the inferred parameters. The quality of the result with VFM depends on the choice of this virtual field by the user, which is arbitrary, problem-dependent, and often requires domain-specific knowledge.

6.5 Bayesian Inverse Problem

In most of the aforementioned approaches, the parameter identification problem is posed in a deterministic setting. A statistical formulation provides an alternative way to address this inverse problem. Namely, the Bayesian perspective to inverse problems provides a systematic framework to combine prior knowledge, experimental data, and noise [46, 47]. Given prior knowledge, experimental data, and

noise statistics, Bayes' rule results in a posterior probability density of the unknown model parameters, blending the experimental data and the model. Bayesian inverse methods have been used in various fields to use data to more accurately inform physics-based models [48–51] and the discovery of constitutive laws in solid mechanics [52, 53].

Here, we shall provide a quick review of the Bayesian formulation of the inverse problem that will be the foundation of the work in Chapter 8. Throughout the rest of this overview, we closely follow Stuart [46].

We want to find an unknown u , which can be full-field or discrete unknown parameters, given y , an observation of a forward model. We solve for u given y , $y \in Y, u \in X$, with X, Y Banach. For the rest of the thesis, we only consider finite-dimensional data, hence $Y = \mathbb{R}^q$. Note that the solution u to $y = G(u)$ is ill-posed due to noisy data; in general, $y \notin \text{Im}_G$, so simply inverting G on data y will not be possible. Before going into the Bayesian formulation of the inverse problem, we remind ourselves of some probability relations.

We consider a finite-dimensional setting, where $u \in \mathbb{R}^n$. We denote $P(\cdot)$ as the probability density function of a random variable, $P(\cdot, \cdot)$ as the joint probability density function of both arguments, and $P(\cdot|\cdot)$ as a probability density function conditional on the second argument. The standard laws of conditional probability are:

$$P(u, y) = P(u|y)P(y), \text{ if } P(y) > 0. \quad (6.23)$$

$$P(y, u) = P(y|u)P(u), \text{ if } P(u) > 0. \quad (6.24)$$

The marginal probability density function on y is then given by

$$P(y) = \int_{\mathbb{R}^n} P(u, y) du, \quad (6.25)$$

$$= \int_{\mathbb{R}^n} P(y|u)P(u) du = Z. \quad (6.26)$$

So, the assumption that $Z > 0$ ensures the data can be obtained from the model. Assuming $Z > 0$, by combining the two identities above, we have

$$P(u|y) = \frac{1}{P(y)} P(u, y), \quad (6.27)$$

$$P(u|y) = \frac{1}{Z(y)} P(y|u)P(u). \quad (6.28)$$

This (6.28) is Bayes' theorem. In *Bayesian inverse problems*, we aim to infer an unknown quantity $u \in X$, from data $y \in Y$, which is assumed to be generated through a noisy observation model

$$y = G(u) + \eta. \quad (6.29)$$

Here, X is a Banach space which can be finite- or infinite-dimensional, $G : X \rightarrow Y$ is a forward and observation operator, and η is observational noise, usually taken to be a Gaussian random variable.

Rather than solving this inverse problem deterministically in the classical sense, we adopt a Bayesian probabilistic framework: (i) place a prior distribution μ_0 on u and then (ii) use Bayes' theorem to compute the posterior distribution μ^y , representing the updated belief about u after observing the data y .

Thus, the goal is to find a probability measure μ_y in X . We consider a finite-dimensional setting, where $u \in X = \mathbb{R}^n$, $y \in \mathbb{R}^q$, $\eta \in \mathbb{R}^q$.

Bayes' Theorem: The random variable $u \in \mathbb{R}^n$ is distributed according to the prior probability density function (p.d.f) $u \sim \pi_0(u)$ and the observational noise, also unknown, $\eta \sim \rho(\eta)$. Then for a fixed u , the distribution of y given u defines the likelihood,

$$y|u \sim \rho(y - G(u)). \quad (6.30)$$

The posterior probability $u|y$ is the solution to the Bayesian formulation of the inverse problem. Assuming the normalization constant

$$Z(y) = \int_{\mathbb{R}^n} \rho(y - G(u))\pi_0(u)du > 0, \quad (6.31)$$

then the posterior p.d.f follows from (6.28)

$$\pi_y = \frac{1}{Z(y)}\rho(y - G(u))\pi_0(u). \quad (6.32)$$

In a generalized infinite-dimensional case, where there are no densities π_y, π_0 with respect to Lebesgue measure, we use Theorem 6.2 in [46] and write in terms of measures. The posterior measure is defined with respect to the reference measure in the Radon-Nikodym derivative sense,

$$\frac{d\mu_y(u)}{d\mu_0(u)} \propto \rho(y - G(u)), \quad (6.33)$$

$$\frac{d\mu_y(u)}{d\mu_0(u)} \propto \exp(-\Phi(u; y)). \quad (6.34)$$

Note that Φ here is a loss potential, usually taken as the negative log-likelihood $\Phi = -\log(\rho(y - G(u)))$.

For the purpose of this thesis, we consider only Gaussian prior and Gaussian noise. Furthermore, u and y are finite-dimensional, typical in the setting of parameter identification problems in mechanics. We specify Gaussian noise statistics $\eta \sim \mathcal{N}(0, B)$ and Gaussian prior $\mu_0 = \mathcal{N}(m_0, C_0)$ for the rest of this Chapter. Going back to (6.32), if the forward map $G(u) = Au$ is linear, then we can obtain the posterior to follow

$$\pi_y(u) \propto \exp\left(-\frac{1}{2} |y - Au|_B^2 - \frac{1}{2} |u - m_0|_{C_0}^2\right), \quad (6.35)$$

which is also Gaussian by definition (one can obtain its mean and covariance (6.36) by completing the square of the above expression). Note here that the posterior involves two squared norms; the first is the discrepancy between the data and the forward model $G(u) = Au$, and the second is the deviation from the prior mean m_0 , each scaled by its respective covariance. The closed-form mean and covariance of the Gaussian posterior above $\mu^y = \mathcal{N}(m, C)$ is given by

$$m = m_0 + C_0 A^* (B + A C_0 A^*)^{-1} (y - A m_0), \quad (6.36)$$

$$C = C_0 - C_0 A^* (B + A C_0 A^*)^{-1} A C_0. \quad (6.37)$$

Extracting information from posterior

Once we have obtained μ^y , the posterior measure for u given the data y , we want to obtain information about this posterior measure. Recall that under the special case of Gaussian assumption for both prior and noise distributions, as well as linear mapping G , the posterior is also Gaussian. In most cases, however, the mapping G is not linear (recall G is a combination of the forward problem and observation operator); hence the posterior is not Gaussian, and computational tools are needed to obtain information from the posterior. Classical approaches include Markov-Chain Monte-Carlo (MCMC) methods, variational methods, and filtering methods.

Markov-Chain Monte-Carlo, or MCMC, is a sampling technique to approximately generate sample functions according to the posterior μ^y . The idea is that we empirically design a Markov chain whose distribution converges weakly to the target posterior measure. Firstly, the state space is explored using a kernel that proposes a point, and secondly, the probability of acceptance of this point depends on the

distribution of the posterior [54, 55]. This is effective and accurate for learning the posterior, but costly since it requires hundreds to thousands of PDE solves of the forward problem.

Another approach is to assume that, instead of sampling the entire posterior distribution directly, the variability in the posterior measure is small and it is tightly concentrated around a single "best" solution. Namely, we look for the MAP (maximum a posteriori) estimate, which is a point that maximizes the posterior probability density, or equivalently minimizes a functional $I(u)$ in (6.39) (also known as the negative log posterior). This treatment allows the use of classical optimization techniques to solve the Bayesian inverse problem. This results in a fast estimation, but the true spread of the posterior is not captured by a single MAP value. The MAP point is a point $u \in E$ that maximizes the posterior measure

$$u_{MAP} = \arg \max_{u \in E} \pi_y(u), \quad (6.38)$$

or,

$$\arg \min_{u \in E} \left(\frac{1}{2} |y - G(u)|_B^2 + \frac{1}{2} |u - m_0|_{C_0}^2 \right). \quad (6.39)$$

This MAP estimator is the minimizer of a regularized data-misfit functional: (i) the first term is the data misfit weighted by the noise covariance that comes from the likelihood, while (ii) the second term is a regularization term derived from the prior. Note that E is an appropriate space of u . This is $\mathbb{R}^n$ for u finite-dimensional, and for u infinite-dimensional is the so-called Cameron-Martin space $E = \text{Range}(C_0^{1/2})$ with inner product $\langle \cdot, \cdot \rangle_{C_0}$ and norm $\|\cdot\|_{C_0}$ associated with the Gaussian prior.

Filtering methods are another class of techniques to obtain information about the posterior measure. It is mainly used for time-dependent problems where data arrive in a stream. The idea is to approximate the posterior in time-dependent data by a sequence of probability measures in time, and the posterior is updated sequentially as the dataset increases. This approach assumes and leverages the Markov structure of the underlying model to break the inference problem into a chain of smaller ones. The idea behind filtering methods is to break up the data sequentially, and the unknown sequentially. After decomposing, at time t_j , we seek to obtain π_{j+1} , the posterior for u_{j+1} given $\{y_j\}_j^{j+1}$. Filtering methods first seek to find an intermediate state $\hat{\pi}_{j+1}$ of the probability distribution of the next state (i.e., π_{j+1}) given observations up to the current time, $\{y_j\}_j^j$. Then, Bayes' update is used to establish the likelihood of the predicted state upon the arrival of new data y_{j+1} . Overall, this is a highly efficient technique, especially since the proposed decomposition reduces the dimensionality

of the inverse problem, but may need careful tuning. One particular method is the Kalman filter, which considers only linear models and Gaussian distributions [47]. Here, the Kalman gain is introduced to recover the posterior mean and covariance upon observation of new data. An interesting note is that the update on posterior covariance does not depend on the mean and is independent of the data, which is only possible due to the linearity of the model. We note that this structure can be leveraged even for nonlinear models by linearizing the model around a point (often the MAP point) and then via the Gaussian assumption, the posterior covariance/uncertainty can be approximated before seeing any data.

Overdetermined and underdetermined Bayesian inverse problem

We now draw attention to the role of prior knowledge as measurements become more precise and data noise becomes small. The role of the prior is especially important depending on whether an overdetermined or an underdetermined inverse problem is being considered. This distinction is relevant to the model discovery problems considered in this thesis. In this section, we illustrate this distinction using the simplest linear, finite-dimensional setting. The calculations below follow closely from Stuart [46], where further generalizations can also be found.

Overdetermined inverse problem. For an overdetermined case, in the limit of zero measurement noise, the solution of the inverse problem is the solution of the least squares problem. In particular, uncertainty disappears, and the prior plays no role. Considering $u \in \mathbb{R}^n$, $y, \eta \in \mathbb{R}^q$ with $q = 2, n = 1$ for the sake of simplicity. We let G linear, $\eta \sim \mathcal{N}(0, \gamma^2 I)$, and $u \sim \mathcal{N}(0, 1)$. Note that the number of measurements is more than the number of unknowns. The posterior measure is then Gaussian:

$$\pi_y(u) \propto \exp\left(-\frac{1}{2\gamma^2} |y - gu|^2 - \frac{1}{2}u^2\right). \quad (6.40)$$

The posterior mean and covariance can be written in closed form as

$$m = \frac{\langle G, y \rangle}{\gamma^2 + |G|^2}, \quad \sigma^2 = \frac{\gamma^2}{\gamma^2 + |G|^2}. \quad (6.41)$$

If noise gets smaller and smaller,

$$m^+ = \lim_{\gamma \rightarrow 0} m = \frac{\langle G, y \rangle}{|G|^2}, \quad (\sigma^+)^2 = \lim_{\gamma \rightarrow 0} \sigma^2 = 0. \quad (6.42)$$

Note that m^+ is a point from the least-squares solution of the overdetermined linear equation $y = Gu$ found from the minimization problem

$$\arg \min_{u \in \mathbb{R}} |y - Gu|^2. \quad (6.43)$$

Underdetermined inverse problem. For an underdetermined case, there exist multiple solutions that can explain all the data collected. Thus, the prior acts to fill in the missing information on parts that have no data measurements.

We have $u \in \mathbb{R}^n$, $y, \eta \in \mathbb{R}^q$ with $q = 1$ and G linear for simplicity. Note that there is one measurement data, less than the number of unknowns.

$$y = \langle G, u \rangle + \eta \quad (6.44)$$

for some $G \in \mathbb{R}^n$. Given $\eta \sim \mathcal{N}(0, \gamma^2)$ and prior $u = \mathcal{N}(0, \Sigma_0)$. Then

$$\pi_y(u) \propto \exp \left(-\frac{1}{2\gamma^2} |y - \langle G, u \rangle|^2 - \frac{1}{2} \langle u, \Sigma_0^{-1} u \rangle \right). \quad (6.45)$$

Since G is linear, the resulting Gaussian posterior mean and covariance can be written in closed form

$$m = \frac{\Sigma_0 G y}{\gamma^2 + \langle G, \Sigma_0 G \rangle}, \quad \Sigma = \Sigma_0 - \frac{(\Sigma_0 G)(\Sigma_0 G)^*}{\gamma^2 + \langle G, \Sigma_0 G \rangle}. \quad (6.46)$$

One can see that as noise gets smaller and smaller,

$$m^+ = \lim_{\gamma \rightarrow 0} m = \frac{\Sigma_0 G y}{\langle G, \Sigma_0 G \rangle}, \quad \Sigma^+ = \lim_{\gamma \rightarrow 0} \Sigma = \Sigma_0 - \frac{(\Sigma_0 G)(\Sigma_0 G)^*}{\langle G, \Sigma_0 G \rangle}. \quad (6.47)$$

Here, $\Sigma^+ G = 0$ and $\langle m^+, G \rangle = y$. The intuition here is that as noise gets smaller, knowledge of the unknown u in the direction of G becomes certain. But in directions not aligned with G , the uncertainty remains. The magnitude of uncertainty is given by both the prior and the likelihood. The prior is still present even with zero noise here.

We close this section by connecting the discussion above to the problem of model discovery in solid mechanics. In many mechanics problems, it is common to assume that the material response can be described by a parametrized constitutive model, so that model discovery reduces to a parameter identification problem [10, 25]. The data are often in the form of full-field experimental measurements (e.g., full-field images of the deformations). Thus, in this setting, we have an overdetermined case, where we seek to identify a relatively small number of parameters from high-dimensional data (data dimension is much larger than the unknowns', $q \gg n$). It follows from the above discussion that, in the small-noise limit and for a linear forward map G , the prior knowledge does not affect the posterior solution. This provides a useful intuition. However, this invariance is not guaranteed when the forward map is

nonlinear. This nonlinearity is always the case in model discovery of materials, since G includes the constitutive model, finite element solve, and observation or imaging operators. The data is also noisy in practice. As a result, the specification of the prior remains an important component of the inference procedure. We should therefore leverage all available prior knowledge about the problem. This includes any governing physics, mechanical balance laws, constitutive assumptions, and any additional information on the material behavior. We note that this is exactly the setting considered in Chapter 8.

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IMAGE TO CONSTITUTIVE MODEL (I2C) METHOD

A. Wihardja and K. Bhattacharya, “Constitutive relations from images”, *Journal of Applied Mechanics* **92**, 081009 (2025).

Contributions: A.W derived the main result of this paper.

Abstract

Constitutive relations close the balance laws of continuum mechanics and serve as a surrogate for a material in the design and engineering process. The problem of obtaining the constitutive relations is an indirect inverse problem where both the relation and the quantities that define the relation have to be inferred from experimental observations. The advent of full-field observation techniques promises a new ability to learn constitutive relations under realistic operational conditions. However, this is done in two steps, first obtaining deformations from the images, and then obtaining the constitutive relation from deformations and forces. This leads to a variety of difficulties. In this paper, we propose a novel approach that enables us to obtain constitutive relations directly from the raw data consisting of images and force measurements.

7.1 Introduction

The design of structural, aerospace, protection, and other thermomechanical engineering systems requires a constitutive relation that describes the material properties [1, 2], and is empirically determined. The essential challenge is that we cannot measure these constitutive relations directly; in fact we cannot even measure the quantities like stress, heat flux, energy density, and state variables that describe the constitutive relations.

The traditional approach relies on uniform states (e.g., uniaxial tension) or universal solutions (e.g., torsion) [2]. Each test provides limited information, and they require repetition. This has been addressed with automation and high-throughput approaches [3]. However, this is not always possible, since some experiments (e.g.,

plate impact) need sterile conditions with precise alignment to obtain the desired state [4]. In any case, all of these still sample idealized states of strain. These limit the fidelity of the resulting models when we use them to model complex situations. We propose to develop methods that can provide high fidelity constitutive relations by probing complex domains and complex states by using full field techniques like digital image correlation (DIC) [5, 6] and thermal imaging.

DIC takes a series of images of a decorated surface undergoing deformation and compares them to infer the deformation [5, 6]. It has emerged as a method of choice due to the ease of using it with the widespread availability of cameras and robust commercial (e.g., [7]) and open source (e.g., [8]) software. The full-field information it provides also gives useful insight. It has been extended beyond optical imaging (e.g., [9] using scanning electron microscopy images), to digital volume correlation [10, 11], and tomography [12]. The deformation is then used to obtain constitutive relations.

This current practice of splitting the inverse problem into two inverse problems, first image to deformation, and second deformation and force to constitutive relation, poses a number of challenges. First, these inverse problems may be ill-posed and have to be regularized. When they are done separately, the regularization of one problem can limit the information available to the other problem. Indeed, DIC as a purely kinematic problem of inferring deformation is ill-posed. In practice, it is either regularized using filters or constrained by finite dimensional (local or global) ansatz. However, finite dimensional approximations do not necessarily converge, and regularization limits the efficacy when there are cracks, shear bands, and shocks. Further, we image a fixed region of space that may correspond to different parts of the body in different snapshots. So, image registration limits us to small displacements. One can stitch together multiple snapshots, but errors may be compounded, and the method is inaccessible to dynamic events where one has very few snapshots.

Second, even when a reasonable deformation is available, the second inverse problem is still a difficult problem since we cannot directly measure stress, but have to infer it from the total force acting on a part of the boundary. So it is still common to use simple configurations, though the full-field inversion is a subject of active research, and we refer the reader to [13] for a recent literature survey.

Third, we may have a mismatch of sensitivities. Even if the overall sensitivity is reasonable, the splitting into two problems leading to situations where the first inversion has low sensitivity, but the second has high sensitivity. This leads to

significant errors.

Finally, there are phenomena where the instrument and the experiment are intimately coupled, rendering a sequential inversion impossible or dependent on unverifiable ansatz. For example, Lawlor *et al.* [14] recently introduced a speckle pattern in the interior of a transparent specimen and were able to observe the interaction between a shock wave and a pore. However, due to the photoelastic effect that changes the refractive index under stress, the image is heavily distorted in the vicinity of the shock as well in the wake of the shock.

In this paper, we propose an alternate approach where we study the integrated inverse problem of inferring constitutive relations directly from raw data in the form of images of speckle patterns and overall forces. We formulate this as an optimization problem and solve it using a gradient-based approach. We focus on finite elasticity and rubber, and demonstrate the method using both synthetic and experimental data.

The study of the mechanical properties of rubber goes back to seminal works of Treloar [15], Rivlin and Saunders [16], and Gent and Rivlin [17]. Assuming incompressibility, one can fully characterize the material by applying two principal stretches independently, and this motivated the biaxial tension test [16] that is widely used [18, 19]. These and other approaches [20–22] seek to create a uniform state. Others use universal solutions [23] like torsion [15] and inflation [17]. More recent efforts study complex deformations in two and three dimensions, exploiting advances in optical microscopy [24–26] and x-ray computed tomography [27]. This leads to the inverse problem that is the focus of this work.

We provide the formulation in Section 7.2. We demonstrate the approach using synthetic data in Section 7.3, and experimental observations in Section 7.4. We conclude in Section 7.5

7.2 Formulation

We assume that the material is hyperelastic, and is governed by a stored energy per unit reference volume W . We assume a parametrized form, so that $W = W(F; P)$, where F is the deformation gradient and $P \in \mathbb{R}^P$ is a set of parameters. Our goal is to find the parameters from an experiment.

Image to constitutive relation

We consider a body occupying the region $\Omega \subset \mathbb{R}^n$ in the natural reference configuration. We fix part of the boundary $\partial_0\Omega$, and apply a time dependent Dirichlet boundary condition $y(x, t) = \bar{y}(x, t)$ on another part $\partial_y\Omega$. The remainder of the boundary is traction-free. At each instant t , the body is in equilibrium, and the deformation $y : \Omega \rightarrow \mathbb{R}^n$ satisfies

$$\nabla \cdot W_F(\nabla y; P) = 0 \text{ on } \Omega \quad (7.1)$$

subject to the boundary conditions where $W_F = \partial W / \partial F$. Equivalently,

$$- \int_{\Omega} (W_F(\nabla y; P) \cdot \nabla \varphi) d\Omega = 0 \quad \forall \varphi \in \mathcal{U} = \{u = 0 \text{ on } \partial_0\Omega \cup \partial_y\Omega\}. \quad (7.2)$$

We decorate a part of the traction-free boundary of the body with a speckle (or other distinctive) pattern, and image a certain region of space $\mathcal{R}$ that contains a part of the traction-free part of the boundary to obtain images $g(y, t)$ (we typically obtain a series of snapshots). We also measure the total reaction force on the part $\partial_y\Omega$.

Our goal is to obtain P from the observations $\{g(y, t), f(t)\}$, by matching them with the corresponding quantities computed with a model with parameters P . We formulate this as an optimization problem

$$P = \arg \min O(P), \quad (7.3)$$

where the objective is

$$\begin{aligned} O = & \int_0^t w(t) \left(\int_{\mathcal{S}} |g_0(x) - g(y(x, t), t)|^2 dA \right. \\ & \left. + \alpha \left| \int_{\partial_y\Omega} W_F(\nabla y(x, t); P) \hat{n} dA - f_{\text{exp}}(t) \right|^2 \right) dt, \end{aligned} \quad (7.4)$$

and $y(x, t)$ satisfies the governing equation (7.2), $w(t), \alpha$ are weights, and $g_0(x) = g(x, 0)$.

We solve this by gradient descent, and therefore seek to calculate the sensitivity of the objective with respect to the parameters. Doing so directly requires us to calculate y_P , the sensitivity of the solution to the governing equation (7.2) to the parameters; this is difficult. Therefore, we use the adjoint equation [28, 29]. To derive this equation, add the left hand side of (7.2) to our objective, and then differentiate with

respect to P . We obtain,

$$\begin{aligned} \frac{dO}{dP} = & \int_0^t \left(-2 \int_S w(t)(g(x, 0) - g(y(x, t), t)) g_y y_P dA \right. \\ & + 2\alpha w(t) \left(\int_{\partial_y \Omega} W_F \hat{n} dA - f_{\text{exp}}(t) \right) \cdot \left(\int_{\partial_y \Omega} (W_{FF} \nabla y_P + W_{FP}) \hat{n} dA \right) \\ & \left. + \int_{\Omega} (W_{FF} \nabla y_P + W_{FP}) \cdot \nabla \varphi dx \right) dt \end{aligned} \quad (7.5)$$

where W_F, W_{FF}, W_{FP} are all evaluated at $(\nabla y(x, t); P)$. This still contains the problematic quantity y_P . However, this expression is true for all $\varphi \in \mathcal{U}$ (cf. (7.2)). We now make a special choice such that we eliminate y_P from above. Specifically, let φ satisfy the *adjoint equation*

$$\begin{aligned} & \int_{\Omega} \nabla \psi \cdot W_{FF} \nabla \varphi dx + \int_S \mu(x, t) \psi dA + \\ & \int_{\partial_y \Omega} \nabla \psi \cdot W_{FF} (\lambda(t) \otimes \hat{n}) = 0 dA \quad \forall \psi \in \mathcal{U} \end{aligned} \quad (7.6)$$

where we have used the fact that W_{FF} has major symmetry and

$$\begin{aligned} \mu(x, t) &= -2w(t)(g_0(x) - g(y(x, t), t))g_y, \\ \lambda(t) &= 2\alpha w(t) \left(\int_{\partial_y \Omega} W_F \hat{n} dA - f_{\text{exp}}(t) \right). \end{aligned} \quad (7.7)$$

Then, the sensitivity of the objective with respect to the parameter is

$$\frac{dO}{dP} = \int_0^t \left(\int_{\Omega} W_{FP}(\nabla y(x, t); P) \cdot \nabla \varphi dx + \lambda(t) \cdot \int_{\partial_y \Omega} W_{FP} \hat{n} dA \right) dt. \quad (7.8)$$

Numerical method

We start with an initial guess for the parameters, and then solve the forward and inverse problem using a finite element method, use the sensitivity to update the parameters, and iterate until a convergence criteria is met. Note that the forward problem is non-linear and we solve this iteratively using Newton-Raphson iteration. We discretize both the forward and the inverse problem with a 3D brick element and a standard piecewise polynomial Lagrange basis function of degree 1. The parameters are updated using method of moving asymptotes (MMA) [30] until either the norm of the sensitivities or the incremental objective reaches $\varepsilon = 10^{-5}$. Algorithm 1 summarizes our approach.

The calculation of μ requires some care. Since this term involves quantities from images that are typically pixelated, the integration is performed by locating pixels in

Algorithm 1 Constitutive relations from images

Input: Reference image g_0 , deformed images g , parametrized model

Output: Displacement y

Step 1: Initiate guess of parameters P ;
Step 2: Pick region of interest ($\mathcal{S}$) to perform correlation;
Step 3: Determine locations of each pixel in each finite element within $\mathcal{S}$;
Step 4: Precompute spatial gradients of the image ∇g ;
while $\|dO/dP\|$ **or** $\|O_k - O_{k-1}\| > \varepsilon$ **do**
 Step 5: Solve forward problem for displacement $y_k(x, t)$;
 for each *pixel in each finite element in $\mathcal{S}$* **do**
 Step 6: Compute $g(y(x, t), t)$, g_y from $y_k(x, t)$;
 end for
 Step 7: Solve adjoint problem for adjoint variable $\varphi_k(x, t)$;
 Step 8: Compute objective $O(y_k, P_k)$;
 Step 9: Compute sensitivity $dO(y_k, P_k)/dP$ for each P ;
 Step 10: Update parameters $P_{k+1} = P_k + \delta P$ with MMA;
end while

each element and summing over these pixels (instead of quadratures). Further, the imaging is performed in the current configuration, and the current image $g(y, t)$ is in pixelated form with uniform pixel spacing in y (current configuration). However, we integrate it over the reference domain. Therefore, we have to pull it back to the reference configuration by finding the values of $g(y(x, t), t)$ where x is sampled uniformly in the reference domain. We use bilinear interpolation to do so. We compute $\nabla_y g$ by pixel differences in the current configuration and then interpolate to pull it back to the reference configuration. While this term can be noisy, it is integrated against the shape function that is smooth on the scale of the pixels, and leading to a stable calculation of μ .

Contrast with the purely kinematic DIC

Before we demonstrate the proposed method in the next section, we comment on the mathematical issues associated with the current practice of treating digital image correlation as a purely kinematic problem of finding deformations. In this approach, we compare the image $g_0(x)$ of a decorated surface before deformation with $g(y)$ after the deformation occurs to obtain the deformation $y(x)$. Specifically, we maximize the correlation between the reference and convected images over all possible deformations, or equivalently, minimize the L^2 norm between the reference

and convected images over all possible deformations,

$$y = \arg \max \int_S g_0(x) g(y(x)) dA = \arg \min \int_S |g_0(x) - g(y(x))|^2 dA. \quad (7.9)$$

The two formulations are equivalent since expanding the second integral gives us the first up to a factor (-2) and the L^2 norm of the images (that are independent of the deformation).

Unfortunately, this problem is ill-posed. Note that the second problem in (7.9) is the classical optimal transport problem of Monge (transporting a mass with density g_0 to a mass with density g), and y is called the transport map. Unfortunately, this problem is not mathematically well-posed, and solutions may not exist and minimizing sequences may not converge. The relaxation, according to Kantorovich, is to look for minimizers over transport plans, $G(x, y)$, such that

$$\min_G \int_S G(x, y) |x - y|^2 dA \times dA \quad (7.10)$$

subject to

$$\int_S G(x, y) dA_y = g_0(x), \quad \int_S G(x, y) dA_x = g(y). \quad (7.11)$$

In essence, the image intensity at some reference point x can get spread over a region of the spatial domain, and the intensity at the spatial point y may come from a region of the reference domain. In short, the solution to the classical DIC problem (7.9) may not exist.

In practice, we make a finite dimensional ansatz on the deformation. Two are common. The first is local, where we select a grid of points $\{\bar{x}^i\}$ and assume that the deformation is piece-wise affine in a sub-domain around these points

$$y(x) = \sum_i (y^i + F^i(x - \bar{x}^i)) \chi_i(x), \quad (7.12)$$

where χ_i is the indicator function of the i^{th} sub-domain around $\bar{x}^i$. The second is non-local where the deformation is given in terms of a finite-dimensional (often finite-element based) basis set $\{\psi_i\}$,

$$y(x) = \sum_i y^i \psi_i(x) \quad (7.13)$$

These constraints yield a solution, but there is no notion of convergence as the number of grid-points or basis functions goes to infinity. Further, this constraint

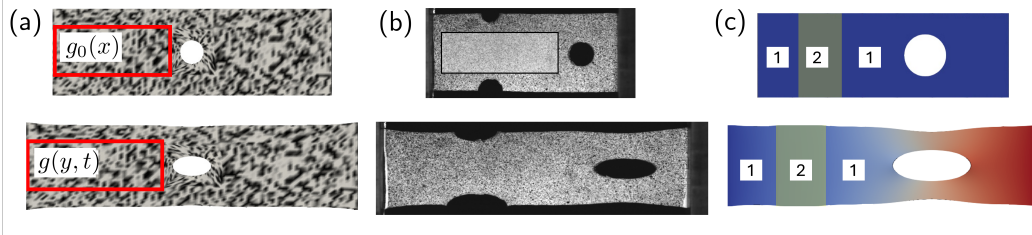


Figure 7.1: Images of reference and deformed configuration for (a) testing, (b) experimental validation, and (c) two-material example. The region of interest to perform correlation is highlighted in red in (a).

to finite dimensions makes the method fail or have poor accuracy when there are cracks, shear bands, and shocks.

There are other difficulties with a purely kinematic approach. We image a fixed region of space that may correspond to different parts of the body in different snapshots. So, image registration limits us to small displacements. One can stitch together multiple snapshots, but errors can be compounded, and the method is inaccessible to dynamic events where one has very few snapshots. Further, there are situations where the deformation and imaging interact as, for example, in the recent work of Lawlor *et al.* [14] mentioned in the introduction.

Note that we overcome these difficulties in our proposed approach, since the deformation is obtained from the equilibrium equation (7.2) and thus is well defined.

7.3 Testing the idea with synthetic data

We test the idea by using synthetic data: we use a known constitutive relation, solve the forward problem with that constitutive relation, and use it to generate synthetic data. We then use the procedure described in Section 7.2 on the synthetic data to obtain the constitutive relation. Comparing the inferred constitutive relation with the known constitutive relation provides us with a verification of the method.

Homogeneous material

We consider a compressible hyperelastic constitutive relation motivated by neo-Hookean:

$$W = \frac{\mu}{2} \left(\text{tr}(F^T F) - 3 - 2 \log(\det F) \right) + \frac{\kappa}{2} (\det F - 1)^2 \quad (7.14)$$

where the parameters are the shear and bulk moduli $P = \{\mu, \kappa\}$. We consider a specimen $(0, \ell) \times (0, w) \times (0, t)$ in the form of a rectangular strip with a central circular hole as shown in Figure 7.1(a). We create a speckle pattern $(g(x, 0))$ from

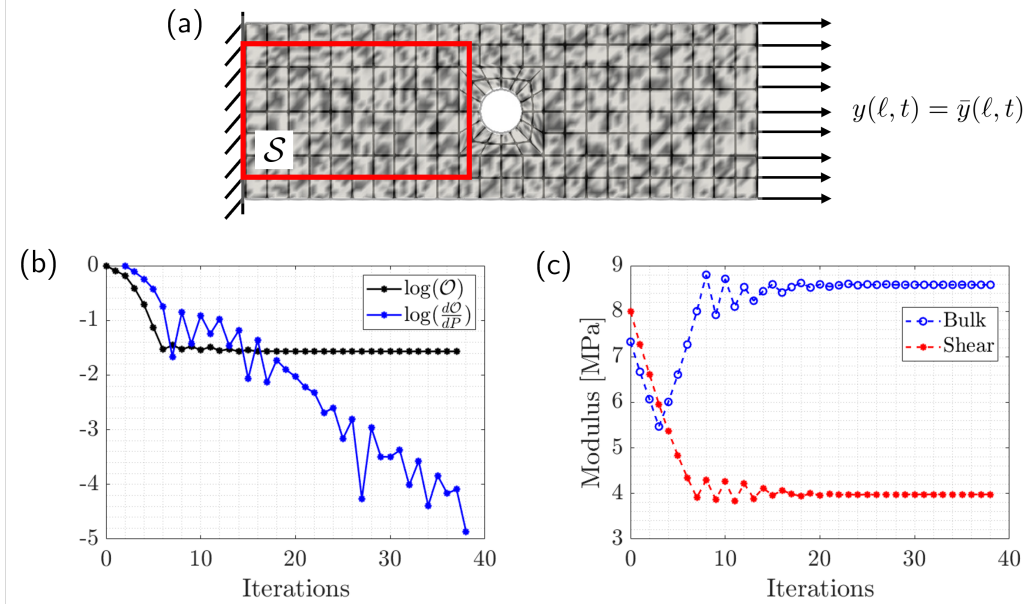


Figure 7.2: Results for quasistatic uniaxial tensile test of specimen shown in (a). The red box is the region to perform image correlation, $\mathcal{S}$. (b) The normalized objective in (7.4) and the combined sensitivity of material parameters with iterations. (c) The evolution of shear and bulk modulus. The synthetic data is generated with $\mu = 4$ MPa, $\kappa = 8.6$ MPa and the optimization is run with initial guess of $\mu = 8$ MPa, $\kappa = 7.3$ MPa.

correlated solutions [7] also as shown. We clamp this specimen with the constitutive relation (7.14) on the left ($y(\{0, x_2\}) = \{0, x_2\}$), and apply a horizontal displacement on the right $y(\{\ell, x_2\}) = \{(1 + \dot{\epsilon}t)\ell, x_2\}$ where the nominal strain rate $\dot{\epsilon}$ is taken to be extremely small. The lateral surfaces, $x_2 = 0, w$, are traction-free. We solve the equilibrium equation (7.2) at each time t to obtain the synthetic deformation $y(x, t)$. We obtain the speckle pattern in the current configuration by convecting the reference image by the deformation:

$$g(y, t) = g_0(x(y, t)) \quad (7.15)$$

where $x(y, t)$ is the inverse deformation. A snapshot is shown in Figure 7.1(a). We also compute the total reaction force $f(t)$ at $x_1 = \ell$ at each instance. We use $\{g_0, \dot{\epsilon}\}$ and a number of snapshots of $\{g, f\}$ as our data.

Known constitutive relation. We start with a situation where the constitutive relation used to generate the data is the same as assumed in the inference, and we seek to find the parameters. A typical result is shown in Figure 7.2. The data is generated with a initial parameters $\mu = 4$ MPa, $\kappa = 8.67$ MPa, and the calculation

Case	Initial guess		Recovered value	
	Shear modulus, μ_i	Bulk modulus, κ_i	Final shear, μ_f	Final bulk, κ_f
Set 1: Synthetic data generated with $\mu = 4$ MPa, $\kappa = 8.67$ MPa				
1a	4 MPa	38 MPa	3.95 MPa	8.63 MPa
1b	1 MPa	9.67 MPa	4.03 MPa	8.35 MPa
1c	0.4 MPa	3.8 MPa	3.95 MPa	8.61 MPa
1d	40 MPa	387 MPa	3.95 MPa	8.50 MPa
1e	0.4 MPa	0.36 MPa	3.98 MPa	8.52 MPa
1f	8 MPa	7.34 MPa	3.98 MPa	8.58 MPa
1g	5 MPa	6.67 MPa	3.99 MPa	8.61 MPa
Set 2: Synthetic data generated with $\mu = 4$ MPa, $\kappa = 38.6$ MPa				
2a	1 MPa	3 MPa	3.99 MPa	33.3 MPa
2b	8 MPa	10 MPa	3.99 MPa	33.3 MPa
2c	2.2 MPa	7 MPa	4.38 MPa	36.0 MPa
Set 3: Synthetic data generated with $\mu = 0.5$ MPa, $\kappa = 4.83$ MPa				
3a	1 MPa	2 MPa	0.55 MPa	4.48 MPa
3b	0.1 MPa	0.8 MPa	0.49 MPa	4.19 MPa
3c	0.3 MPa	1.0 MPa	0.50 MPa	4.16 MPa

Table 7.1: Performance for different initial guesses and different sets of synthetic data.

is initialized with $\mu = 8$ MPa, $\kappa = 7.3$ MPa. Figure 7.2(a) shows the reference image of the specimen with the finite element grid used for the simulations, the speckle pattern and the region of correlation $\mathcal{S}$ marked. The overall dimensions are $0.036 \times 0.012 \times 0.005$ m. Figure 7.2(b) shows the objective and the sensitivity, while Figure 7.2(c) shows the material parameter as the iteration proceeds. We observe a quick drop in the objective, a steady drop in the sensitivity, and a stable value for the parameters. The recovered values are $\mu = 3.98$ MPa, $\kappa = 8.58$ MPa, in close agreement with the values that generated the data.

We repeat the simulation with several values of the initial guess, and data generated with three sets of parameters. The results are shown in Table 7.1. We see that we obtain very good recovery in each case.

We now turn to noisy data. Images obtained from an experiment have noise depending on the experimental setup and pixel accuracy, and so we seek to test our approach against noisy data. We generate images with 720×240 pixels and 360×120 pixels within the domain and add 10% uniformly distributed random noise to the

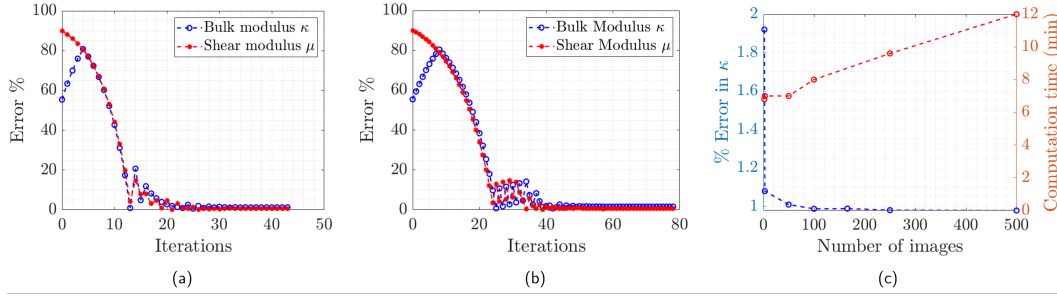


Figure 7.3: Performance with (a) 10% random noise in synthetic images with 720×240 pixels and (b) 360×120 pixels. (c) Various number of images for inversion.

% Error	Total pixels			Mesh refinement	
	180 × 60	360 × 120	720 × 240	1 × mesh	2 × mesh
Shear modulus, μ	0.5%	0.4%	0.4%	0.8%	0.4%
Bulk modulus, κ	2.4%	1.3%	0.6%	1.5%	1.3%

Table 7.2: Performance tests: Errors for various image and mesh resolutions.

speckle values for all pixels. The range of speckle values is between 0 and 1, hence, the maximum absolute noise value is 0.1. As before, the synthetic data is generated with $\mu = 4$ MPa and $\kappa = 8.67$ MPa. The initial guess of parameters is $\mu = 0.4$ MPa and $\kappa = 0.44$ MPa. The evolution of material parameters can be seen in Figure 7.3 (a,b). We note that the method is able to recover both parameters with just 4% error.

We study the performance of our method with 2, 3, 50, 100, 250, and 500 snapshots of $g(y, t)$, resulting in errors of 1.9%, 1.1%, 1.0%, 0.99%, 0.98%, 0.97% in the bulk modulus, respectively. The error in bulk modulus and computational time per iteration can be seen in Figure 7.3(c). In all cases, the number of optimization iterations using MMA method is 35, 35, 37, 39, 39, and 34, respectively. Thus, we get reasonable recovery with two snapshots and this improves mildly with more snapshots.

We conclude this study by looking at a few computational issues. We study the performance of the method under refinement for both the image and the finite element discretization. In each case, the synthetic data is developed using parameters $\{\mu, \kappa\} = 4$ MPa, 8.67 MPa, and the simulations are initialized with parameters $\{\mu, \kappa\} = 8$ MPa, 7.34 MPa. Table 7.2 summarizes the effect of refining the image (pixel resolution) while keeping the finite element discretization fixed. We see that the errors are still small, though it degrades a little with decreasing image resolution. The effect of fixing the image resolution (at 360×120 pixels) and refining the mesh

can also be seen in Table 7.2. We find a slight improvement in the recovery with a refined mesh.

Putting all these together, we conclude that the proposed method successfully recovers the parameters of a known constitutive relation.

Unknown constitutive relation. We apply our approach to synthetic data generated as described above with (7.14). However, in contrast to the previous example, we do not assume a knowledge of the form of the constitutive relation. Instead, we assume a different parametrized form, one motivated by a compressible Mooney-Rivlin relation

$$W = C_1(\bar{I}_1 - 3) + C_2(\bar{I}_2 - 3) + C_3(\bar{I}_1^4 - 3^4) + \kappa/2(\det F - 1)^2, \quad (7.16)$$

with parameters $P = \{C_1, C_2, C_3, \kappa\}$ where $\bar{I}_1, \bar{I}_2$ are the principal invariants of the left Cauchy-Green tensor, normalized by $(\det F)^{-2/3}$. We proceed as before and obtain the parameters $C_1 = 1.6$ MPa, $C_2 = 0.017$ MPa, $C_3 = 0.004$ MPa, and $\kappa = 12.4$ MPa.

To verify that the recovered constitutive relation still captures the actual constitutive behavior, we perform an independent simulation on a validation specimen with a different geometry as shown in Figure 7.1(b), with overall dimensions $0.05 \times 0.02 \times 0.005$ m. Figure 7.4(a) compares the measured and computed force between synthetic and recovered constitutive relations. Figure 7.4(b-d) shows the comparison of σ_{11} , σ_{22} , and σ_{33} at a nominal strain of 25%. We define the overall total error in the stress and strain fields to be

$$\begin{aligned} e_\sigma &= \frac{\int_0^t \int_\Omega (\sigma_{\text{synthetic}} - \sigma_R(P))^2 d\Omega dt}{\int_0^t \int_\Omega (\sigma_{\text{synthetic}})^2 d\Omega dt}, \\ e_\epsilon &= \frac{\int_0^t \int_\Omega (\epsilon_{\text{synthetic}} - \epsilon_R(P))^2 d\Omega dt}{\int_0^t \int_\Omega (\epsilon_{\text{synthetic}})^2 d\Omega dt}, \end{aligned} \quad (7.17)$$

and find 0.015% error in stress and 0.014% error in strain.

Therefore, we conclude that the proposed method successfully recovers the constitutive relation even without *a priori* knowledge of the form of the constitutive relation.

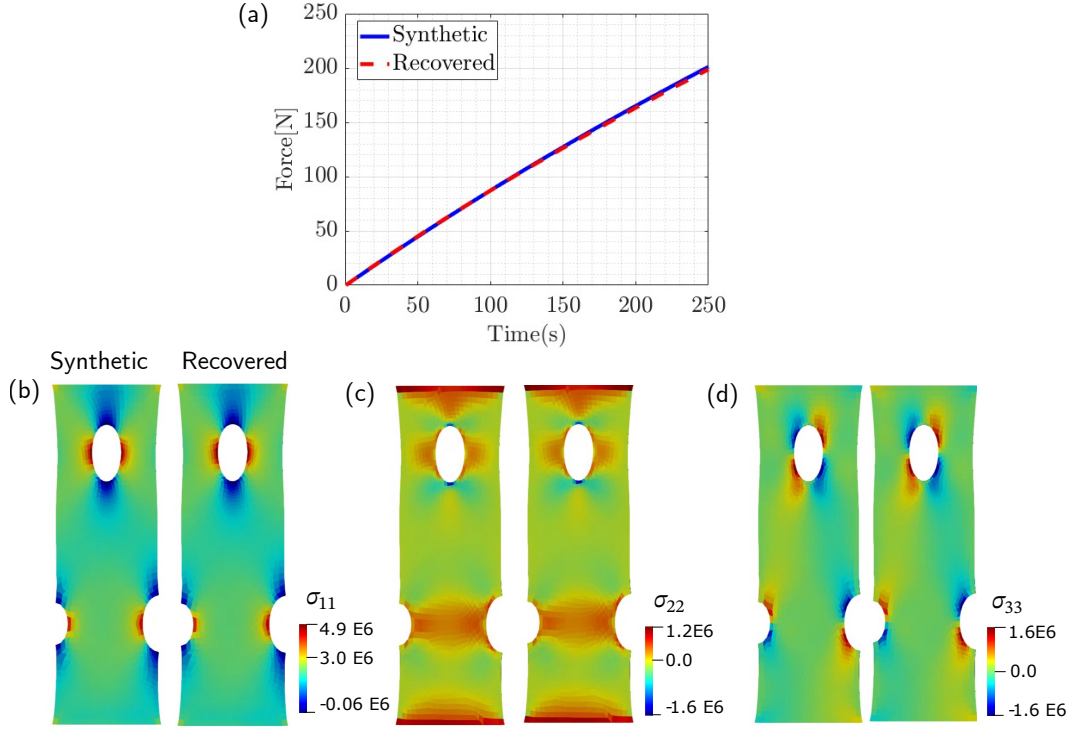


Figure 7.4: Comparison of the synthetic data and recovered constitutive relation using an independent test. (a) Force and (b-d) stress for the verification specimen at a nominal strain of 0.25.

Heterogeneous material

The method above can also be used when the material is piecewise uniform, as long as the domains of uniformity are identified *a priori*. The derivation of the sensitivity using the adjoint equation described earlier can easily be generalized to this case. The adjoint equation and the sensitivity are exactly as in (7.6) and (7.8) respectively, except that W depends on x . Importantly, the adjoint problem is solved only once per iteration, and the parameters for all the materials are updated simultaneously from the resulting sensitivity.

We now demonstrate the performance of our method when we have two materials as shown in Figure 7.1(c). We assume the constitutive law (7.14) for each material, but with different parameters. We generate synthetic data with $(\mu_1, \kappa_1) = (4 \text{ MPa}, 8.67 \text{ MPa})$ and $(\mu_2, \kappa_2) = (20 \text{ MPa}, 136 \text{ MPa})$ as described above. The results of our algorithm for various initial guesses are shown in Table 7.3. We observe excellent recovery as before.

Case	Initial guess		Recovered value	
	Shear modulus, μ_i	Bulk modulus, κ_i	Final shear, μ_f	Final bulk, κ_f
Material 1: Synthetic data generated with $\mu = 4$ MPa, $\kappa = 8.67$ MPa				
1	1 MPa	2.17 MPa	4.03 MPa	8.42 MPa
2	20 MPa	9.34 MPa	4.01 MPa	8.28 MPa
3	8 MPa	7.34 MPa	4.16 MPa	8.44 MPa
4	1 MPa	1.67 MPa	3.95 MPa	8.59 MPa
5	0.8 MPa	7.73 MPa	3.96 MPa	8.09 MPa
6	8 MPa	3.73 MPa	4.28 MPa	8.06 MPa
Material 2: Synthetic data generated with $\mu = 20$ MPa, $\kappa = 136$ MPa				
1	15 MPa	45 MPa	19.8 MPa	127 MPa
2	40 MPa	36.7 MPa	20.1 MPa	119 MPa
3	15 MPa	145 MPa	20.1 MPa	135 MPa
4	40 MPa	187 MPa	20 MPa	130 MPa
5	10 MPa	96.7 MPa	19.7 MPa	145 MPa
6	25.0 MPa	242 MPa	22.4 MPa	201 MPa

Table 7.3: Performance in the two-material case for different initial guesses.

7.4 Experimental demonstration

We now apply the proposed method on experimental data obtained using a natural rubber specimen (McMaster-Carr, 87145k411). We consider a specimen of size $0.036 \times 0.012 \times 0.0015$ m with a hole in the center as shown in Figure 7.1(a) and apply a speckle pattern of similar size to that in the figure. We clamp the left end and apply a uniaxial displacement to the right at a nominal strain rate of $\dot{\epsilon} = 0.001$ /s up to a nominal strain of 45% on the Instron E3000. As the sample deforms, we record the boundary force and image the sample at 2 frames per second, with a resolution of 1024×1024 pixels of the field of view (Photron Fastcam NOVA S12 camera and Tokina AT-X PRO lens, 100F 2.8D). We use bilinear interpolation of the image to downsample the images to $720\text{px} \times 240\text{px}$.

We then perform our method on the series of images collected. We assume that our material is described by a compressible constitutive relation motivated by the Mooney-Rivlin constitutive law,

$$W = C_1(\bar{I}_1 - 3) + C_2(\bar{I}_2 - 3) + \kappa/2(\det F - 1)^2, \quad (7.18)$$

with parameters $P = \{C_1, C_2, \kappa\}$ where $\bar{I}_1, \bar{I}_2$ are the principal invariants of the left Cauchy-Green tensor, normalized by $(\det F)^{-2/3}$. We obtain $C_1 = 0.33$ MPa, $C_2 = 0.21$ MPa, and $\kappa = 7.53$ MPa as the parameters from the experiment.

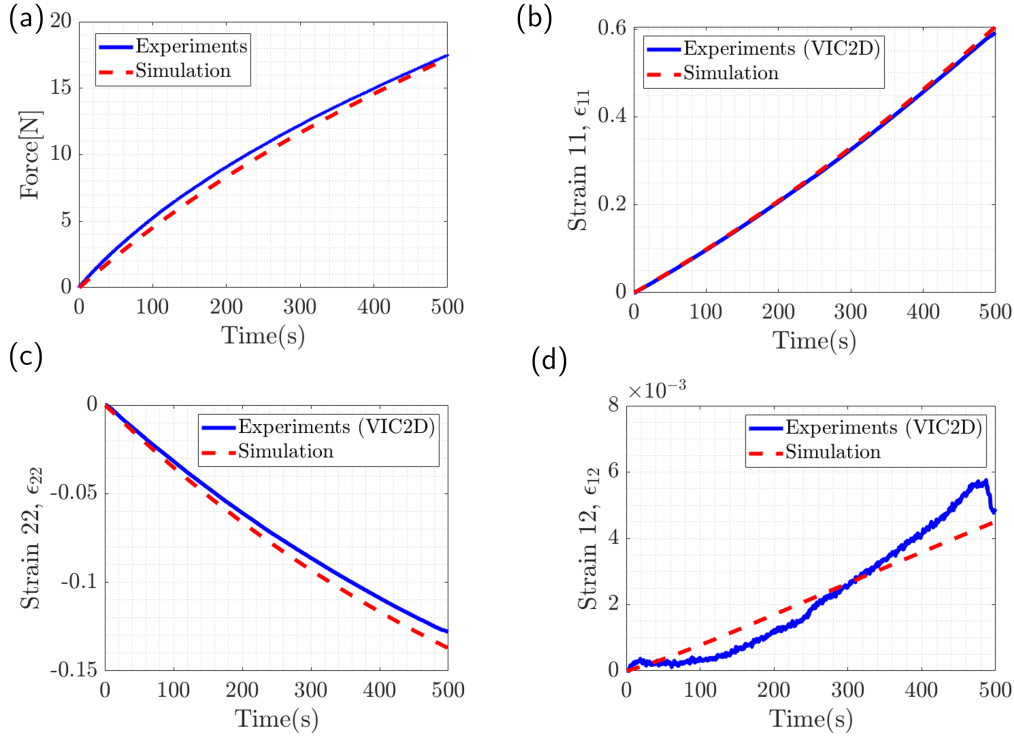


Figure 7.5: Comparison of the measured and computed (a) force and (b-d) average strain response for the verification specimen. The computed response uses the recovered parameters, and the strains are averaged over a region marked in Figure 7.1 using the recovered parameters for the verification specimen.

To verify that these recovered constitutive behavior accurately describes the material, we conduct an independent experimental test on a specimen with a different geometry shown in Figure 7.1(b), with overall dimensions $0.05 \times 0.02 \times 0.0015$ m. As before, the sample is deformed at $\dot{\epsilon} = 0.001/\text{s}$ up to 50% strain, and we record the force and image the specimen every 2 frames per second. Figure 7.5(a) compares the measured and computed force response using a finite element calculation and the recovered parameters. We find an excellent match.

We then use the recorded images and the commercial VIC2D software (Correlated Solutions, Columbia, SC) to obtain the (Lagrangian) strain field. Figure 7.6 compares the strain field obtained from VIC2D with that computed using finite element analysis and the recorded parameters at three loading stages. Figure 7.5(b-d) compares the strains averaged over a region marked in Figure 7.1(b). Again, we find an excellent match. We note that the discrepancy in ϵ_{12} is comparable to the strain uncertainty of VIC-2D. The uncertainty is determined by measuring the noise floor of strain values in VIC2D by taking a series of five still images of a control sample.

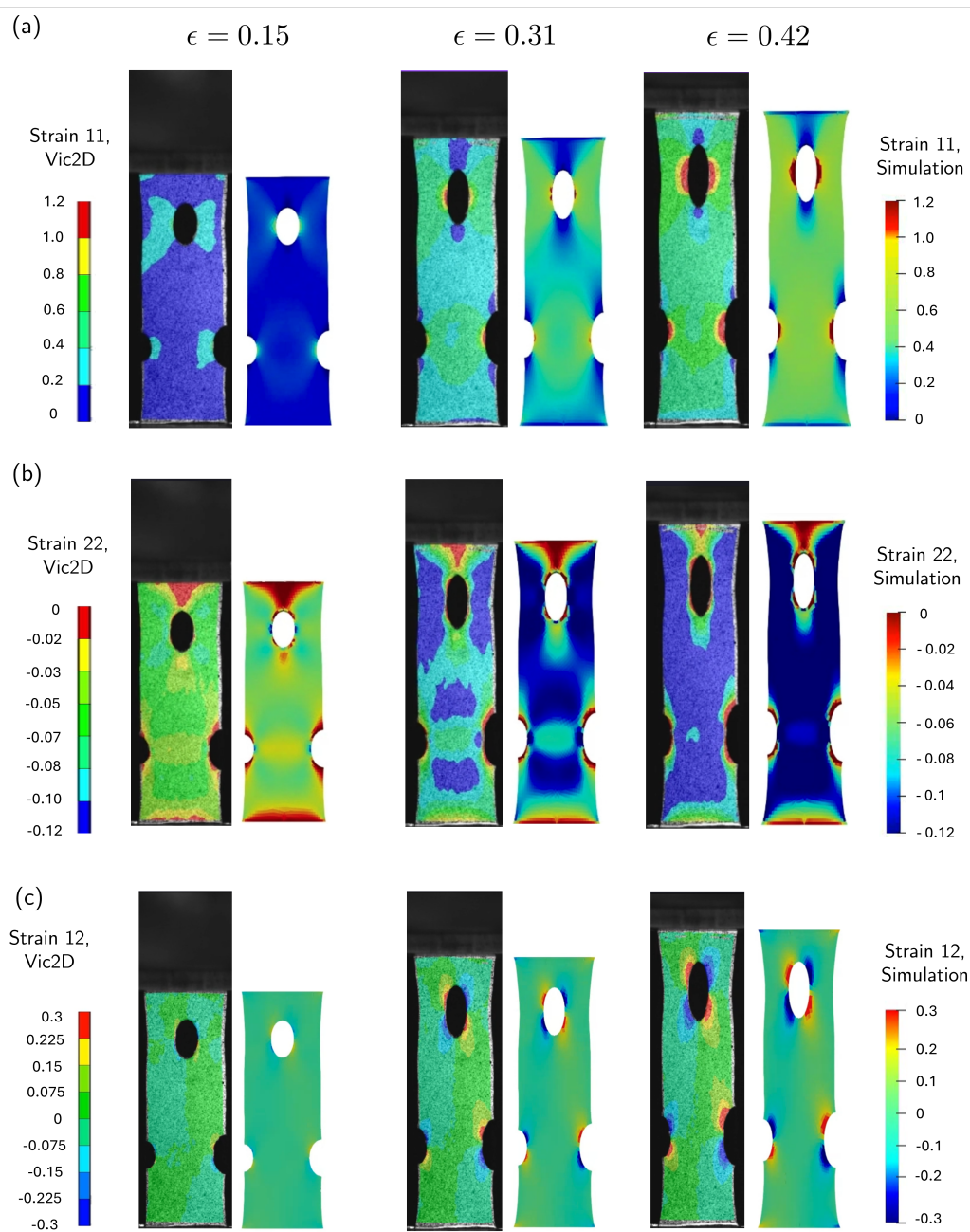


Figure 7.6: Experimental images used for verification of material parameters of a natural rubber specimen.

Under no deformation, the standard deviation of ϵ_{11} , ϵ_{22} , ϵ_{12} are 0.0006, 0.0005, and 0.0004 respectively.

We conclude that the proposed method successfully recovers the constitutive relation from an experimental test.

7.5 Conclusion

We address the problem of learning constitutive relations of materials from experiments. We seek to use the power of full-field observation techniques like digital image correlation. In a departure from previous work that breaks down the inverse problem into two nested optimization problems, first obtaining deformation from images and second obtaining constitutive relations from deformation and force, we propose an integrated approach. Specifically, we formulate the problem of obtaining constitutive relations from raw data (images and force) as a partial differential equation constrained optimization problem, and solve it using the adjoint method. The integrated approach overcomes many of the problems associated with treating digital image correlation as a purely kinematic problem of obtaining deformation from images, as well as the problems associated with a multi-level optimization. We demonstrate this approach on finite elasticity using both synthetic and experimental data. We view this as a first step in a larger program, and plan to address history-dependent behavior, shocks, fracture, and other phenomena in future work.

Acknowledgement

We are grateful for the financial support of the National Science Foundation (2009289) and the Army Research Office (W911NF-22-1-0269).

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Chapter 8

SPECIMEN DESIGN FOR MATERIAL PARAMETER IDENTIFICATION USING TOPOLOGY OPTIMIZATION

A. Wihardja and K. Bhattacharya, “Specimen design for material parameter identification using topology optimization.” arXiv:2607.16865, 2026.

Contributions: A.W. derived the main results of this paper.

Abstract

Constitutive relations close the equations of continuum mechanics, and serve as a surrogate for a material in the design and engineering process. They are often specified in a parameterized form with parameters identified by experiment. In this paper, we propose a framework for identifying experimental configurations that are maximally informative for constitutive model discovery. The framework strongly couples modeling and experimentation: the model leverages high-dimensional data from full-field measurements, while the current uncertainty in the model guides the design of future experiments. We formulate this goal by integrating Bayesian optimal experimental design with topology optimization. The Bayesian design criterion quantifies expected information gain, which drives the topology optimization of the specimen geometry.

8.1 Introduction

Constitutive relations, a map of kinematic quantities (e.g., strain, deformation history) to dynamic quantities (e.g., stress), are essential to close the equations of continuum mechanics [1, 2]. These relations therefore serve as a descriptor of the underlying, complex physics of a material. High-fidelity constitutive relations are essential for predictions in many engineering fields. In practice, identifying constitutive relations of a material involves selecting an appropriate model form, often parameterized with a set of parameters. This particular choice of model form is usually informed by the knowledge of the basic underlying physics of the material, while the parameters associated with the model form are then estimated, usually

from experiments. Once a complete model is established and calibrated, it is then deployed to predict material response in more complex real-world scenarios. The quality of these predictions thus depends on the fidelity of the chosen constitutive model, which in turn depends on the quality of its parameters identified.

Unfortunately, identifying constitutive laws is challenging, particularly because the quantities being mapped by these relations (stresses, strains, internal states) are not directly measurable in an experiment [3]. In material testing experiments, the directly observable and measurable quantities are boundary loads and boundary displacements, and one has to infer stress and strains from the measured data to then determine model parameters. Thus, in general, reconstructing stress and strain fields (or other internal physical quantities in the model) becomes part of the inversion. Therefore, traditional mechanical testing addresses this difficulty by relying on simple configurations and specimen geometries that furnish a state of homogeneous deformation in the sample, which corresponds to one strain state. The measured boundary force then provides the stress state corresponding to that strain point. This procedure identifies a single stress and strain data point, and when repeated over various loading conditions, allows the determination and calibration of a constitutive model. Rigorous experimental studies following this approach have been used to model isotropic rubber-like materials [4–6].

However, materials are often anisotropic and may require a significantly large number of experiments. To overcome these limitations, recent approaches leverage advances in optics, imaging, and electronics to obtain full-field data, often in the form of images, from which the material deformation state can be inferred. With full field images comes also the potential to sample nonuniform, heterogeneous, complex deformation states within a single experiment to provide richer information for model identification—this leads to a more accurate and cost-efficient modeling approach. However, many established image-based inversion methods still have limitations that limit the fidelity of the inferred constitutive models when they are used to predict complex responses in engineering applications [7, 8].

The first limitation arises due to limitations in the inversion analysis. Images provide rich full-field data, but using them to identify a constitutive law means one has to solve a more complex inverse problem. This inverse problem is inherently ill-posed: the material parameters and mechanical fields must be inferred from noisy, indirect measurements. As a result, regularization is often required, and the inferred parameters and model fidelity will depend sensitively on both the

data noise and the chosen regularization. Current methods divide this into two smaller inverse problems and follow a sequential procedure. First, images are converted to displacement fields using digital image correlation (DIC) or related techniques [9–12]. Second, the inferred displacement fields are then used to identify model parameters, commonly done with methods such as the virtual fields method (VFM) [13] or finite element model updating (FEMU) [14]. More recently, single-step approaches that directly infer constitutive models from images have also been developed [15].

In most of these approaches, however, the parameter identification problem is posed in a deterministic setting. These may be ill-posed and a regularization is introduced to stabilize the inversion. However, the choice of regulation can be somewhat arbitrary without making further modeling assumptions. Consequently, the fidelity of the inferred model depends on the regularization strategy, and selecting an appropriate regularization can require extensive domain knowledge that is often not available for new materials or complex problem settings.

A statistical formulation provides an alternative way to address the ill-posedness of the inverse problem in a more explicit fashion. In particular, the Bayesian approach to inverse problems provides a systematic framework to combine prior knowledge, experimental data, and noise; we refer readers to [16, 17] for a detailed discussion. An advantage of this Bayesian perspective over classical approaches is that it makes the modeling assumptions about prior knowledge and experimental noise statistics both clear and explicit. A second important advantage is that it serves as a natural way to quantify uncertainties in the inferred parameters. In short, the idea behind this Bayesian framework is to assimilate experimental data into models while explicitly quantifying the uncertainties in both model and data. The result of this data assimilation is the posterior probability density of the unknown parameters, blending the experimental data and the model. Bayesian inverse methods have been used in various fields to use data to more accurately inform physics-based models. Examples include fluid mechanics [18], groundwater flow [19], seismic [20], and thermoacoustic [21] problems, and the discovery of constitutive laws in solid mechanics [22, 23]. Despite those advantages, one downside to Bayesian approach is the high numerical cost in evaluating the resulting posterior probability density. This evaluation requires repeated solutions of the forward model, and sampling methods such as Markov Chain Monte Carlo (MCMC) [24, 25] though accurate, require hundreds to thousands of PDE solves. Consequently, many recent work on Bayesian

inference has also focused on developing fast and scalable numerical approaches to reduce the number of model evaluations for expensive physics models [20, 23, 26].

A second limitation arises from the design of the experiment itself [27]. Even when high-dimensional image data are available, often current experimental procedures still rely on specimen geometries chosen using intuition, standard testing protocols, or simple modifications of conventional geometries (e.g., dogbones and notched specimens). Such standard and conventional topologies were originally developed for reliable data and straightforward interpretation, but do not necessarily maximize the information content of the experiment. So, we are still accessing a limited number of deformation states, leading to sparse data. Access to high-dimensional full-field measurements create an opportunity to utilize rich information in these measurements by deliberately designing complex geometry specimens or loadings to probe multiple deformation states within a single test. This allows for a more accurate model inference while reducing experimental cost, giving rise to a natural design optimization problem: selection of specimen geometries or loadings that maximize information gain. This optimal design problem is nontrivial, since the relationship between design variables (for example, sample geometry) and the measured full-field response is not direct: it is governed by balance laws and is dependent on the constitutive relationship itself, which we seek to infer. Bayesian framework can provide a quantitative path to optimal design of experiments by accounting explicitly for knowledge of model, parameter uncertainty, and noisy data. This is another advantage of using Bayesian perspective to inverse problems, as it not only quantifies uncertainty in the inferred material parameters, but also provides a principled foundation for optimal experimental design.

Optimal experimental design (OED) for model inference seeks to determine experimental configurations that are maximally informative for the inference problem. To do optimal design, one typically defines the overarching goal through a utility function that is often related to a Fisher information matrix or a covariance. Descriptions of utility functions and their relation to various experimental goals can be found in [28–30]. Different design criteria result in various optimal design strategies, for example, D-, A-, E-, or c- optimality, among others.

In the Bayesian perspective, we focus on works by Lindley [31]. According to Lindley, the expected information content of the next experiment can be used as a measure of the utility of experimental design for model inference. This expected information gain (EIG) is defined as the expected Kullback-Leibler divergence from

the prior distribution to the posterior distribution. This approach has been the foundation of many experimental design strategies in material model identification [32], subsurface flow [33], thermoacoustic instability [34], shock tube experiments [35], and general advection–diffusion problems [36], for example. This approach, while meaningful, often struggles with costly evaluation of the utility function, which involves high-dimensional integration evaluated with sampling approaches, requiring many evaluations of the forward problem over many data realizations.

So the OED problem inherits all the challenges of solving the Bayesian inverse problems, and these challenges are exacerbated for nonlinear PDE-constrained inverse problems. Some studies have tried to answer the optimal design problem using Markov Chain Monte Carlo sampling [35] or other approaches that better reduce numerical cost [33]. Among these, Gaussian or Laplace approximations are often used to furnish a tractable closed-form approximation of the EIG [37, 38]. This leads to an optimal design with an optimality criterion that can be interpreted as minimizing the volume of the parameter uncertainty ellipsoid [28]. This is the D-optimal design criterion that is the focus of this work. In the context of mechanical testing, the D-optimal criterion has been used in [32, 39, 40] to develop optimal sample geometry or loading path. In the context of mechanical testing, other optimality criteria such as minimizing the average variance of the inferred parameters (related to A-optimality [41]) has been explored in shape optimization for linear elastic materials [42]. Other approaches of sample geometry design instead rely on optimizing some measure of strain heterogeneity, for example, [43–45].

In this paper, we propose a framework to design optimal specimen geometries for constitutive model identification by coupling Bayesian optimal experimental design with topology optimization. When coupled with Bayesian OED, topology optimization enables the exploration of a much larger design space, allowing the discovery of non-intuitive specimen topologies that can generate informative heterogeneous deformation fields. Instead of prescribing a small number of geometric parameters, such as hole size, boundary shape, or notch radius, we optimize over a high-dimensional material density field. In this way, the proposed approach moves beyond conventional geometries that sample sparse material states and instead provides complex geometry specimens to obtain information-rich data. Density-based topology optimization is a natural framework for this task, where a spatially varying material density field is the design variable and the objective is evaluated through a PDE-constrained forward problem. It has been widely developed for structural

mechanics problems; we refer readers to extensive discussions in [46]. Many engineering applications have leveraged density-based topology optimization, including minimizing the compliance of linear elastic structures [47], optimizing damage resistance [48], and designing phononic tailored band gaps in materials [49].

We demonstrate examples of the proposed framework through the design of specimen topologies for hyperelastic materials tested in a uniaxial tension device. Specifically, we seek specimens that are confined within a rectangular region, gripped at two edges and pulled apart. Note that the complex geometry creates complex states of stress, and thus one can gather a comprehensive range of information. We focus on anisotropic materials. Many biological soft tissues exhibit strongly anisotropic mechanical behavior as a result of collagen fibers in the tissue [50]. The orientations and arrangement of these collagen fibers result in a complex, direction-dependent behavior of the tissue. The determination of the fiber orientations and arrangements is essential to guide the development of high-fidelity biological implants. For example, in abdominal wall tissues, such as the human linea alba and rectus sheath, there exist interwoven collagen fibers with varying orientation [51], and determining the orientation of these fiber families is pertinent to guide the design of surgical meshes that better reproduce the native tissue mechanics and aid recovery [52]. Similarly, human skin tissue consists of different collagen fiber families with a particular orientation and arrangements [53]. However, identifying anisotropic constitutive behavior from macroscopic experimental data is challenging because the experiment must excite deformation states that are sensitive to the underlying preferred directions. Further, the constitutive models involve a significant number of material constants that must be inferred from those experiments. Approaches in the field incorporate microscopic imaging either during or before the mechanical testing [52, 54], which can provide more information, but often requires specialized experimental equipment and substantial post-processing that takes hours. These limitations motivate an alternative strategy to design complex specimen geometry so that a simple loading condition generates heterogeneous deformation fields that are informative about anisotropic material parameters.

The remainder of the paper is organized as follows. Section 8.2 formulates the general abstract formulation, with the parameter identification formulated as a Bayesian inverse problem in Section 8.2, and optimal experimental design in Section 8.2. We specialize to the problem of material parameter identification in Section 8.3. We introduce the general setting using topology optimization in 8.3, the material

parameter identification in Section 8.3 and the optimal experimental design in Section 8.3. The gradient based optimization approach is developed in Section 8.3. The implementation is summarized in Section 8.3. The method is demonstrated in Section 8.4 with isotropic materials in Section 8.4 and anisotropic materials in Section 8.4.

8.2 General Framework

We provide the background for the problem of optimal design of experiments for model discovery in this section. We assume that we have a parametrized model, and therefore model discovery reduces to parameter identification. We formulate this as a Bayesian inverse problem, and use it to formulate the problem of optimal experimental design. This section draws from [16, 31, 55].

Parameter identification as a Bayesian inverse problem

We consider the Bayesian inverse problem of estimating unknown parameters $P \in \mathbb{R}^{d_P}$ from experimental data. For a given experimental design ρ , we assume that the measured data $\tilde{y} \in \mathbb{R}^d$ follows the model equation

$$\tilde{y} = \mathcal{G}(P, \rho) + \eta, \quad (8.1)$$

where $\mathcal{G}$ is parameter-to-observable map that maps the unknown parameter vector P and the design variable ρ to observations, and η is the noise. $\mathcal{G}$ involves the forward model of the experiment, the finite element discretization, and the observation operator, and is nonlinear in general ¹. In this work, we assume that the noise is Gaussian with mean zero and covariance $\Gamma_{\text{noise}} \in \mathbb{R}^{d \times d}$, i.e., $\eta \sim \mathcal{N}(0, \Gamma_{\text{noise}})$. Under these assumptions, the data likelihood, or the distribution of experimental data $\tilde{y}$ for a given parameter set P and specified design ρ , is

$$\pi_{\text{like}}(\tilde{y}|P; \rho) \propto \exp\left(-\frac{1}{2}(\mathcal{G}(P, \rho) - \tilde{y})^T \Gamma_{\text{noise}}^{-1}(\mathcal{G}(P, \rho) - \tilde{y})\right). \quad (8.2)$$

We assume that we have some prior knowledge about the unknown variables, typically obtained from previous experiments, existing measurements, or domain knowledge, and then use π_{like} to refine our knowledge. We assume that we have an uncorrelated Gaussian prior on the parameters, with measure $\mu_0 = \mathcal{N}(P_0, C_0)$, characterized by mean P_0 and covariance C_0 . The prior mean represents the best guess about the unknown, while the covariance represents the uncertainty in that

¹We provide a characterization of this map for our problem in Section 8.3.

estimate. For the rest of this work, we only consider the case of a Gaussian prior and noise distribution.

According to Bayes' rule in our finite dimensional setting, the posterior distribution of the unknown parameter given the prior and the experimental observations admits a probability density

$$\pi_{\text{post}}(P|\tilde{y}; \rho) \propto \pi_{\text{like}}(\tilde{y}|P; \rho) \pi_{\text{prior}}(P), \quad (8.3)$$

where π_{prior} is the probability density associated with prior measure μ_0 . In general, this posterior probability distribution cannot be evaluated analytically when the forward map $\mathcal{G}$ is nonlinear. Still, there are a couple of useful observations that we can make.

The value of the parameter that maximizes the posterior distribution, the maximum a posteriori (MAP) estimate, is given by the variational problem,

$$\begin{aligned} P_{\text{MAP}} &= \arg \min O_P(P), \\ \text{where } O_P(P; \tilde{y}, \rho) &= \Phi(P; \tilde{y}, \rho) + \frac{1}{2} \left((P - P_0)^T C_0^{-1} (P - P_0) \right), \\ \text{and } \Phi(P; \tilde{y}, \rho) &= \left(\frac{1}{2} (\mathcal{G}(P, \rho) - \tilde{y})^T \Gamma_{\text{noise}}^{-1} (\mathcal{G}(P, \rho) - \tilde{y}) \right). \end{aligned} \quad (8.4)$$

The expression Φ in O_P is the negative log-likelihood (see Eq. 8.2) from a Bayesian perspective. From the perspective of classical optimization, this objective is the regularized data misfit functional Φ with a quadratic regularization.

Further, if $\mathcal{G}$ were linear, the posterior would be Gaussian with mean P_{MAP} . Therefore, it is common to use a Laplace approximation (see [34, 37, 38]) where we approximate the posterior with a Gaussian (or $\mathcal{G}$ with a linear map). This Gaussian approximation of the posterior is $\mathcal{N}(P_{\text{MAP}}, C_{\text{post}})$ where

$$C_{\text{post}} = \left(H(P_{\text{MAP}}) + C_0^{-1} \right)^{-1}, \quad (8.5)$$

and H is the Hessian of the negative log-likelihood ($\Phi(P; \tilde{y}, \rho)$) evaluated at the MAP point.

Optimal experimental design (OED)

The Bayesian inverse problem discussed previously assumes a fixed experimental design ρ . We now seek to determine the experimental design ρ such that the

resulting observed data are maximally informative about the unknown variables. We use the expected information gain (EIG) as the suitable utility function that provides us with a measure of the information content of the next experiment [31]. This expected information gain (EIG) is defined as the expected change in the Shannon entropy between the prior to the posterior distributions [31] over all possible realizations of data. Equivalently, it can be defined by the expected Kullback-Leibler divergence (KL divergence) over all possible experimental data realizations[55]. The KL divergence itself provides a distance measure between the posterior and prior probability density [56]. We have,

$$EIG := \int_{\mathbb{R}^{d_P}} \int_{\mathbb{R}^d} D_{kl}(\pi_{\text{post}}(P|\tilde{y}; \rho) || \pi_{\text{prior}}) \pi_{\text{like}}(\tilde{y}|P; \rho) d\tilde{y} \pi_{\text{prior}}(P) dP, \quad (8.6)$$

$$\text{where } D_{kl}(\pi_{\text{post}} || \pi_{\text{prior}}) = \int_{\mathbb{R}^{d_P}} \log \left(\frac{\pi_{\text{post}}(P|\tilde{y}; \rho)}{\pi_{\text{prior}}(P)} \right) \pi_{\text{post}}(P|\tilde{y}; \rho) dP. \quad (8.7)$$

Note that the evaluation of the EIG requires knowledge of the posterior measure after obtaining data in the next experiment. Typically, this is computed by sampling across all possible future data realizations, which is numerically expensive. However, in the linear setting where $\mathcal{G}$ is linear with Gaussian noise and prior, the resulting posterior measure is also Gaussian with covariance given in (8.5). It follows that we can now characterize the EIG as

$$EIG \approx O_{\text{EIG}} := \frac{1}{2} \log \left(\frac{\det C_0}{\det C_{\text{post}}} \right) = \frac{1}{2} \log \det(I + C_0 H(P_{\text{MAP}})). \quad (8.8)$$

This is often commonly known as D-optimal criteria [28], i.e., maximizing the EIG is equivalent to minimizing the determinant of the posterior covariance. We note that this expression is exact if $\mathcal{G}$ is linear; for $\mathcal{G}$ nonlinear, (8.8) is obtained by linearizing about the current estimate and using the Gauss-Newton approximation for H .

As we shall see in the following section, $\mathcal{G}$ is nonlinear, and therefore, we adopt the local Bayesian D-optimal approximation. We start with the current best estimate of the parameter and the covariance; this is our prior $\mu_{\text{prior}} = \mathcal{N}(P_0, C_0)$. We then create a design to maximize the expected information gain (8.8), conduct an experiment, and then update our model parameters. This OED approach can be iterated, leading to a sequential local Bayesian D-optimal approach. After k iterations, the prior parameter estimate $P_0 = P_k$ and prior covariance $C_0 = C_k$ act as priors in the problem to design specimen with ρ_{k+1} for the next experiment.

8.3 Application to materials parameter identification

Setting

We apply the framework of Section 8.2 to the problem of identifying the material parameters of a hyperelastic material using mechanical tests. We seek to design the shape of the specimen that enables us to optimally identify the material properties.

We assume that our specimen is confined to a region $\Omega \subset \mathbb{R}^n$. We specify the natural reference configuration of the body to be $\Omega_\chi = \{x \in \Omega : \chi(x) = 1\}$ where $\chi : \Omega \rightarrow \{0, 1\}$, and assume that the constitutive behavior of the body is described by a stored energy density $W(F; P)$ where $F = I + \nabla u$ is the deformation gradient, and u is the displacement field. We assume that the specimen is fixed at $\partial_0\Omega$, and subject to a time-dependent Dirichlet boundary condition $y(x, t) = \bar{y}(x, t)$ on another part $\partial_u\Omega$ (so we restrict $\chi = 1$ on $\partial_u\Omega \cup \partial_0\Omega$). The remainder of the boundary of the specimen is traction-free. We limit ourselves to the quasistatic setting and therefore the displacement $u : \Omega_\chi \rightarrow \mathbb{R}^n$ satisfies

$$\nabla \cdot W_F(F; P) = 0 \text{ on } \Omega_\chi \quad (8.9)$$

subject to the boundary conditions where $W_F = \partial W / \partial F$.

Our goal is to design the specimen or the function χ . Unfortunately, optimal design problems are known to be nonconvex, and therefore we relax this problem using topology optimization and SIMP interpolation [46]. We approximate χ using a function $\rho : \Omega \rightarrow [\rho_{\min}, 1]$ and assume that the body occupies the entire domain with energy density

$$W(x, F; P) = \rho^p(x) W(F; P), \quad (8.10)$$

where p is the SIMP exponent to aid topology optimization. Equilibrium is given by (8.9) on Ω , or

$$-\int_{\Omega} (\rho^p W_F(F; P) \cdot \nabla \varphi) \, d\Omega = 0 \quad \forall \varphi \in \mathcal{U} = \{u = 0 \text{ on } \partial_0\Omega \cup \partial_u\Omega\}. \quad (8.11)$$

Now, our design is given by the function ρ .

Parameter identification

We now specify the objective $\mathcal{O}_p$ that we use for parameter identification. In a typical mechanical test, we decorate part of the boundary with a speckle pattern $g_0(x)$, and image the convected pattern $g(y, t)$ during deformation (where $y = x + u$), and also measure the reaction force $f(t)$ in some direction e_f on $\partial_u\Omega$ where the displacement is applied. There are two possible formulations.

- *Image-based formulation.* We follow Wihardja and Bhattacharya [15] and consider the raw images and boundary forces as the experimental data $\tilde{y} = \{g(y, t), f(t)\}$. We consider time-continuous measurements common in material testing (though this is easily specialized to a discrete time measurement). The parameter to observable map (8.1) is

$$P \rightarrow \{g(x + u(x, t), t), f_u(t)\}, \quad (8.12)$$

$$\text{where } f_u(t) = \int_{\partial\Omega_u} W_F(\nabla u(x, t); P) n \cdot e_f dA \quad (8.13)$$

is the reaction force on $\partial_u\Omega$ in the direction e_f and u solves the governing equation (8.11). Note that this map is nonlinear due to the nonlinearity of the governing equation (8.11) as well as the speckle pattern g . Therefore, the functional O_P in (8.4 for Bayesian parameter identification becomes

$$\begin{aligned} O_P &= \Phi_d + \Phi_f + \frac{1}{2}(P - P_0)^T C_0^{-1}(P - P_0), \\ \Phi_d &= \frac{1}{2} \int_0^t \int_{\Omega} \rho^{2p} (g_0(x) - g(y(x, t), t))^T \Gamma_i(t)^{-1} \\ &\quad (g_0(x) - g(y(x, t), t)) d\Omega dt, \\ \Phi_f &= \frac{1}{2} \int_0^t \left(f_u(t) - f_{\text{exp}}(t) \right)^T \Gamma_f(t)^{-1} \left(f_u(t) - f_{\text{exp}}(t) \right) dt, \end{aligned} \quad (8.14)$$

where $\Phi = \Phi_d + \Phi_f$ is the total negative log-likelihood/data misfit with time-continuous data, Φ_d is the image mismatch between model prediction and experiments, Φ_f is the force mismatch, Γ_i is the noise covariance associated with grayscale intensity values of images, and Γ_f is the noise covariance associated with measured force. The prior mean of the parameter is P_0 with prior covariance C_0 . Note that we have weighted the images in Φ_d with ρ since these are only observed in the actual design domain.

- *Displacement-based formulation.* It is common practice to use a two-step approach in experimental mechanics. We first use the images to infer the displacement using Digital Image Correlation (DIC) [9–11, 57]. We then regard the displacement field as the experimental data in the parameter identification problem. We note that the first inverse problem (image to displacement) is known to be ill-posed [15], and so is regularized by constraints and filters. Consequently, it is difficult to quantify the noise associated with the displacement field data. In particular, the noise may be correlated based on the constraint. Still, this is widely used, and therefore we study this as well.

In this setting, the data are $\tilde{y} = \{u_{DIC}(x, t), f(t)\}$ and the objective in Eq. 8.14 has different data misfit term,

$$\Phi_d = \frac{1}{2} \int_0^t \int_{\Omega} \rho^{2p} (u_{DIC}(x, t) - u(x, t))^T \Gamma_u(t)^{-1} (u_{DIC}(x, t) - u(x, t)) d\Omega dt, \quad (8.15)$$

where Γ_u is the noise covariance associated with the displacement-field measurements.

In what follows, we use a displacement-based formulation for our development since this leads to more compact expressions. However, the approach extends easily to the image-based formulation, and we evaluate both in our examples.

Given a proposed design ρ , since no data from a ρ -experiment exists yet, we predict the covariance from that design at the current best estimate P_0 ,

$$C_{\text{post}}(\rho) = \left(H(\rho) + C_0^{-1} \right)^{-1}, \quad (8.16)$$

where the Hessian in our setting is

$$H = \int_0^t \left(\int_{\Omega} \left(\rho^{2p} J^T \Gamma_u^{-1} J \right) d\Omega + J_f^T \Gamma_f^{-1} J_f \right) dt, \quad (8.17)$$

with $J = u_P := \frac{\partial u}{\partial P},$

$$J_f = \frac{\partial f}{\partial P} = \int_{\partial_u \Omega} (W_{FF}(\nabla u, P_0) \nabla u_P + W_{FP}(\nabla u, P_0)) \hat{n} \cdot e_f dA.$$

Once a design ρ_{best} is selected and an experiment is performed, we can now compute the updated MAP estimate as the minimizer of (8.14),

$$P_{\text{MAP}} = \arg \min_P \mathcal{O}_{\mathcal{P}}(P; u, \rho) \quad \text{such that } u \text{ satisfies (8.11) for } \rho. \quad (8.18)$$

Note that the Hessian H is a functional of ρ , u and u_P , or $H(\rho, u, u_P)$ at the current estimate P_0 . Also, (8.18) is a PDE-constrained optimization problem, and (8.17) requires the solution of (8.11) as well as the sensitivity of this solution with respect to P .

Optimal experimental design

We obtain the optimal experimental design for parameter identification under the local Bayesian D-optimal approximation. At the start of the problem, we have

the current parameter estimate P_0 and covariance C_0 . We can now specialize the expected information gain (8.8) to our setting,

$$O_{EIG}(\rho, u, u_P) = \frac{1}{2} \log \det(C_0 C_{\text{post}}^{-1}(\rho)) = \frac{1}{2} \log \det(I + C_0 H(\rho, u, u_P)), \quad (8.19)$$

where H is given in (8.17). We now have our optimal experimental design problem,

$$\rho_{\text{best}} = \arg \min_{\rho} -O_{EIG}(\rho, u, u_P) \quad \text{such that } u \text{ satisfies (8.11) for } \rho. \quad (8.20)$$

Note that this is a PDE-constrained optimization problem where u, u_P depend on ρ through (8.11).

Gradient-based optimization with adjoint method

The considerations so far lead to an iterative method where we solve (8.20) for the design, perform an experiment, and then solve (8.18) for estimating the parameters. Note that both (8.20) and (8.18) are PDE-constrained optimization problems. We solve them using a gradient-based approach that exploits the adjoint method to compute the gradients.

The functional O_{EIG} depends on u and $\partial u / \partial P$. Therefore, we have two forward problems,

F1. The displacement field u satisfies

$$\int_{\Omega} (\rho^P W_F(\nabla u, P_0)) \cdot \nabla \varphi \, d\Omega = 0 \quad \forall \varphi \in \mathcal{U}. \quad (8.21)$$

F2. The sensitivity of displacement to the parameter u_P satisfies the following equation obtained by differentiating the above:

$$\int_{\Omega} \left(\rho^P (W_{FP}(\nabla u, P_0) + W_{FF}(\nabla u, P_0) \nabla u_P) \right) \cdot \nabla \varphi \, d\Omega = 0 \quad \forall \varphi \in \mathcal{U}, \quad (8.22)$$

where u is the solution of (8.21). We note in passing that (8.22) may be viewed as an adjoint equation to (8.21) if we were confined to parameter identification.

We now turn to finding the gradient of O_{EIG} with respect to ρ . This requires care, since u, u_P depend on ρ , and we use the adjoint method. Given any $\lambda, \beta_P \in \mathcal{U}$, we

use the forward problems (8.21 and (8.22) to rewrite our objective as

$$\begin{aligned} \mathcal{O}^*(\rho, u, u_P) &= -\mathcal{O}_{EIG}(\rho, u, u_P) + \int_{\Omega} (\rho^p W_F(\nabla u, P_0)) \cdot \nabla \lambda \, d\Omega \\ &+ \int_{\Omega} (\rho^p (W_{FP}(\nabla u, P_0) + W_{FF}(\nabla u, P_0) \nabla u_P)) \cdot \nabla \beta_P \, d\Omega. \end{aligned} \quad (8.23)$$

We perturb ρ in the direction μ ($\rho \mapsto \rho + \varepsilon \mu$ for small ε) and assume that the solutions of the forward problems F1 and F2 are continuous with respect to ρ so that the corresponding solutions are $u + \varepsilon v$ and $u_P + \varepsilon v_P$. The variation of $\mathcal{O}^*$ with respect to ρ in the direction μ is given by

$$\langle \delta_{\rho} \mathcal{O}^*, \mu \rangle := \left. \frac{d}{d\varepsilon} \mathcal{O}^*(\rho + \varepsilon \mu, u + \varepsilon v, u_P + \varepsilon v_P) \right|_{\varepsilon=0}. \quad (8.24)$$

A long, but straightforward calculation shown in Appendix 8.A shows that the sensitivity of the objective to the design is given by

$$\begin{aligned} \langle \delta_{\rho} \mathcal{O}^*, \mu \rangle &= \int_{\Omega} p \rho^{p-1} \left(-\frac{1}{2} C_0 (I + C_0 H)^{-T} \cdot \int_0^t 2 \rho^p u_P^T \Gamma_u^{-1} u_P \, dt \right. \\ &\quad + W_F(\nabla u, P_0) \cdot \nabla \lambda \\ &\quad + \left(W_{FP}(\nabla u, P_0) \right) \cdot \nabla \beta_P \\ &\quad \left. + \left(W_{FF}(\nabla u, P_0) \nabla u_P \right) \cdot \nabla \beta_P \right) \mu \, d\Omega, \end{aligned} \quad (8.25)$$

where λ, β_P satisfy the following adjoint equations.

A1. The adjoint equation for β_P is

$$\begin{aligned} &-\frac{1}{2} C_0 (I + C_0 H)^{-T} \cdot \left(\int_0^t \int_{\Omega} 2 \rho^{2p} u_P \Gamma_u^{-1} v_P \, d\Omega \, dt \right. \\ &\quad \left. + \int_0^t 2 \left(\int_{\partial_u \Omega} (W_{FF} \nabla v_P) \hat{n} \cdot e_f \, dA \right) \Gamma_f^{-1} J_f \, dt \right) \\ &\quad + \int_{\Omega} (\rho^p W_{FF} \nabla v_P) \cdot \nabla \beta_P \, d\Omega = 0 \quad \forall v_P \in \mathcal{U}. \end{aligned} \quad (8.26)$$

A2. With β_P as in (8.26), the adjoint equation for λ is

$$\begin{aligned}
& -\frac{1}{2}C_0(I + C_0H)^{-T} \cdot \\
& \left(\int_0^t 2J_f^T \Gamma_f^{-1} \int_{\partial_u \Omega} ((W_{FFF} \nabla u_P + W_{FFP}) \nabla v) \hat{n} \cdot e_f \, dA \, dt \right) \\
& + \int_{\Omega} \rho^p (W_{FF} \nabla v) \cdot \nabla \lambda \\
& + \int_{\Omega} \rho^p (W_{FFF} \nabla v \nabla u_P + W_{FFP} \nabla v) \cdot \nabla \beta_P \, d\Omega = 0 \quad \forall v \in \mathcal{U}.
\end{aligned} \tag{8.27}$$

Implementation

We choose a suitable reference domain or ground structure that allows for the definition of boundary conditions. We specify parts of the ground structure that can be designed or left as solids/voids. We construct a sufficiently fine finite element mesh of the ground structure such that the structure can be described in a reasonable bit-map representation. This mesh is unchanged throughout the entire optimization procedure. We discretize the displacement finite element as a standard piecewise polynomial Lagrange basis function of degree 1, and the design variable ρ as piecewise constant on each element. All PDEs are solved using a finite element method in deal.II [58].

In performing topology optimization, we use the SIMP interpolation method [59], where the design density is scaled by an interpolation power ρ^p . We initialize the optimization with $p = 1$ and increase it gradually to $p_{\max} = 3$, following the standard practice [46] to promote convergence toward a solid-void design. In all examples, p is increased by 0.1 every 20 iterations. At each optimization iteration, we use the method of moving asymptotes (MMA) [60] based on the sensitivity (8.25) to update the design. We then filter it with a renormalized filter with a linear weight function [61] to avoid the development of fine-scale structures due to the ill-posedness of the topology optimization. Once $p_{\max}$ is reached, the optimization is continued until either the design sensitivity or the incremental objective reaches $\varepsilon = 10^{-4}$ and the design is sufficiently well-defined. We summarize our approach in Algorithm 2.

All computations were performed on a workstation with a 12th Gen Intel Core i9-12900 CPU (16 cores / 24 threads, up to 5.1 GHz) and 64 GB RAM. For the mesh size used here (96×32 elements, with a total of 6402 DOFs), one MMA iteration consisting of the two forward solves ($F1, F2$), assembly of the Hessian, two adjoint solves ($A1, A2$), and the design update itself requires approximately 50 seconds.

Algorithm 2 Optimal Experimental Design

Input: Reference domain Ω with triangulation, current estimate of parameters of the model P_0 with covariance C_0 and current design ρ

Output: Updated design field ρ_{best}

Initialize Design $\rho(x)$, current model parameters P_0 , and initial SIMP power $p = 1$;

while $p < p_{\max}$ **and** $\|\delta_\rho O_{\text{EIG}}\|$ **or** $\|O_{\text{EIG},k+1} - O_{\text{EIG},k}\| > \varepsilon$ **do**

 Step 1: Filter the design density field ρ ;

 Step 2: Solve forward problems F1, F2 for u and u_P ;

 Step 3: Compute $C_{\text{post}}(\rho)$ (Eq. 8.16) and objective O_{EIG} (Eq. 8.19);

 Step 4: Solve the adjoint problems A1, A2 for β_P and λ ;

 Step 5: Compute design sensitivity $\delta_\rho O_{\text{EIG}}$ at each element;

 Step 6: Update design density field ρ with MMA at each element;

 Step 7: Every 20 MMA iterations, update SIMP power $p \leftarrow \min(p + 0.1, p_{\max})$

end while

Convergence of the final solid-void design (SIMP exponent increased from 1 to 3 over 400 iterations) therefore took approximately 5 hours on this hardware.

8.4 Demonstration on hyperelastic materials

We now demonstrate the proposed approach on hyperelastic materials. We demonstrate both the image-based and displacement-based approaches. We study two examples, one isotropic and one anisotropic. In all our examples, the reference domain is rectangular, $\Omega = 0.036 \times 0.012 \text{ m}^2$, with one edge held fixed and the other subjected to uniaxial extension as shown in Figure 8.1. In some of our examples, we fix a strip near the boundary of the domain to be fixed at $\rho = 1$ (solid), as shown in Figure 8.1(a), and design only the interior. In other examples, we only fix the edges at the grips.

While the method above used an iterative procedure, we will conduct one iteration in this demonstration. We show that a single iteration already yields meaningful designs.

Isotropic material

We assume

$$W = \frac{\mu}{2} \text{tr}(F^T F - 3 - 2 \log J) + \frac{k}{2} (J - 1)^2, \quad (8.28)$$

where $J = \det F$ in three dimensions, and specialize to plane stress. The parameters to be estimated are $P = \{\mu, k\}$.

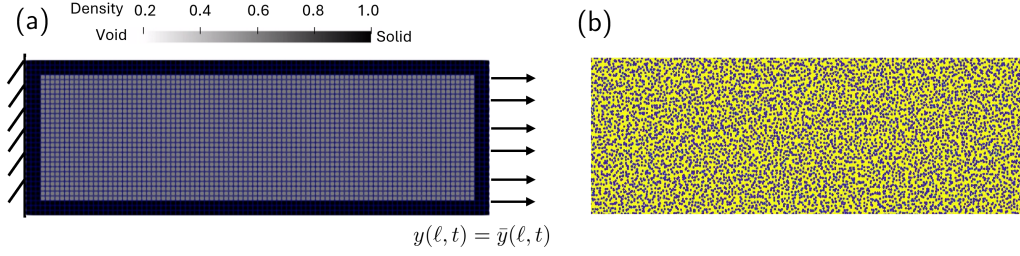


Figure 8.1: (a) Reference domain of $0.036 \times 0.012 \text{ m}^2$, with finite element mesh consisting of 48×16 elements. An example density assignment to initiate the optimization can be seen in the scale bar. (b) Representative of speckle pattern to perform image-based optimal designs (15×15 pixels within each mesh), with 4×4 pixels per speckle.

We assume that our prior (assumed Gaussian) is characterized by

$$m_0 = \{\mu^0, k^0\} = \{4 \text{ MPa}, 8.67 \text{ MPa}\},$$

$$C_0 = \begin{bmatrix} \sigma_\mu^2 & 0 \\ 0 & \sigma_k^2 \end{bmatrix}, \quad \sigma_\mu = \sigma_k = 1 \text{ MPa}. \quad (8.29)$$

Similarly, we assume that our noise (assumed Gaussian and uncorrelated) has zero mean with covariance

$$\Gamma_i = 0.01^2 \mathbf{I}, \quad \Gamma_u = (10^{-5})^2 \mathbf{I}, \quad \Gamma_f = (0.025t)^2 \quad (8.30)$$

for the images (in image-based formulation), displacements (in displacement-based formulation), and force, respectively. Note that we have taken the force to increase with time; we anticipate our force to increase, and the noise on the typical load cells increases with force magnitude. Note that we consider a uniaxial extension setting, so we take $e_f = e_1$ in (8.13) and neglect the contribution of the other components.

The results of our simulations are shown in Figure 8.2. The top row shows the designs when we constrain the entire boundary to have fixed density and start with a uniform initial guess. The second row retains the same constraint, but starts with an initial configuration consisting of a hole. The resulting designs are similar, showing that in this case the design is independent of the initial guess. Both the image- and displacement-based formulations lead to initial designs that have two strips that are largely uniform through the length. The third row shows a design when we use an initial dogbone design and remove the constraint on the top and bottom.

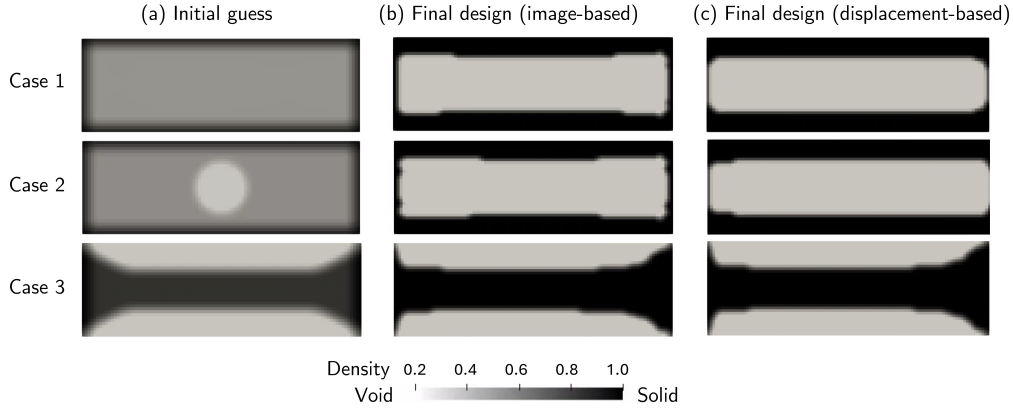


Figure 8.2: Optimal design for isotropic material. (a) Initial design (density assignment) for optimization. (b) Resulting optimal topology with images as raw data, and (c) with displacements as raw data.

We follow the SIMP interpolation as described in Section 8.3 and increase the SIMP exponent from 1 to 3 over the first 400 iterations. We obtain the final solid-void designs within the next 100 to 400 iterations, depending on the initial design guess.

We now turn to evaluate the performance of the optimal design in inferring parameters from experiments by using synthetic data. We set the reference parameters and perform numerical simulations to compute the experimental observables (image and force) and add noise to them consistent with our assumptions. Then, these synthetic data are used to recover the parameters following the image-based inference procedure [15]. We chose the image-based design (case A of Figure 8.2b) as an example to compare with a random design with the same volume fraction. The inferred shear modulus has a 0.3% error for both the optimal design and the random design, while the error in bulk modulus is 4% and 10% for the optimal and random design. We see that the optimized design can lead to more accurate parameter inference.

Fiber-reinforced material

We next consider an anisotropic hyperelastic material with two fiber families in the plane of the specimen, with stored energy density

$$W = C_1(\bar{I}_1 - 3) + C_4(\bar{I}_4 - 1)^2 + C_6(\bar{I}_6 - 1)^2 + \frac{\kappa}{2}(J - 1)^2, \quad (8.31)$$

where $\bar{I}_1 = \text{tr}(\bar{C})$, $\bar{I}_4 = a \cdot \bar{C}a$, $\bar{I}_6 = b \cdot \bar{C}b$, $J = \det F$ for $\bar{C} = J^{-4/3}C$, $a = (\cos \theta_1, \sin \theta_1, 0)$ and $b = (\cos \theta_2, \sin \theta_2, 0)$. We specialize to plane stress. The parameters to be estimated are $P = \{C_1, C_4, C_6, \kappa, \theta_1, \theta_2\}$.

Case	Description
Various priors	
1	Baseline (8.32): $\{C_1^0, C_4^0, C_6^0, \kappa^0, \theta_1^0, \theta_2^0\} = \{4 \text{ MPa}, 8 \text{ MPa}, 8 \text{ MPa}, 13 \text{ MPa}, \pi/4, -\pi/6\}$
2	$\theta_1^0 = \pi/3$, remaining parameters identical to the baseline.
3	$C_6^0 = 16 \text{ MPa}$, remaining parameters identical to the baseline.
4	$\{C_1^0, C_4^0, C_6^0, \kappa^0\} = \{4.5 \text{ MPa}, 9 \text{ MPa}, 9 \text{ MPa}, 15 \text{ MPa}\}$, remaining parameters identical to the baseline.
Various initial designs and constraints	
5	Initial design I1; edge constraint.
6	Initial design I2; edge constraint.
7	Initial dogbone design; grip constraint.

Table 8.1: Cases considered using the displacement formulation.

Unless otherwise stated, the mean and covariance of our prior (Gaussian and uncorrelated) are given by

$$\begin{aligned}
m_0 &= \{C_1^0, C_4^0, C_6^0, \kappa^0, \theta_1^0, \theta_2^0\} \\
&= \{4 \text{ MPa}, 8 \text{ MPa}, 8 \text{ MPa}, 13 \text{ MPa}, \pi/4, -\pi/6\}, \\
C_0 &= \text{diag}(\sigma_{C_1}^2, \sigma_{C_4}^2, \sigma_{C_6}^2, \sigma_{\kappa}^2, \sigma_{\theta_1}^2, \sigma_{\theta_2}^2),
\end{aligned} \tag{8.32}$$

where the variance is 20% of mean value (for example, $\sigma_{C_1} = 20\% C_1^0 = 0.8 \text{ MPa}$). We assume a Gaussian noise distribution $\eta \sim \mathcal{N}(0, \Gamma)$ for all data measurements.

The image-based formulation often led to non-convergence. The sensitivity $\delta_\rho \mathcal{O}_{\text{EIG}}$ requires us to differentiate the images, and this leads to extremely noisy sensitivity.

Therefore, we confine ourselves to displacement-based formulation in this section. We assume spatio-temporally uncorrelated Gaussian noise $\Gamma_f = (0.01t)^2 \mathbf{I}$ for the force measurements: as before, it increases with time since the noise in a typical load cell increases with force. We consider two types of noise for the displacement, both spatio-temporally uncorrelated and Gaussian. The first is constant noise $\Gamma_u = (10^{-5})^2 \mathbf{I}$, and the second is linearly increasing $\Gamma_u = (10^{-6} \times t)^2 \mathbf{I}$ to reflect the increasing noise with increasing displacement in a typical DIC inversion.

The results are shown in Figure 8.3 for various priors. Each of these designs is subject to the constraint that the edges are solid, and the initial guess is uniform. We observe that despite the fact that the priors vary by about 15%, the resulting designs are similar. This is significant since it suggests that we get meaningful designs

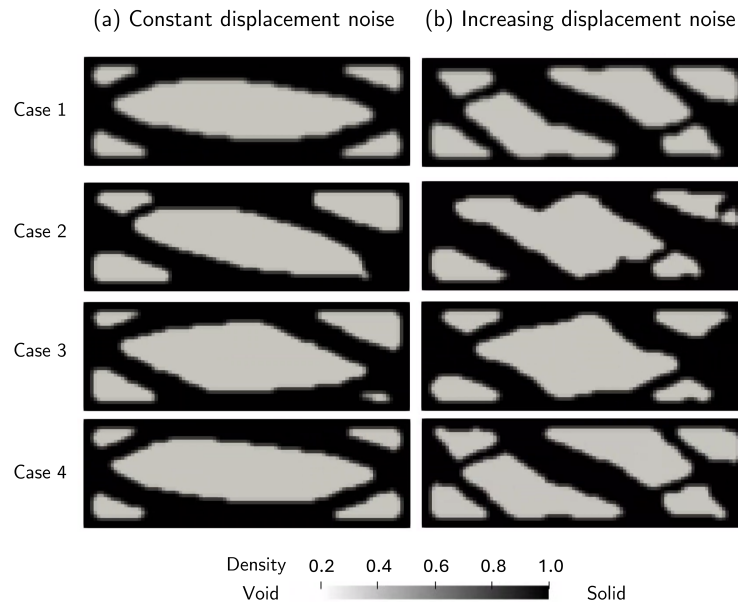


Figure 8.3: Optimal design for fiber-reinforced hyperelastic model in the displacement formulation with (a) constant and (b) increasing noise. The rows depict different priors as noted in Table 8.1. All these designs use a uniform density as an initial guess and constrain the edges to be solid.

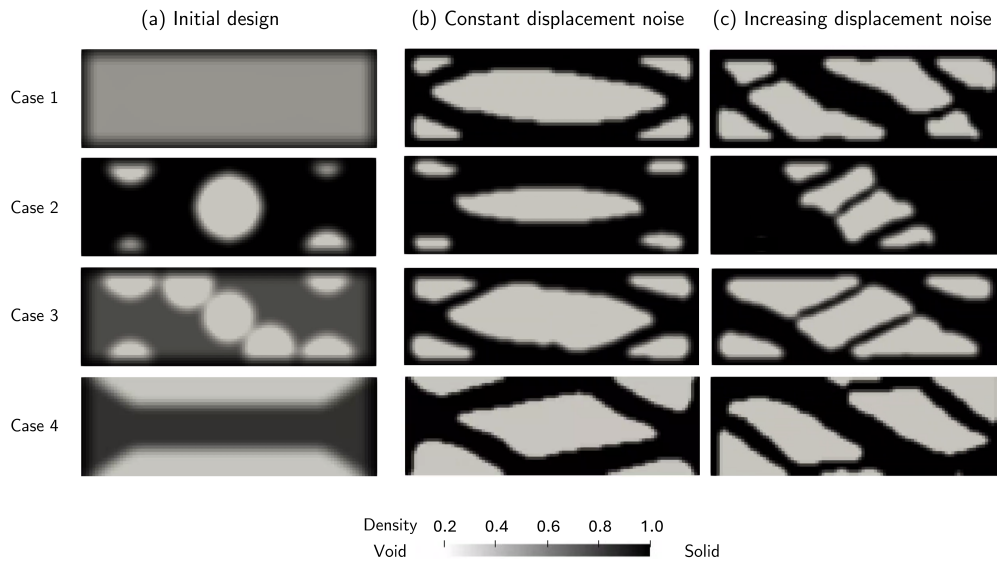


Figure 8.4: Optimal design for fiber-reinforced hyperelastic model in the displacement formulation with (b) constant and (c) increasing noise. The rows depict different initial designs and constraints as noted in Table 8.1.

even without iteration. Turning to the designs, we observe that they all have linear elements aligned in multiple directions to be able to assess the anisotropy. Finally, the uniform displacement noise results in a simpler design compared to the case of increasing displacement noise.

Figure 8.4 shows the result for various initial guesses, and for different design constraints. While the details of the design depend on these, the overall designs are all very similar. Again, we observe that they all have linear elements aligned in multiple directions to be able to assess the anisotropy.

We now turn to understanding the efficacy of our optimal designs. As a representative example, we consider the optimal grip-constrained design (case 7, Figure 8.4). For constant noise, the expected information gain O_{EIG} increases from 9.5 for the initial to 11.3 for the final. For increasing noise, O_{EIG} increases from 11 for the initial to 12 for the final. Thus, the optimization does lead to an increase in the expected information gain. We compare the performance of our optimized design with a number of arbitrary designs in Table 8.2. In each case, the optimized design has the highest expected information gain.

Another indicator is the trace of the expected posterior covariance. This is often termed the A-optimal criterion that minimizes the mean square error of the inferred parameters [62]. We split this into the trace of the part related to moduli and the part related to orientations, and note that these are related to mean square error. These are also shown in Table 8.2. Again, in each case, we notice that our optimized designs perform better than the arbitrary design. Thus, we observe that optimizing under the D-optimal criterion also leads to a better performance on the A-optimal criterion.

These indicators above depend only on the prior and do not involve any experimental evaluation. However, our goal is to infer the parameters from the experiment. We use synthetic data to evaluate this. We use a set of reference parameters and numerical simulation to compute the experimental observables (image, displacement and force), and add noise to them consistent with our assumptions to create synthetic data. We use this synthetic data to recover our parameters. We can do so in two ways: either using the image-based formulation or the displacement-based formulation for parameter inference². Table 8.3 shows the error in the inferred parameters,

$$\frac{P_{\text{MAP}} - P_{\text{true}}}{P_{\text{true}}}, \quad (8.33)$$

²The designs are optimized using displacement-based formulation for both cases.

for the optimal design (case 7 in Figure 8.4) for two sets of reference parameters. Finally, this table also compares the inference using a couple of arbitrary designs (A2 and A4 in Table 8.2) with that of the optimized design in case 1 (Figure 8.3) under constant noise. We see that the optimized designs lead to more accurate parameter inference in each case.

In summary, our optimization leads to better expected information gain as well as more accurate parameter inference.

8.5 Conclusion

We have developed an approach to design specimens to enable efficient and accurate recovery of material parameters from experiments. We pose the problem of material parameter identification as a Bayesian inverse problem, and cast the problem of specimen design as a D-optimal experimental design problem solved using topology optimization. We demonstrate our approach on isotropic and anisotropic hyperelastic materials, and show that the optimized specimens increase the expected information gain and provide more accurate parameter inference compared to conventional or non-optimized designs.

The results in this work suggest that direct image-based inference is valuable to obtain high-fidelity constitutive models, while displacement-based data may provide a more stable and practical route for OED, leading to more reliable designs. For model parameter inference, raw images contain rich, fine-scale information and, when used within the image-to-constitutive integrated framework (e.g., [15]), avoid the intermediate step to obtain displacements. However, using raw images to guide optimal experimental design is challenging since design sensitivity depends on the image intensities, and differentiating them leads to noisy gradients that poorly influence the topology optimization.

We close by discussing future extensions building on this framework. A natural extension is to consider history-dependent materials, such as plasticity and rate-dependent soft materials. In these complex systems, the constitutive response depends not only on the current deformation but also on internal states and loading history, and is thus more challenging to characterize. This added complexity opens interesting directions. First, the design problem should consider not only the specimen topology, but also the loading path and rate to sufficiently explore relevant evolution laws. Second, this increased complexity calls for the development of an adaptive and fast data assimilation framework that continuously integrates experi-

Edge constraint, $V_f = 0.6$						
	Case 1 (Fig. 8.3)	Design A1	Design A2	Design A3	Design A4	Design A5
<i>Constant noise</i>						
$\mathcal{O}_{EIG}$	12	10.5	11.4	10.4	11	10.6
MSE_{mod}	8.2	10	9.75	10	9.6	10
MSE_{orient}	0.002	0.009	0.006	0.012	0.006	0.01
<i>Increasing noise</i>						
$\mathcal{O}_{EIG}$	11	9.7	10	9.6	9.9	9.8
MSE_{mod}	10	10.8	10.7	10.9	10.9	11
MSE_{orient}	0.005	0.04	0.035	0.035	0.034	0.034
Grip constraint, $V_f = 0.5$						
	Case 7 (Fig. 8.4)	Design B1	Design B2	Design B3	Design B4	Design B5
<i>Constant noise</i>						
$\mathcal{O}_{EIG}$	12	11	10.9	9.9	10.4	11
MSE_{mod}	8.7	10	10.2	10.4	9.8	9.8
MSE_{orient}	0.003	0.008	0.0083	0.016	0.01	0.007
<i>Increasing noise</i>						
$\mathcal{O}_{EIG}$	11.3	9.6	9.6	9.4	9.7	9.9
MSE_{mod}	9.8	11	11	11	11	10.8
MSE_{orient}	0.001	0.06	0.07	0.04	0.025	0.04

Glossary:

$MSE_{\text{mod}} = \text{Posterior } (\sigma_{C_1}^2 + \sigma_{C_4}^2 + \sigma_{C_6}^2 + \sigma_{\kappa}^2) \text{ MPa}^2$

$MSE_{\text{orient}} = \text{Posterior } (\sigma_{\theta_1}^2 + \sigma_{\theta_2}^2)$

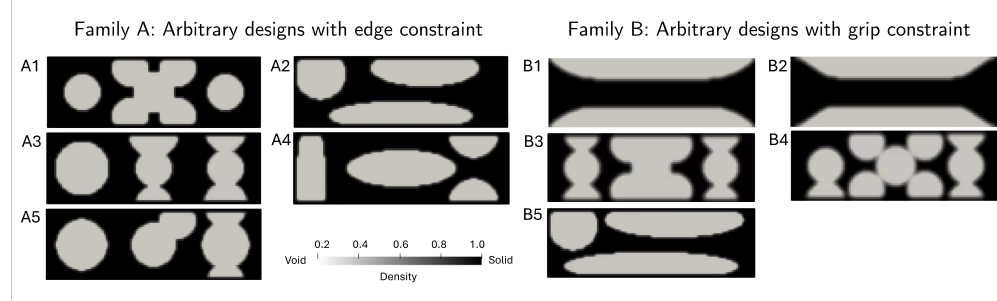


Table 8.2: Evaluation of the optimal designs: comparison of the expected information gain and uncertainty reduction of the optimized designs against those of arbitrary designs.

	C_1	C_4	C_6	κ	θ_1	θ_2
Case 7: Displacement-based design, Displacement-based recovery						
$P_{\text{true}}=\{C_1 = 4.5, C_4 = C_6 = 9, \kappa = 14.6 \text{ MPa}, \theta_1 = 40^\circ, \theta_2 = 27^\circ\}$						
Initial	3.1	2.2	2.6	2.6	15	0.3
Final (constant noise)	2.6	1.0	1.6	1.2	15	0.3
$P_{\text{true}}=\{C_1 = 3.5, C_4 = C_6 = 7, \kappa = 11.4 \text{ MPa}, \theta_1 = 43^\circ, \theta_2 = 31^\circ\}$						
Initial	3.7	6.7	5.9	6.4	9.6	3.8
Final (constant noise)	0.6	5.5	4.6	5.5	7.4	3.7
Case 7: Displacement-based design, Image-based recovery						
$P_{\text{true}}=\{C_1 = 4.5, C_4 = C_6 = 9, \kappa = 14.6 \text{ MPa}, \theta_1 = 40^\circ, \theta_2 = 27^\circ\}$						
Initial	2.7	3.7	9.8	3.0	4.0	8.8
Final (constant noise)	2.2	2.3	3.5	1.5	4.0	8.7
Final (increasing noise)	2.2	0.3	1.8	2.0	5.0	8.75
$P_{\text{true}}=\{C_1 = 3.5, C_4 = C_6 = 7, \kappa = 11.4 \text{ MPa}, \theta_1 = 43^\circ, \theta_2 = 31^\circ\}$						
Initial	5.0	13.6	5.0	4.8	8.9	4.3
Final (constant noise)	0.7	6.6	6.6	4.0	4.7	4.2
Final (increasing noise)	4.0	6.6	5.0	3.4	2.4	4.2
Case 1 vs. arbitrary: Displacement-based design, Image-based recovery						
$P_{\text{true}}=\{C_1 = 4.5, C_4 = C_6 = 9, \kappa = 14.6 \text{ MPa}, \theta_1 = 40^\circ, \theta_2 = 27^\circ\}$						
Design A2 (Table 8.2)	4.4	7.6	8.4	3.0	6.5	8.8
Design A4 (Table 8.2)	5.4	6.4	6.2	2.3	6.0	8.8
Optimal design, case 1 (Fig. 8.3)	4.4	5.9	6.1	1.9	3.8	8.8

Table 8.3: Evaluation of the optimal designs: comparison of the percentage error in parameter inference using the optimal designs versus non-optimal designs.

ments with model inference. In particular, as we gain more knowledge about the material history from experiments (e.g., evolution of yield surface), we can perform follow-up optimal experiments using this updated knowledge in real time. This necessitates a scalable Bayesian OED framework. For strongly nonlinear, history-dependent materials, the adjoint-based gradient methods presented here may become prohibitively expensive, making gradient-free ensemble methods, such as ensemble Kalman inversion or ensemble Kalman filtering, attractive for coupling with the Bayesian optimal experimental design framework here.

Beyond extension to more complex materials, an important consideration is robustness to real experimental conditions. In practice, in any fabrication and experiment, divergence from design to execution is inevitable. This challenge can be addressed in two interesting ways. First, robustness can be incorporated directly into the design problem by optimizing specimens that remain informative under expected manufac-

turing and experimental variability (e.g., printing defects, material heterogeneity). Second, robustness can be handled sequentially, where after each experiment, the actual fabricated geometry, noise, and measured response can be assimilated into the entire model update, and the next design can be chosen based on the accumulated data and observed deviations. Such an integrated closed-loop framework would make the proposed approach more practical for real experiments to capture materials with complex physics, while further reducing the number of tests required for high-fidelity constitutive model discovery in solid mechanics.

Acknowledgement We are delighted to acknowledge useful discussions with Andrew M. Stuart. We gratefully acknowledge the financial support of the Caltech MCE Center on AutoFab and the Office of Naval Research through MURI grant Number N00014-23-1-2654.

Author contributions: CRediT Adeline Wihardja: Conceptualization, Formal analysis, Investigation, Methodology, Software, Validation, Visualization, Writing – original draft, Writing – review and editing. Kaushik Bhattacharya: Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Supervision, Validation, Visualization, Writing – original draft, Writing – review and editing.

APPENDIX TO CHAPTER 8

8.A Derivation for sensitivity and adjoint PDEs

We seek to find the gradient of O_{EIG} with respect to ρ . The details of this gradient calculation to obtain the sensitivity (8.25) and the adjoints (8.26,8.27) are given here. Given any $\lambda, \beta_P \in \mathcal{U}$, we use the forward problems (8.21 and (8.22) to rewrite our objective as

$$O^*(\rho, u, u_P) = -O_{EIG}(\rho, u, u_P) + \int_{\Omega} (\rho^P W_F(\nabla u, P_0)) \cdot \nabla \lambda \, d\Omega \quad (8.34)$$

$$+ \int_{\Omega} (\rho^P (W_{FP}(\nabla u, P_0) + W_{FF}(\nabla u, P_0) \nabla u_P)) \cdot \nabla \beta_P \, d\Omega. \quad (8.35)$$

For the rest of the calculations in this section, we define

$$u_{P_m} := \frac{\partial u}{\partial P_m}, \quad v_{P_m} := \delta u_{P_m}, \quad v := \delta u, \quad (8.36)$$

and define

$$H_{mn} = \int_0^T \left(\int_{\Omega} \rho^{2p} u_{P_m}^T \Gamma_u^{-1} u_{P_n} \, d\Omega + J_m^T \Gamma_f^{-1} J_n \right) dt, \quad (8.37)$$

where m, n denote parameter indices. In general, H is a finite-dimensional matrix depending on the number of material parameters $P \in \mathbb{R}^P$ that are considered, i.e., $H \in \mathbb{R}^{P \times P}$. For the rest of this section, we explicitly denote H as H_{mn} . We follow Einstein summation over the parameter indices. The EIG is

$$O_{EIG} = \frac{1}{2} \log \det(I + C_0 H), \quad H \text{ evaluated with } P_0 \text{ for a given design } \rho. \quad (8.38)$$

Thus,

$$\delta O_{EIG} = \frac{1}{2} [C_0(I + C_0 H)^{-T}]_{mn} \delta H_{mn}. \quad (8.39)$$

and

$$A_{mn} := \left[\frac{1}{2} C_0(I + C_0 H)^{-T} \right]_{mn} = \left[\frac{1}{2} C_{\text{post}} \right]_{mn} \quad (8.40)$$

Now, perturb ρ in the direction μ , such that $\rho \mapsto \rho + \varepsilon \mu$, $u \mapsto u + \varepsilon v$, $u_{P_m} \mapsto u_{P_m} + \varepsilon v_{P_m}$. The full variation of O^* with respect to ρ in the direction μ is given by

$$\langle \delta_{\rho} O^*, \mu \rangle := \left. \frac{d}{d\varepsilon} O^*(\rho + \varepsilon \mu, u + \varepsilon v, u_P + \varepsilon v_P) \right|_{\varepsilon=0}. \quad (8.41)$$

Carrying out the calculation:

$$\begin{aligned}
\langle \delta_\rho \mathcal{O}^*, \mu \rangle = & \\
& - A_{mn} \left(\int_0^t \int_\Omega 2p \rho^{p-1} u_{P,m}^T \Gamma_u^{-1} \rho^p u_{P,n} d\Omega dt \right) \mu \\
& - A_{mn} \int_0^t \int_\Omega \rho^{2p} u_{P_m}^T \Gamma_u^{-1} v_{P_n} + \rho^{2p} v_{P_m}^T \Gamma_u^{-1} u_{P_n} d\Omega dt \\
& - A_{mn} \int_0^t \left(\int_{\partial_u \Omega} ((W_{FF} \nabla v_{P_m}) \hat{n}) \cdot e_f dA \right)^T \Gamma_f^{-1} J_{f_n} + J_{f_m}^T \Gamma_f^{-1} \left(\int_{\partial_u \Omega} ((W_{FF} \nabla v_{P_n}) \hat{n}) \cdot e_f dA \right) dt \\
& - A_{mn} \int_0^t J_{f_m}^T \Gamma_f^{-1} \left(\int_{\partial_u \Omega} ((W_{FFF} \nabla u_{P_n} + W_{FFP_n}) \nabla v) \hat{n} \cdot e_f dA \right) dt \\
& - A_{mn} \int_0^t \left(\int_{\partial_u \Omega} ((W_{FFF} \nabla u_{P_m} + W_{FFP_m}) \nabla v) \hat{n} \cdot e_f dA \right)^T \Gamma_f^{-1} J_{f_n} dt \\
& + \int_\Omega \rho^p W_{FF} \nabla v \cdot \nabla \lambda d\Omega \\
& + \int_\Omega (p \rho^{p-1} W_F \cdot \nabla \lambda) \mu d\Omega \\
& + \int_\Omega p \rho^{p-1} (W_{FP_m} + W_{FF} \nabla u_{P_m}) \cdot \nabla \beta_{P_m} \mu d\Omega \\
& + \int_\Omega \rho^p (W_{FFF} \nabla v \nabla u_{P_m} + W_{FFP_m} \nabla v + W_{FF} \nabla v_{P_m}) \cdot \nabla \beta_{P_m} d\Omega
\end{aligned}$$

Grouping terms with μ , we obtain the sensitivity to be

$$\begin{aligned}
& - A_{mn} \left(\int_0^t \int_\Omega 2p \rho^{p-1} u_{P,m}^T \Gamma_u^{-1} \rho^p u_{P,n} d\Omega dt \right) \mu \\
& + \int_\Omega (p \rho^{p-1} W_F \cdot \nabla \lambda) \mu d\Omega \\
& + \int_\Omega p \rho^{p-1} (W_{FP_m} + W_{FF} \nabla u_{P_m}) \cdot \nabla \beta_{P_m} \mu d\Omega.
\end{aligned} \tag{8.42}$$

Grouping terms of v_{P_m} and setting to zero,

$$\begin{aligned}
0 = & -A_{mn} \int_0^t \int_{\Omega} \rho^{2p} v_{P_m}^T \Gamma_u^{-1} u_{P_n} d\Omega dt \\
& -A_{nm} \int_0^t \int_{\Omega} \rho^{2p} u_{P_n}^T \Gamma_u^{-1} v_{P_m} d\Omega dt \\
& -A_{mn} \int_0^t \left(\int_{\partial_u \Omega} ((W_{FF} \nabla v_{P_m}) \hat{n}) \cdot e_f dA \right)^T \Gamma_f^{-1} J_{f_n} dt \\
& -A_{nm} \int_0^t J_{f_n}^T \Gamma_f^{-1} \left(\int_{\partial_u \Omega} ((W_{FF} \nabla v_{P_m}) \hat{n}) \cdot e_f dA \right) dt \\
& + \int_{\Omega} \rho^p (W_{FF} \nabla v_{P_m}) \cdot \nabla \beta_{P_m} d\Omega \quad \forall v_{P_m} \in \mathcal{U},
\end{aligned} \tag{8.43}$$

furnishes the adjoint equation A1 for β_{P_m} for each parameter m .

Grouping terms of v and setting to zero,

$$\begin{aligned}
0 = & -A_{mn} \int_0^t J_{f_m}^T \Gamma_f^{-1} \left(\int_{\partial_u \Omega} ((W_{FFF} \nabla u_{P_n} + W_{FFP_n}) \nabla v) \hat{n} \cdot e_f dA \right) dt \\
& -A_{mn} \int_0^t \left(\int_{\partial_u \Omega} ((W_{FFF} \nabla u_{P_m} + W_{FFP_m}) \nabla v) \hat{n} \cdot e_f dA \right)^T \Gamma_f^{-1} J_{f_n} dt \\
& + \int_{\Omega} \rho^p W_{FF} \nabla v \cdot \nabla \lambda d\Omega \\
& + \int_{\Omega} \rho^p (W_{FFF} \nabla v \nabla u_{P_m} + W_{FFP_m} \nabla v) \cdot \nabla \beta_{P_m} d\Omega \quad \forall v \in \mathcal{U},
\end{aligned} \tag{8.44}$$

results in the adjoint equation A2 for λ . We note that all the derivatives above, i.e., $W_F, W_{FF}, W_{FFF}, W_{FP}, W_{FFP}$ are all evaluated at $(\nabla u, P_0)$, and density ρ is the proposed design.

CONCLUSION AND FUTURE OUTLOOK

This thesis addressed the challenge of developing high-fidelity constitutive models for complex materials. Such materials often exhibit large deformation, anisotropy, rate dependence, temperature dependence, and coupled and complex physics that are often only partially understood. The work presented in this thesis addressed this challenge from two complementary directions.

The first direction addressed this challenge by following a traditional route in mechanics: designing experiments, identifying important physical mechanisms, and using the resulting experimental observations to develop and validate a physics-based constitutive model. We demonstrated this approach for liquid crystal elastomers (LCEs), a class of soft active materials whose response is governed by the coupling between rubber elasticity, viscosity, liquid crystalline order, and deformation. They are promising for impact absorption structures, though their dynamic response remains largely unexplored. Specifically, there is a gap in experimental characterization in the exact strain rate regime ($10^1\sim 10^3\text{ s}^{-1}$) under large deformation that is of interest to applications. To address this inaccessible regime, in Chapter 3 we developed a novel tensile drop-tower setup that can probe, for the first time, large strains of 100% at dynamic, 10^2 s^{-1} regimes. These experiments provided new insights into the rate-dependent multiphysics coupling between liquid crystalline order evolution and viscoelasticity. However, there was no engineering model that captured this coupling; thus, in Chapter 4, we incorporated these new insights into the previously developed model for LCE while remaining faithful to the underlying microscale physics. The resulting model accurately describes the behavior across all strain rates (static 10^{-2} to dynamic 10^3 s^{-1}). We continued in Chapter 5 to further investigate the coupled effects of temperature and strain rate, providing a broader experimental characterization of the complex behavior of LCEs.

The second direction addressed the broader problem of constitutive model discovery for new, emerging materials. Many contemporary multifunctional and structural materials have physics that are not well understood and thus not amenable to classical constitutive models. While advances in imaging optics have enabled data-rich experiments, established inversion methods still suffer from two major deficiencies.

First, we often use idealized experiments or geometries to overcome the limitations of the inversion analysis, resulting in sparse data since we only probe special states of the material. Second, we use image-based methods to obtain displacements. Unfortunately, the most widely used method (Digital Image Correlation or DIC) is an ill-posed method that requires ad hoc regularization and still leads to a multi-level optimization in obtaining constitutive models (images to displacements, displacements to models). These two limitations lead to large errors in regimes of interest where there are complex states. To overcome these deficiencies, in Chapter 7, we developed the Image to Constitutive Model (I2C) method—an integrated, unified inversion framework to infer constitutive laws directly from raw images and forces. This formulates the problem of finding constitutive relations as an indirect inverse problem that is solved using partial-differential-equation constrained optimization to minimize the difference between the experimental images and computed images. This method bypasses the DIC step and its inherent ill-posedness, and is amenable to any general form of constitutive relations under large deformations, non-pristine test conditions, and complex loadings. We showed that this method can successfully recover the constitutive relation from both synthetic and experimental tests on finite elasticity.

Finally, in Chapter 8, we developed an approach that uses the current model and physics knowledge of the material to design optimal experiments that maximize information gain. This approach reformulates the model discovery problem from a Bayesian perspective. Coupled with topology optimization, it leads to a quantitative framework to design specimen geometries that produce maximally informative data within a single experiment. We showed that using the proposed optimized specimens leads to more accurate model inference in comparison to using conventional specimens.

Through these studies, we have addressed major challenges related to the broader goal of model discovery for emerging materials with complex physics. Together, these contributions create a closed-loop framework between modeling and experimentation for learning and developing the next-generation materials.

9.1 Extensions and Future Outlook

Our contributions presented in this thesis can be extended toward several interesting and exciting future directions. We detail these avenues and discuss general pathways moving forward.

A first direction is the general development of coupled physics models for complex materials that consider thermomechanical coupling, damage, and microstructure evolution. The studies in this thesis show, through LCEs as an example, that strain rate, temperature, microstructural evolution, and dissipation can be strongly coupled. More broadly, many complex engineering materials used in dynamic and impact applications need models that couple mechanical deformation with heat generation and transport, microstructural evolution, and rate-dependent mechanisms. Future avenues involve developing novel experimental setups that provide information on mechanical and thermal behavior by combining mechanical testing with, for example, calorimetry, infrared thermography, diffraction measurements, and other field measurements. In addition, measurements at different scales can also be integrated to inform the development of high-fidelity constitutive models that remain physically faithful.

A second direction is to extend the data-driven frameworks developed here to consider more complex constitutive behavior. Our demonstration focused on hyper-elastic materials, where the material response is only dependent on the current deformation state. Many important engineering materials are history-dependent, where effects like plasticity, viscoelasticity, damage, and rate-dependence exist. In these complex systems, the constitutive response depends not only on the current deformation but also on internal states and loading history, and is thus more challenging to characterize. Developing constitutive laws for elastic-plastic materials, for example, traditionally requires hundreds of mechanical tests on statically determinate specimens to map their yield surface and its evolution. Exploiting full-field measurements with deliberately designed specimen geometries can accelerate and automate the discovery of models for these complex materials by providing richer information within a single experiment. This added complexity opens interesting yet challenging directions to extend the proposed frameworks. I2C can be extended to such materials, requiring inference of both the material parameters and any hidden internal states. This creates a more challenging inverse problem, but it is also where the direct use of raw images and physics-based constraints may be most valuable. The OED framework would be a higher-dimensional design problem, considering not only the specimen topology, but also the loading path and rate to excite and sufficiently explore relevant evolution laws. This complexity also necessitates the development of a fast data assimilation framework that continuously integrates experiments with model inference. Gradient-free and ensemble-based methods can be pursued here.

A third direction of continuation is to couple the inverse I2C framework with physics-based machine learning approaches for constitutive model discovery. For many emerging materials, the appropriate constitutive form may not be known in advance. Highly parametrized machine-learning models would allow raw images to inform unknown constitutive behavior. A complementary direction is model-free discovery, where admissible material response databases are constructed directly from the raw experimental data. These approaches could be especially useful for architected materials, biological materials, and additively manufactured materials, where the effective behavior is governed by complex microstructure and cannot be easily represented by classical constitutive models.

A fourth direction is to extend the inverse framework to three-dimensional measurements and additional experimental data sources. The I2C method was developed around two-dimensional raw images and force measurements, but the same philosophy can be extended to volumetric data and other experimental measurements (temperature fields, diffraction measurements). This would allow the inverse problem to incorporate richer information about the multiphysics and multiscale behavior of complex materials. To enable this extension, developing a unified inversion framework that can combine these measurements is necessary. A central component of this effort is scalability. This also leads to an interesting direction to develop a computationally tractable inversion framework. Processing and integrating full-field data measurements with various modalities is computationally expensive and memory-intensive. One can pursue integration of efficient solvers and high-performance computing for fast image registration and numerical simulations within the inversion framework. This will enable a practical framework that can iteratively guide data collection and accurate model discovery for multiphysics, multiscale materials.

A fifth direction is to consider the realization of the optimal specimen designs within the framework developed in Chapter 8. The numerical results show that topology-optimized specimens can improve model parameter inference. However, realistically, practical experiments introduce additional constraints such as manufacturability, grip design, material defects, thickness variations, and other noise. Future work should incorporate these effects directly into the design problem. One approach is to develop a topology optimization framework that results in robust designs that remain informative under these perturbations. This is a challenging problem, and it is an open problem in the field. In tandem, developing a data

assimilation framework that takes this into account is another exciting avenue.

Lastly, the long-term outlook is a closed-loop framework for experimental mechanics and constitutive modeling. One can use an initial experiment to infer a preliminary model and quantify uncertainty. The current model and uncertainty are then used to design the next experiment. Once an experiment is performed, the new data are assimilated to update the model, and the process repeats. This data-driven, iterative setting requires the development of new experimental campaigns for a variety of complex material systems, as well as the development of scalable and adaptive inversion approaches that leverage a high volume of data.

9.2 Closing remarks

Overall, this thesis integrates the development of constitutive models with experiments and experimental design. By combining current physics knowledge, data-rich measurements, inverse methods, and optimal design, one can move toward a more integrated approach that enables the development of high-fidelity constitutive models for next-generation materials. This work provides a path toward new, alternative approaches in experimental mechanics and materials characterization: pairing data-rich experiments and novel data-driven inversion techniques that extract models directly and automatically from data. Although the specific methods presented here are still in an early stage, I hope that they can motivate the development of new integrated experimental, computational, and theoretical approaches in mechanics.