Research Article

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Mapping linear viscoelasticity for design and tactile intuition

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Abstract: We propose and study methods to improve tactile intuition for linear viscoelastic fluids. This includes (i) Pipkin mapping with amplitude based on stress rather than strain or strain-rate to map perception to rheological test conditions; and (ii) data reduction of linear viscoelastic functions to generate multi-dimensional Ashby-style cross-property plots. Two model materials are used, specifically chosen to be easily accessible and safe to handle, with variable elastic, viscous, and relaxation time distributions. First, a commercially available polymer melt known as physical therapy putty, reminiscent of Silly Putty, designed for a range of user experiences (extra-soft to extra-firm). Second, a transiently cross-linked aqueous polymer solution (Polyvinyl alcohol-Sodium Tetraborate, PVA-Borax). Readers are encouraged to procure or produce the samples themselves to build intuition. The methods studied here reduce the complexity of the function-valued viscoelastic data, identifying what key features we sense and see when handling these materials, and provide a framework for tactile intuition, material selection, and material design for linear viscoelastic fluids generally.

Keywords: Viscoelasticity, Psychorheology, Ashby Diagram

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1 Introduction

Intuition with viscoelastic materials is central to integrating them into the design toolbox. Engineering design with elastic solids and Newtonian fluids is well established, as

demonstrated through extensive cross-property Ashby Diagrams [1–3] and materials databases [4, 5]. This is facilitated by the simplicity of describing these materials by single value material properties (such as viscosity for fluids and modulus or hardness for solids). Materials experts and non-experts alike have developed intuition of the value of these properties for common materials such as those shown in Figure 1.

Soft viscoelastic materials, those between the bounds of pure Hookean solids and Newtonian fluids, have mechanical/rheological properties that are more complex which likely could meet a wide variety of design functionality; biology demonstrates this [9–11] as do engineering applications such as soft robotics [12–21], polymer processing [22–25], sporting goods [26], automotive [27], soft armor [28], 3D printing [29–32], and injectable hydrogels [33–37].

Yet, unique challenges still inhibit the use of rheologically-complex materials in engineering design. First, even simple viscoelastic material properties are functions, not single values. Thus, even at a basic level, design with these materials is a high-dimensional problem (e.g. cross-property Ashby Diagrams would require either more dimensions or lower-dimensional descriptions [12, 32, 38–40]). Second, while design intuition with solid and fluid materials is well developed, the same intuition with viscoelastic materials is lacking. Take for example the viscoelastic polysilicone putties in Figure 2. The qualitative description of these samples has not yet been associated with specific viscoelastic rheological parameters.

The putties in Figure 2 are marketed on a range from extra-soft to extra-firm. However, the viscoelastic character of the putties implies that the materials have complex, function-valued properties. What rheology correlates to perception? Are there simplified material descriptions that can be used to develop the intuition needed to *design* with viscoelastic liquids?

We are motivated by *design* which is built upon *analysis* (Figure 3). Currently, most work in the soft matter/rheology literature is *analysis* of existing or new materials, proceeding from left to right in Figure 3. True *design* with rheological properties is an inverse problem shown from right to left in Figure 3. Starting from the required

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Figure 1: Simple solids and Newtonian fluids can be easily described by single-valued material properties, such as a Young's modulus or a Newtonian viscosity [6]. This enables easy intuition when designing with these materials as well as easy communication of the material properties. Figure adapted from [6], [7] and [8].

Changing viscoelasticity: relaxation timescale? elasticity? viscosity?

Figure 2: Polysilicone materials designed for a range of user experience from “extra-soft” (left) to “extra-firm” (right). But this naming convention from the supplier does not clarify what viscoelastic features change across the spectrum of materials: elasticity, or viscosity, or relaxation timescales? (Photos taken 10 minutes after initial spherical state.)

performance, the enabling properties are then identified (e.g. initially agnostic to any material structure [41]); rheological properties can be achieved by multiple microstructures (e.g. the various structures that achieve a yield stress fluid [32, 39]) and the design trajectory iterates by choosing/refining a particular material structure [42]. This design trajectory is shown from right to left in Figure 3.

Here we propose materials for tactile intuition of linear viscoelasticity, study their rheological properties, identify low-dimensional descriptions, and demonstrate a frame-

work for Ashby-style plots of linear viscoelasticity. The materials we propose (detailed in Sec. 3) are readily available, easy to make, non-toxic, and non-hazardous.

With these materials, we will probe the question *When you feel a material, can you draw its material functions?* or its inverse, *If you see the rheological material functions, can you intuitively know how the material will feel?* The link between subjective assessment of the rheological behavior of the material (i.e. user experience) and objective data from rheological tests is known as psychorheology[44]

Figure 3: Engineering design is an inverse process with multiple options to meet an objective. Function-valued rheological properties are at the center of this structure-properties-performance design paradigm (adapted from [43]). Our work here aims to improve design intuition for linear viscoelastic materials by simplifying property representations and improving the mapping of properties to performance.

which combines introspective psychology, sensory evaluation, physics, and rheology [45]. Beyond rheological data, the field is generalized to psychophysics. Psychophysics and psychorheology have a strong basis in the literature, particularly in their applications to the food and cosmetics industries [44–51]. In this work we lay out a framework to change nebulous psychorheological characteristics of viscoelastic materials, such as ‘soft’ or ‘firm’, into quantitative links to material properties, aiding the use of viscoelastic materials in design.

Building intuition helps engineers and rheologists alike. The ideas presented in this work have been used in multiple short courses on rheology to help scientists and engineers build intuition for viscoelastic properties. More details are provided in Appendix A.¹

¹ The polysilicone putty results here have been included as a hands-on lesson module for building intuition with linear viscoelasticity for rheology short courses including: Houston, TX (July 2015), Buenos Aires, Argentina (April 2016), Minneapolis, MN (June 2016, August 2018), Istanbul, Turkey (April 2017), Irvine, CA (May 2017), Stanford, CA (June 2019), and at KU Leuven, Belgium (September 2017, September 2019). Figures as they appear in the module can be found in Appendix A.

2 Background

2.1 Design

2.1.1 Engineering design and the role of intuition

The design science literature makes it clear that to improve design, designers need to understand and have experience with these systems [52–55]. Herein lies the problem for design with rheologically-complex materials: rheology is not a topic widely understood by those without graduate-level experience in the field.

Like design, education is also reliant on multiple layers of information and understanding. It is widely understood that effective learning and understanding requires a variety of experiences and teaching styles: visual, auditory, and *tactile* [56].

A large task within this work and future design work with rheologically-complex material is to develop multiple levels of experience, particularly tactile experience, with these materials to aid in education and give designers the necessary subject matter expertise.

2.1.2 Materials Selection for Design

Single-valued properties are those for which a single data point can describe the flow behavior of a material, e.g. the viscosity of water at room temperature (in contrast to the flow curve of mayonnaise) or the modulus of rubber (in contrast to the full shape of decaying relaxation modulus of molten plastic). Single-valued material descriptions allow for the creation of cross-property plots for intuition and material selection, such as Figure 1, or the widely used Ashby-style coplots [1–3] for Material Selection for Design. Ashby presents cross plots of a number of simple material properties (e.g. Young's modulus, density, cost) for thousands of commonly used materials [57]. Ashby diagrams give designers and engineers guidance for selecting materials that need to achieve a specified performance and thus have specific material properties.

With rheologically complex materials, properties are inherently function valued. For some types of rheological complexity, there has been an effort to distill this complex behavior into single characteristic quantities such as yield-stress fluids (yield stress and post-yielding viscosity) [38] or food products (springiness) [58], to develop tools to guide materials selection similar to Ashby [1]. However, viscoelastic behavior presents an even bigger challenge.

2.2 Continuous relaxation spectra and viscoelastic constants

The concept of a continuous distribution of relaxation timescales [59, 60] will be used as a means to describe the function-valued linear viscoelastic observations here and facilitate data reduction to lower-order descriptions. It is convenient to develop the definition of a continuous relaxation spectrum by first understanding a discrete relaxation spectrum [60]. The viscosity-weighted discrete relaxation spectrum is defined as (notation following that used by Martinetti *et al.* in [61]).

$$\hat{H}(\tau) = \sum_{i=1}^N \eta_i \cdot \delta(\tau - \tau_i). \quad (1)$$

where η_i is the viscous strength of the relaxation mode at timescale τ_i . This definition can be visualized as a superposition of N Maxwell models combined in parallel. At the limit $N \rightarrow \infty$, this becomes the definition of the viscosity-weighted continuous relaxation spectrum

$$H(\tau) = \lim_{N \rightarrow \infty} \sum_{i=1}^N \eta_i \cdot \delta(\tau - \tau_i). \quad (2)$$

This viscosity-density spectrum $H(\tau)$ has dimensions of Pa·s/s. The spectrum can likewise be defined as a modulus-weighted spectrum

$$Q(\tau) = \lim_{N \rightarrow \infty} \sum_{i=1}^N G_i \cdot \delta(\tau - \tau_i) \equiv \frac{H(\tau)}{\tau} \quad (3)$$

Importantly, low-dimensional viscoelastic constants that describe viscoelastic qualities (elasticity, viscosity, compliance) can be calculated directly from the integral moments of the modulus-weighted spectrum $Q(\tau)$ and their equivalence with the viscosity-weighted spectrum $H(\tau)$ [60]

$$M_0 = \int_0^\infty Q(\tau) d\tau = \int_0^\infty \frac{H(\tau)}{\tau} d\tau = G_0 \quad (4)$$

$$M_1 = \int_0^\infty Q(\tau) \tau d\tau = \int_0^\infty H(\tau) d\tau = \eta_0 \quad (5)$$

$$M_2 = \int_0^\infty Q(\tau) \tau^2 d\tau = \int_0^\infty H(\tau) \tau d\tau = J_0 \eta_0^2. \quad (6)$$

From the zeroth, first, and second moments and their associated viscoelastic constants we calculate further low-dimensional descriptions of the characteristic relaxation times of the material,

$$\tau_1 = \frac{M_1}{M_0} = \frac{\eta_0}{G_0} \quad (7)$$

$$\tau_2 = \frac{M_2}{M_1} = J_0 \eta_0. \quad (8)$$

These timescales are referred to in literature [60] as “number average” $\tau_n = \tau_1$ (the mean relaxation time of the modulus weighted spectrum $Q(\tau)$) and “weight average” $\tau_w = \tau_2$ (the mean relaxation time of the viscosity weighted spectrum $H(\tau)$) respectively but here we prefer the numerical subscript notation which better corresponds to the different moment definitions given in Eqs. 4–6. We use the timescales of Eqs. 7–8 to define the polydispersity index

$$PDI = \frac{\tau_2}{\tau_1}. \quad (9)$$

3 Materials and Methods

3.1 Materials

Materials for this study were chosen for their range of viscoelastic behavior and ease of access (one is commercially

Figure 4: Aqueous polyvinyl alcohol, transiently crosslinked, displays a wide range of elastic modulus but a comparatively narrow range of relaxation timescales. From left to right, concentrations (PVA wt%:Borax wt%) are: 3.50:1.25, 2.75:1.25, 3.50:0.10, 2.75:0.10, 2.00:0.50. (Photos taken 10 minutes after initial spherical state.)

available, while the other is easily formulated in the lab). While the materials fall within different classes (polymer melt and transiently cross-linked aqueous polymer solution), both materials show approximately Maxwellian linear viscoelastic behavior including a dominant relaxation timescale and fluid-like terminal regime behavior within the range of human perception.

3.1.1 Polysilicone Physical Therapy Putties

This study will first describe six different types of putty manufactured by Depco, Incorporated sold under the name Thera-Flex Brand TheraPutty which were ordered through Isokinetics, Inc. (at the time of writing, available through the retailer Amazon.com). These materials are used for physical therapy, *e.g.* hand strengthening after an injury. Per manufacturer specification, the putties range from “extra-soft” to “extra-firm”. The six putties tested, shown in Figure 2, were extra-soft (tan), soft (yellow), medium (pink), medium-firm (red), firm (green), and extra-firm (blue). Ingredients of each putty are not specified by the manufacturer, but similar materials are known to be made of dimethyl silicone polymer [62–64]. The viscoelasticity of the putties is therefore likely due to polymer entanglement in addition to transient cross-linking of the silicone polymer with the use of a cross-linking compound such as boric acid and additional filler materials [65–68].

3.1.2 PVA-Borax

Aqueous solutions of Poly(vinyl alcohol) (PVA) (Aldrich Chemical Co., molecular weight = 85,000 - 124,000, 99+% hydrolyzed) were mixed with aqueous solutions of Sodium Tetraborate (Borax) (Aldrich Chemical Co.) to form multiple transient polymer networks with ion-assisted thermoreversible crosslinks [69]. The mixture was prepared as follows. PVA was dissolved in deionized water under continuous stirring at a temperature of about 95°C for approximately four hours to form a 4wt% stock solution. Borax was dissolved under similar conditions to form a 4wt%

stock solution. The two solutions were allowed to cool to room temperature and were mixed together in different ratios to form multiple compositions. Mixing was done by combining measured weights of each component in a closed test tube and shaking them until the components were mixed thoroughly. The gel was then centrifuged at 3000 rpm for 10 minutes to remove any air bubbles (CL2 Centrifuge, Thermo Scientific). The resulting mixture was then used for rheometry tests.

3.2 Rheometry

Stress-controlled creep and oscillatory shear measurements on the viscoelastic therapy putties were performed on a single-head, combined motor-transducer rotational rheometer (Discovery Series Hybrid Rheometer (DHR), model HR-3, TA Instruments). Stress-relaxation measurements were performed on a strain-controlled separated motor-transducer rotational rheometer (ARES-G2, TA Instruments). The instrument was used with a 20 mm diameter flat plate geometry at an operating gap of 800 μm . The temperature for the tests was set at 25°C and controlled using a Peltier bottom plate.

Oscillatory stress sweep tests and creep tests were performed to ensure that data collected was within the linear regime. For each test, a sample was rolled into a ball and placed onto the bottom plate and the geometry was lowered onto it. Excess sample was trimmed at a gap of 850 μm before the final lowering of the geometry.

Three repeat tests were performed to judge experimental uncertainty due to loading and trimming the sample, as well as sample relaxation within the rheometer geometry. To ensure that the samples had relaxed sufficiently prior to testing, each putty was allowed to relax for a specified duration. These tests ensured that the total error due to the loading procedure and subsequent relaxation was less than or equal to five percent. Multiple gap heights were also tested to check for the presence of slip with the materials. No significant effect was found.

Measurements on the PVA-Borax systems were performed on the same combined motor-transducer rota-

tional rheometer using a plate-plate geometry (40mm, sandblasted). Dynamic linear viscoelastic measurements were performed using an oscillation frequency sweep (small amplitude oscillatory shear) at 1% strain amplitude at 25°C maintained by a Peltier system in the lower plate. The strain amplitude was chosen from a forward and reverse oscillatory strain amplitude sweep at 1 rad/s to identify the linear viscoelastic regime. For each test, the sample was poured onto the bottom plate to form a round puddle and the upper geometry was lowered onto it. After lowering the geometry, excess sample was trimmed to match the upper plate diameter. The sample was allowed to relax for approximately three minutes prior to taking measurements. A solvent trap was used during testing to prevent evaporation losses.

4 Pipkin Mapping of Perception-to-Rheology

When discussing tactile perception and how people interact with materials, it seems natural to speak about stress inputs. There is precedence in psychorheological literature that human inputs for understanding material qualities (such as softness/firmness) correlate with stress-controlled experiments [44, 48, 70, 71]. This contrasts with how rheologically-complex materials are often described with material functions based on strains and strain-rates being the controlled input (which is convenient for understanding/solving some constitutive models, *e.g.* ordered fluid expansions where stress is an explicit function of multiple terms involving strain rate [22, 72]).

4.1 Stress-Amplitude Pipkin Space

The two-dimensional operational space of Pipkin, of amplitude (commonly strain or strain rate) and transient timescale, is exceedingly useful in mapping out nonlinear viscoelastic constitutive models and characterization [69, 73, 74]. Traditionally, the space is presented as a function of two non-dimensional representations of input: the Deborah number ($De = \frac{\tau}{t} = \tau\omega$) for viscoelasticity and the Weissenberg number ($Wi = \tau\dot{\gamma}$) for nonlinearity. These terms allow for a description of the relevant timescale and the strength of the deformation. The Pipkin space can be generalized for dimensional inputs of timescale t and amplitude $\dot{\gamma}$ [75, 76] (allowing for arbitrary or various τ to be considered), or extended to extensional deformations. However, the use of *stress* amplitude for the or-

dinate is much less common being used only for stress-controlled LAOS response mapping [77] and recently described in [74].

Here, we introduce a stress-amplitude Pipkin space to use as an operational map to connect human perception to rheological tests, shown in Figure 5. In the stress-amplitude Pipkin space of Figure 5, the strength of the flow is described simply as the stress amplitude in the place of the traditional strain or strain-rate (Weissenberg number).

The stress-amplitude perspective will relate more naturally to tactile perception intuition (squeezing, pulling, poking). After mapping human tactile perception, we will use this operation map to guide rheological testing that is likely to be relevant to user experience with the materials.

4.2 Perception Limits

The conditions for hand-held tactile and visual perception can be mapped to a Pipkin space, as shown in Figure 5. The colored regions show the bounds of relevant stresses and timescales that we have estimated for human handling. Conditions for rheological characterization are also shown, as trajectories through the Pipkin space [74]. A similar methodology could be followed for other user-experience problems, *e.g.* mouthfeel, palpation, ergonomic hand grip material, etc. where different ranges might be relevant. Table 1 documents the range limits identified here for mapping tactile and visual perception of viscoelastic liquids such as the therapy putties and PVA-Borax, which are described in more detail as follows.

Visual perception, shown in orange in Figure 5, is particularly important for viscoelastic materials which can visually appear solid over short times and flow over long times. Here, the visual perception is bound at short timescales by approximate ranges of human perception. We use 13 ms in Figure 5, the minimum viewing time for visual comprehension [84]. The minimum bound could be set smaller with reasonable assumptions, *e.g.* for the case of bouncing viscoelastic putty the bounce impact time is 4 ms [12], and the consequences are visually apparent. Or, if we consider shooting Silly Putty with a bullet [85] this imposes even shorter transient timescales with visually observable consequences, but this is outside the scope here. The longest timescales for visualization are set by the patience of human observation, which we show as unbounded. Different limits could be applied, *e.g.* the time lapse in Figure 2 was stopped at 10 min, and the famous pitch drop experiment has been active since 1930 [86].

The stress-axis limits for visual perception are set by any environmental conditions that can be visually

Figure 5: Pipkin diagram [73] adapted to map perception from Table 1 using stress for the amplitude (rather than strain or strain-rate). Stress-amplitude σ_0 is a more natural mapping for user-experience. Regions relevant to human perception guide rheological characterization, shown as locations or trajectories through the Pipkin space.

Table 1: Perception limits relevant to human handling of putties and gels. See text for details.

	Min & Max	Comments
Stress (tactile)	200 Pa 50 kPa	Light touch [78, 79] Hand grip strength [80, 81]
Time (tactile)	200 ms 10 min	Neuro-muscular max frequency [82, 83] Patience while squeezing
Time (visual)	13 ms -	Visual comprehension [84] Patience while observing

observed. Example scenarios include the gravitational stresses in Figure 2, the hand-squeezing stresses suggested in the photo of Figure 5, impact stress when dropping balls of putty from a height of tens of centimeters [12] or dropping a heavy ball of putty from the top of a building² [87], or shooting Silly Putty with a bullet [85]. For visual perception related to hand-held intuition, here we have taken a simple approach of arbitrarily setting the visual perception stress range slightly outside the bounds of the tactile perception limits, which are detailed below.

² available videos include <https://www.youtube.com/watch?v=4kxUXAWkUmM> and <https://www.youtube.com/watch?v=2Zez3v0fGi8>

Tactile perception, shown in blue in Figure 5, is more limited than visual perception since the stresses and timescales are imposed by human hands. Long timescales are chosen as estimates relevant to handling the materials, e.g. during physical therapy, on the order of a few minutes. The shortest possible timescale can be estimated in two ways. One is based on the highest frequencies that can be perceived by passively resting human hands, as studied in the haptic literature [88, 89]. This gives transient timescales of 1-10 ms. However, this is a lower bound estimate and not relevant to handling viscoelastic materials because we expect that the hands must actively impose the deformation, rather than passively perceive it. A better lower bound timescale estimate comes from neuro-muscular maximum frequency of motion. A well-documented and widely used test of motor function (and impairment) is the “Finger Tapping Test” [82]. It measures the maximum rate at which a person can tap their finger. For healthy adults, the “intertap interval” time is generally on the order of 150-220 ms, e.g. Hubel *et al.* [83] report timescales on the order of 200 ms for a broad range of adult participants. We interpret this 200 ms as a minimum characteristic transient time of fingers (and hands) to interact with the viscoelastic putties, e.g. hands or fingers attempting to quickly deform the material to observe the viscoelastic relaxation. Based on this estimate, viscoelastic relaxation times would need to be larger than 200 ms to be perceived in this type of scenario.

Tactile stress amplitude limits are calculated from relevant literature on grip strength measured with a Jamar dynamometer [80] and haptic perception limits [78, 79]. This gives a minimum stress on the order of 200 Pa, based on light touch perception, and a maximum stress around 50 kPa, based on hand grip strength. Stresses could potentially be larger by poking the materials with a finger, around 460 kPa [90]. It is important to note that the numbers and limits of these regions cannot be exact but these ranges can act as an approximate guide to the relevant conditions of use, and conditions relevant for rheometric characterization.

4.3 Co-location of rheological characterization

Our Pipkin map allows for the co-location of simple rheological tests corresponding to the regions relevant to user experience. Stress-input rheological tests such as creep (step-stress), oscillatory tests (fixed stress amplitude, or fixed frequency) can be mapped over the regions of relevance to insure that rheological characterization best corresponds to how a material will be processed. We recommend this co-location mapping as a general technique for rationalizing rheometric test conditions.

For designers, these regions can guide in the selection (or design) of viscoelastic materials given rheological property targets. Conditions outside of these regions are hypothesized to be unimportant to the behavior and performance within the region of interest. In-use conditions should define property targets.

In Figure 5, the operational map does not include any material-specific limits, such as the onset of nonlinear behavior at a critical stress, or the longest relaxation time. In that sense, the current Pipkin space is material agnostic and application-specific. If a material has already been selected, then these limits (or normalizations to make dimensionless groups) could be added to further describe the space and the anticipated material response.

5 Experimental Results

5.1 Rheology of Therapy Putties

5.1.1 Step-input Experiments

Step-stress shear rheometry (creep tests) may closely correlate with the use of the physical therapy putties. In use,

Figure 6: Creep compliance $J(t, \sigma_0)$ from experiments, analogous to the Figure 2 time-lapse conditions subjected to gravitational forcing, at increasing stress amplitude. $\sigma_0 = 10^2, 10^3, 10^4$ Pa for each polysilicone putty. For the softest putty, only the lowest stress amplitude is in the linear region. Experimental limits for maximum torque prevent some higher amplitudes for the two firmest putties.

a reasonably constant stress may be applied for a few seconds or more. The step-stress experiment moves through the Pipkin map (Figure 5) as a horizontal line trajectory from right-to-left as time passes.

Figure 6 shows the creep compliance $J(t; \sigma_0)$ for various input stress amplitudes ($\sigma_0 = 10^2, 10^3, 10^4$ Pa). These are within the perception bounds outlined on the Pipkin space (Figure 5 and Table 1, maximum tactile stress $5 \cdot 10^4$ Pa) implying that they may reveal information about how the material feels to the user.

Only the softest two putties show nonlinear behavior at stresses accessible to the instrument used. For that reason, as well as other experimental issues (see Supplementary Information) relating to the stiffness of the putty, no comparative conclusions can be drawn from the nonlinear step-input data.

As shown in Figure 6, 100 Pa is the only stress for which all six putties remain linear. This sets the bounds of the linear viscoelastic regime for further testing. The linear creep-compliance, $J(t)$, and creep-fluidity, $\phi(t) = \dot{\gamma}(t)/\sigma_0$, are both shown in Figure 7. At long times, all materials show flow behaviors of a viscoelastic fluid. The largest dif-

Figure 7: Linear step-stress data from Figure 6 (for $\sigma_0 = 100$ Pa). Compared on log-scales as (a) creep compliance $J(t) \equiv \frac{\gamma(t)}{\sigma_0}$ and (b) creep fluidity $\phi(t) \equiv \frac{\dot{\gamma}(t)}{\sigma_0}$. The curves show significantly more flow at long time for the softest putties. There is very little difference observable in the creep response for the firmest four putties. (Gray area indicates region of significant system inertia [91].)

ferences are seen in the extra-soft and soft putties. There are only very slight differences among the firmest four putties, particularly when looking at the creep-compliance. Clearly, the step-stress material response is not enough to distinguish the differences in all the materials.

Step-strain (stress relaxation) experiments were also performed on some of the putties. These results are shown in Appendix A Figure A1 comparing the green (firm) and yellow (soft) putties, and in Supplementary Information for the red (medium) putty. Differences are apparent. The relaxation timescale appears to be shorter for the softer putty in this representation. The flow viscosity is not as apparent in this representation of $G(t)$ compared to the creep tests, e.g. specifically comparing green and yellow putties

in Figure A2. A short time (rubbery) plateau modulus is not observable in the $G(t)$ data due to the minimum step time of the instrument. This is resolved by considering oscillatory characterization.

5.1.2 Oscillatory Experiments

Oscillatory experiments can provide higher signal-to-noise ratios compared to step tests, and can also span the Pipkin perception space where frequency corresponds to inverse transient time. Long times are measured at low frequency and short times are measured at high frequencies. Just as in the step-stress experiments conducted, it is important to choose experimental parameters appropriate and significant for human perception. Guided by the Pipkin space in Figure 5 (Table 1) and the results of the creep tests described in Sec. 5.1.1, a stress amplitude of 100 Pa is chosen. This stress is shown to be within the linear viscoelastic limit for these materials, thus all linear viscoelastic descriptions (moduli, compliances, viscosities, fluidities, etc.) can be used interchangeably [59].

For reference, non-linear material behavior is shown in Supplementary Information. Over the range of frequencies used, all materials are within the linear viscoelastic limit.

A frequency-sweep with conventional strain-input perspective (G' , G'') is shown in Figure 8a for the six therapy putties, isolated individually in Supplementary Information. The stress-input perspective of dynamic compliance J' and fluidity ϕ' is shown in Figure 9, and also isolated for individual samples in Supplementary Information. Each putty can be distinguished with these oscillatory material functions due to the high signal-to-noise ratios and availability of sub-dominant components.

The response of each putty appears to have a rubbery plateau modulus $G' \rightarrow G_0$ at high frequency (Figure 8b) and a nearly terminal regime at low frequency, resulting in a low-frequency plateau dynamic viscosity $\eta' \rightarrow \eta_0$ (Figure 8c). In this sense, the responses all look similar to the familiar Maxwell model. However, a single-mode fit is not completely satisfactory, see Figures 13 and A4 for examples with the green and yellow putties.

Another check of how well a single-mode description is satisfactory comes from the stress-input material functions signatures (J' , ϕ'), which would be constant for a single-mode Maxwell model. Figure 9 reveals that the (J' , ϕ') signatures are nearly constant, as detailed with the green putty in Figure 9b,c. Thus, the (J' , ϕ') material functions act as a type of data reduction compared to (G' , G''), since they are approximately constant for each putty.

Figure 8: Linear viscoelastic frequency sweeps for the physical therapy putties; input stress amplitude 100 Pa. Uncertainty bars are associated with multiple trials; repeatability falls within the symbol size for all tests. (a) Viscoelastic moduli G' (closed) and G'' (open) for all samples. (b,c) the “firm” putty data emphasizing the low dimensional quantities G_0 , η_0 , and τ_1 as calculated from the moment of the continuous spectrum shown in an inset to (b). The instrument inertia (dashed line), as defined in [91].

Figure 9: Stress-input paradigm of linear viscoelastic functions. Elastic (J' , closed) and viscous (ϕ' , open) measures for the therapy putties in the stress-input paradigm. Both measures show a plateau (low-frequency for ϕ' , high-frequency for J') when viewed from the stress-input perspective. The elastic plateau converges to the same value for all putties. Right: stress-input measurements for the “firm” viscoelastic therapy putty. Low-dimensional viscoelastic descriptions for the elastic (J_0) and viscous (ϕ_0) measures are easily found as the data plateaus over multiple decades of input frequencies in this stress-input paradigm.

5.2 Rheology of PVA-Borax

Similar oscillatory data for the PVA-Borax material system is used to provide context and comparison with the putty results. Oscillatory linear viscoelastic data is shown in Figure 10 for ten concentrations of PVA-Borax mixture, courtesy of Olivia Carey-De La Torre [92–94]. All PVA-Borax samples also approach a high frequency elastic modulus plateau $G' \rightarrow G_0$ and a low frequency dynamic viscosity plateau $\eta' \rightarrow \eta_0$. The plateau moduli are all much lower than the putties and vary considerably among the samples. We will see that all PVA-Borax samples have a dominant relaxation time within a small range of each other. Similar data reduction challenges occur for these samples, which we address below.

6 Data reduction for intuition and Ashby-style plots

The data reduction from plateaus is useful for understanding what is different between the therapy putties. But, how should apparent plateaus be quantitatively identified, what timescales can be defined, and what about the polydispersity of timescales when materials are not precisely single-mode Maxwell?

We address these questions with a data reduction approach based on relaxation spectra $H(\tau)$ and moments of its distribution, as defined in Eqs. 4–9. These moments provide measures of viscosity η_0 , elastic modulus G_0 , relaxation timescales τ_1 and τ_2 , and the polydispersity index PDI associated with the range of probed frequencies. These measures will clarify and quantify the differences between the putties, identify which differences might actually be perceived when handling them, and provide multi-dimensional data for making Ashby-style plots of the viscoelastic properties.

Although spectrum calculations can yield non-unique solutions [95–101], the moments of the resulting distribution are well-defined and we compute them from only the Davies-Anderssen range of timescales [102] $\tau_{\min} < \tau < \tau_{\max}$, related to the minimum and maximum experimental frequencies as $\tau_{\min} = e^{2\pi}/\omega_{\max}$ and $\tau_{\max} = e^{-2\pi}/\omega_{\min}$. Importantly, the spectrum description allows for polydisperse relaxation timescales without assumption of an underlying model form or form of the data. This contrasts other possible approaches. For example, we do not force a single-mode Maxwell fit (since this may not be satisfactory), nor do we force a parameterized shape of the spectrum (since different materials may have different shapes

Figure 10: Linear viscoelastic moduli (G' , η') for a selection of concentrations of PVA-Borax systems show Maxwell-like behavior of the system. Concentration of PVA (2, 2.75, 3 wt%) is indicated by color intensity (low to high). Concentration of Borax (0.05, 0.1, 0.5, 1.25, 1.5) is indicated by symbol size. Data collected by Olivia Carey-De La Torre [92–94]

of the spectra), nor do we fit only the plateaus of the frequency-dependent data (since plateaus may not always be achieved or apparent within the range of available data).

A spectrum of relaxation times can be found from experimental G' , G'' data with many different techniques. Here, a high-density discrete approximation to the continuous relaxation spectrum $H(\tau)$ is calculated using the TRIOS software from TA Instruments. A representative example is shown for a single putty in the inset of Figure 8b. All spectra and their associated viscoelastic moduli are shown in Supplementary Information.

Integral moment calculations of $H(\tau)$, Eqs. 4–6, are truncated to include only the window of timescales τ that

Table 2: Low-dimensional linear viscoelastic descriptions of the physical therapy putties, as calculated from the moments of the relaxation spectrum.

Material	η_0 [Pa·s]	G_0 [Pa]	τ_1 [s]	τ_2 [s]	PDI
Extra-soft	$0.15 \cdot 10^5$	$0.43 \cdot 10^6$	0.036	0.092	2.55
Soft	$0.58 \cdot 10^5$	$0.79 \cdot 10^6$	0.073	0.125	1.71
Medium	$1.18 \cdot 10^5$	$0.92 \cdot 10^6$	0.128	0.221	1.73
Medium-firm	$1.20 \cdot 10^5$	$0.91 \cdot 10^6$	0.132	0.228	1.73
Firm	$1.51 \cdot 10^5$	$1.11 \cdot 10^6$	0.136	0.238	1.75
Extra-firm	$1.49 \cdot 10^5$	$0.92 \cdot 10^6$	0.162	0.306	1.89

can be confidently determined from the range of frequencies probed. This is based on the analysis of Davies & Anderssen [102], $e^{2\pi}/\omega_{\max} < \tau < e^{-2\pi}/\omega_{\min}$. These bounds of τ used for integration are indicated by vertical lines in Figure 8b (inset) and for all the spectrum plots in Supplementary Information. This truncated window eliminates the tails where the spectra show the most variability between repeat tests. Inclusion of these tails can have a significant contribution to some of the moment calculations. Omitting these tails results in more consistent moments from repeat tests, though the truncation loses some information. For example, the reconstructed G' , G'' from the truncated $H(\tau)$ are lower than the experimental data (especially the sub-dominant components) in the regions of high and low frequency. The plateau values are generally recovered very well from the moments of the truncated spectra, e.g. in Figure 8b,c. The extra-soft putty shows the most noticeable loss of information with a G_0 that is slightly lower than the experimental data at the highest measured frequencies. However, no other sample suffers from any significant issue.

Table 2 summarizes the moment calculations for the therapy putties in terms of low-dimensional viscoelastic properties. Of the five measures shown for each putty, only three are independent, e.g. $\tau_1 = \eta_0/G_0$ and $PDI = \tau_2/\tau_1$.

6.1 Viscosity, Modulus, Timescale

Figure 11 compares the putties in terms of η_0 , G_0 , and τ_1 . From this and Table 2 it is clear that the difference felt between putties is primarily the viscosity η_0 , not the elastic modulus G_0 or relaxation timescale τ_1 . The viscosity η_0 spans an order of magnitude, showing a clear differentiation between the therapy putties. There is also separation in the relaxation timescales, but it is important to refer back to the Pipkin space in Figure 5 and see that the timescales for the materials (36–162 ms) are below the range of human perception (around 200 ms) thus it is un-

Figure 11: The difference felt between putties is primarily the viscosity η_0 , not the elastic modulus G_0 or relaxation timescale τ_1 . The low dimensional descriptions are calculated from moments of the relaxation spectra. Only a small difference in elastic nature is observed while an order of magnitude change is observed across the viscous description. There is significant difference in the relaxation timescale, however these timescales are well below the estimated limits of active perception outlined in Table 1.

likely that the relaxation timescale differences are important to a user's experience with the materials. The elastic modulus changes slightly among the putties, but the change is not dramatic and all putties have shear elastic modulus G_0 on the order of 1 MPa. Moreover, this modulus is only relevant at transient timescales shorter than the relaxation timescale, which is below our estimate of human tactile perception.

It may seem odd that viscosity differentiates the range of putties that are marketed as “soft” to “firm”, since these descriptive words are more likely to evoke concepts of elastic rather than viscous properties. However, the timelapse images of Figure 2, and the squeezing images of Figure 5, show flow on timescales much longer than any measured relaxation time, and the increase in flow for the “softer” putties is consistent with the idea of viscosity determining the response.

6.2 Relaxation timescale dispersity

The low-dimensional descriptions in Figure 11 naturally omit information about the full material response. They imply a material with a single dominant relaxation time. Simple checks for Maxwellian-type material behavior include Cole-Cole plots [103–106] (see Supplementary Information) as well as checking for long, well-defined plateaus in the stress-input material functions J' and ϕ' (Figure 9). A more quantitative approach is to report the timescale polydispersity index PDI , Eq. 9, i.e. the ratio of average relaxation timescales based on different moments of the relaxation spectrum. $PDI = 1$ represents a perfect single-mode Maxwell response and sets the lower bound of possible PDI values. In a logarithmic scale, the PDI can be viewed as the distance between the timescales τ_2 and τ_1 .

Figure 12 shows the PDI for all therapy putties and PVA-Borax samples. The majority of samples examined here have dispersities near 1.7. The exception is the extra-soft therapy putty with a larger PDI of 2.5. For reference, the same analysis applied to digitized data for a wormlike micelle system described in [107] is also included, with a $PDI = 1.1$ (see Supplementary Information for details).

For intuition purposes, what constitutes a “large” PDI ? This is a challenging question since PDI is likely to increase as the frequency range considered increases, revealing additional modes. For example, we observe larger PDI when the $H(\tau)$ moment integrals are not truncated. As a reference for “large” polydispersity, consider power-law relaxation of a critical gel [108], with relaxation spectrum

Figure 12: The polydispersity index PDI for all samples is small, but noticeably larger than 1. Therapy putties shown as colored circles, PVA-Borax as dark gray circles, and a wormlike micelle solution [107] as hexagon symbol.

given by

$$H(\tau) = \frac{S}{\Gamma(n)} \tau^{-n} \quad (10)$$

where S and n are positive constants, $0 < n < 1$, and $\Gamma()$ is the gamma function. This is clearly a broad distribution of timescales. For a finite window of timescales ranging from $\tau_A - \tau_B$, where we define $X = \tau_B/\tau_A$, we derive analytical results for the truncated moment integrals, resulting in

$$PDI = \frac{(1-n)^2}{n(2-n)} X^n \left[\frac{(1 - \frac{1}{X^{2-n}})(1 - \frac{1}{X^n})}{(1 - \frac{1}{X^{1-n}})} \right]. \quad (11)$$

Note this PDI is independent of the specific values of τ_A and τ_B and only depends on the range of timescales via X . The square-bracket term approaches 1 in the limit of large X . As a specific example, consider the therapy putty range of frequencies $\omega = 0.1 - 500$ rad/s. After applying the Davies-Anderssen window, this gives $X = 216$. Using this range and a reference critical gel exponent $n = \frac{1}{2}$ gives $PDI = 5.3$. This can serve as a reference for “large” PDI for comparison in Figure 12 (see Supplementary Information for additional details).

Another route to interpret PDI , and its importance in describing the G' , G'' data, is via the three-parameter relaxation spectrum derived from the modulus weighted log-normal distribution

$$H(\tau) = H_{\max} \exp \left[-\frac{1}{2} \left(\frac{\ln(\tau) - \ln(\tau_{\max})}{\sigma} \right)^2 \right] \quad (12)$$

where $H_{\max}$ is the peak of the spectrum $H(\tau)$, $\tau_{\max}$ is the log-median relaxation timescale, and σ is the stan-

Figure 13: Three parameters are satisfactory to describe the linear viscoelastic behavior of therapy putties and PVA-Borax samples. The log-normal (3 D.O.F) adds only one parameter related to polydispersity but when fit to the data by a reduction of the residuals, is a marked improvement over the single-mode Maxwell model (with 2 D.O.F). (left) ‘firm’ putty and (right) 2.75 wt%PVA - 1.5wt% Borax sample.

dard deviation of the log-normal distribution. This three-parameter distribution analytically maps to the three parameters (η_0 , G_0 , PDI) as [61], $PDI = e^{\sigma^2}$, $G_0 = H_{\max}\sigma\sqrt{2\pi}$, and $\eta_0 = G_0\tau_{\max}e^{\frac{1}{2}\sigma^2}$. In addition to providing interpretation of PDI , the log-normal based spectrum can be used to check if the three-parameter description based on only (G_0 , η_0 , PDI) is reasonable to describe the data. Figure 13 shows the log-normal description of the experimental data, which is excellent for the putties and the PVA-Borax samples. This suggests that, for these materials at least,³ the three degree of freedom (D.O.F.) description is satisfactory and reasonably complete. We therefore move forward to propose how to represent this three-parameter description on an Ashby-style property map.

7 Ashby-style mapping and discussion

Ashby diagrams are widely used in design and material selection [3], though they rely on single-valued material properties. This is a challenge for rheological data, which is described by functions, not constants. Ashby-style plots for reduced rheology data have been demonstrated with

yield-stress fluids [12, 38–40] but not with linear viscoelastic fluids.

Here we have reduced the linear viscoelastic description of these materials to three independent parameters and we propose a method of visualizing this information on a two-dimensional cross-property plot in the style of Ashby diagrams. Figure 14 shows this visualization: symbols are located at coordinate (τ_1, G_0) , and a “tail” extends from the symbol, which terminates at the value τ_2 . This combines all the information from Table 2 and Figures 11–12 into a single visualization.

All five measures shown in Table 2 can be extracted from the two-dimensional plot in Figure 14. In addition to (τ_1, G_0, τ_2) as described, the viscosity is extracted as $\eta_0 = G_0\tau_1$, and reference values are shown as dashed lines. The $PDI = \tau_2/\tau_1$ and on the logarithmic scale in Figure 14 this directly relates to the length of the line, $\log(PDI) = \log(\tau_2) - \log(\tau_1)$.

A key concern in reducing rheologically-complex materials to low-dimensional descriptions is the loss of the important function-valued material response. Here, the PDI helps indicate the loss of information if only (τ_1, G_0) are used to describe the material. Higher-order measures could also be used to quantify how satisfactory the three-parameter description is, e.g. quantifying the disagreement between a log-normal description and the data (as in Figure 13), or quantifying higher moments of the relaxation spectrum $H(\tau)$.

The mapping in Figure 14 reveals several trends and patterns with the two materials considered here. The putties span a range of timescales τ_1 in addition to an order

³ Of course, PDI alone may not be satisfactory to describe the dispersity of relaxation timescales, particularly in the case of materials with vastly different relaxation behavior such as power-law relaxation [109, 110] or a multi-modal distribution of relaxation times [111, 112].

Figure 14: Ashby-style plot of high-dimensional linear viscoelastic behavior onto two-dimensional coordinates for the putties (color symbols at top) and the PVA-Borax (gray symbols in middle). Symbols are centered at coordinate location (τ_1, G_0) with a “tail” extending to a value of τ_2 to indicate the polydispersity of relaxation timescales.

of magnitude of viscosity η_0 . However, they have a near constant elasticity ($G_0 \approx 1$ MPa) across the different “firmnesses” (except extra-soft, due to the truncation window). The nearly constant G_0 for all putties facilitates some interpretation about what may be changing in the formulation and microstructure. It suggests that the number density of effective crosslinks is constant, a la rubber elasticity theory, be they from transient crosslinks of Borax or filler, or entanglements of the polymer. This could be tested by changing the crosslink density, for example, by varying the amount of a crosslinking additive, which should change the modulus G_0 . To maintain a constant G_0 (i.e. keep the crosslink number density constant), but change the relaxation timescale, different approaches could be used. For example, if entanglements dominate the relax-

ation timescale, then increasing molecular weight of the polymer would primarily lead to changes in η_0 and τ_1 [104, 113]. However, this may also change the distribution of timescales. A simpler hypothesis, in terms of changing formulation, is that different oils could be used with varying viscosity. This could potentially shift the timescales and viscosity η_0 , yet retain the same G_0 and a similar shape of the relaxation time distribution.

In contrast to the putties with constant modulus and changing timescale, the PVA-Borax timescales vary only from $\tau_1 = 0.2 - 0.8$ s. Thus, the material is a candidate for providing tactile intuition to answer the question: how does it “feel” to keep τ_1 reasonably constant (and within human perception) but change η_0 and G_0 . The nearly constant timescale implies that the relaxation behavior is somewhat independent of concentration implying that the timescale is primarily set by the thermoreversible crosslinking timescale [93, 114, 115].

Together, the PVA-Borax and physical therapy putties demonstrate two ways to span the linear viscoelastic property space.

8 Conclusions

In this work, we have demonstrated the use of a stress-input perspective Pipkin diagram to guide experimental characterization of physical therapy putties in the stress amplitude and time ranges relevant to the user experience of those materials. The stress-perspective Pipkin space proposed in this work aids in guiding the characterization and design of materials for which human perception is important. The space shown in Figure 5 is created specifically to be relevant to the tactile perception related to physical therapy putties. It can be further modified for other problems of interest such as food rheology by adjusting relevant regions to processes important to other user experiences (i.e. chewing, spreading, etc.).

Rheological characterization of complex materials is often a high-dimensional question. Using a model system of therapy putties designed for a range of user experiences and a PVA-Borax gel, we are able to show that viscoelastic constants calculated from a general continuous relaxation spectrum (found from simple linear viscoelastic tests) can reduce the complexity of the rheological description.

We show that in these systems, linear viscoelastic rheological properties are appropriate to differentiate between the user experiences of the putties. Reducing the complexity of the data allows for a clearer understanding of the differences in the materials and aids in data visual-

ization which leads to improved tactile intuition with viscoelastic materials.

With lower-dimensional descriptions of the linear viscoelastic response, we propose Ashby-style cross property plots that allow for the plotting of three proposed independent linear viscoelastic measures $[G_0, \tau_1, \tau_2]$. Such plots can be populated with a wide range of viscoelastic materials. Similar to traditional Ashby diagrams, these plots can be used to guide material selection in diverse applications including food and consumer products, vibration isolation, and soft robotics. These plots can also be used to guide the development of new materials into spaces not populated by existing materials.

Reducing the complexity of the data is not without its challenges. As we show, important information can be lost by this reduction from function-valued properties to low-dimensional parameters. Going forward, it will be important to find a systematic way to choose which low-dimensional representations are most useful for a specific design problem, be it correlation with tactile perception or with a performance metric (e.g. vibration isolation [41, 116]) and to account for any material behavior not included in the described low-dimensional metrics. The potential role of *nonlinear* rheological properties further widens the design space while exacerbating these important challenges.

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References

- [1] Ashby M.F., Material Selection in Mechanical Design, Butterworth-Heinemann, Boston, MA, 2 edition, 1999
- [2] Ashby M.F., Materials Selection in Mechanical Design, Elsevier, Oxford, 3 edition, 2005
- [3] Ashby M.F., Johnson K., Materials and Design: The Art and Science of Material Selection in Product Design, Elsevier, Oxford, 2 edition, 2010
- [4] MatWeb, MatWeb Material Property Data, available at <http://www.matweb.com>
- [5] United Laboratories, UL Prospector, available at <http://www.urlprospector.com>
- [6] Macosko C.W., Rheology: principles, measurements, and applications, Wiley-VCH, New York, 1994
- [7] Barnes H.A., Hutton J.F., Walters K., An Introduction to Rheology, volume 1, Elsevier, 1989
- [8] Smooth-on, Durometer Shore Hardness Scale, available at <https://www.smooth-on.com/page/durometer-shore-hardness-scale>
- [9] Shadwick R.E., Mechanical Design in Arteries, Journal of Experimental Biology, 202, 1999, 3305–3313
- [10] Autumn K., Liang Y.A., Hsieh S.T., Zesch W., Chan W.P., Kenny T.W., Fearing R., Full R.J., Adhesive Force of a Single Gecko Foot-hair., Nature, 405(6787), 2000, 681–5, 10.1038/35015073
- [11] Denny M., The Role of Gastropod Pedal Mucus in Locomotion, Nature, 285, 1980, 160–161
- [12] Ewoldt R.H., Extremely Soft: Design with Rheologically Complex Fluids, Soft Robotics, 1(1), 2014, 12–20, 10.1089/soro.2013.1508
- [13] Aksak B., Murphy M.P., Sitti M., Adhesion of Biologically Inspired Vertical and Angled Polymer Microfiber Arrays., Langmuir, 23(6), 2007, 3322–32, 10.1021/la062697t
- [14] Kim S., Sitti M., Biologically Inspired Polymer Microfibers with Spatulate Tips As Repeatable Fibrillar Adhesives, Applied Physics Letters, 89(26), 2006, 261911, 10.1063/1.2424442
- [15] Kim D.H., Rogers J.A., Stretchable Electronics: Materials Strategies and Devices, Advanced Materials, 20(24), 2008, 4887–4892, 10.1002/adma.200801788
- [16] Murphy M.P., Sitti M., Waalbot: An Agile Small-Scale Wall-Climbing Robot Utilizing Dry Elastomer Adhesives, IEEE/ASME Transactions on Mechatronics, 12(3), 2007, 330–338, 10.1109/TMECH.2007.897277
- [17] Qu L., Dai L., Stone M., Xia Z., Wang Z.L., Carbon Nanotube Arrays with Strong Shear Binding-On and Easy Normal Lifting-Off, Science, 322(5899), 2008, 238–42, 10.1126/science.1159503
- [18] Seok S., Onal C.D., Wood R., Rus D., Kim S., Peristaltic Locomotion with Antagonistic Actuators in Soft Robotics, IEEE International Conference on Robotics and Automation, 2010, 1228–1233
- [19] Laschi C., Cianchetti M., Mazzolai B., Margheri L., Follador M., Dario P., Soft Robot Arm Inspired by the Octopus, Advanced Robotics, 26(7), 2012, 709–727, 10.1163/156855312X626343
- [20] Lin H.T., Leisk G.G., Trimmer B., GoQBot: A Caterpillar-Inspired Soft-Bodied Rolling Robot., Bioinspiration & Biomimetics, 6(2), 2011, 026007, 10.1088/1748-3182/6/2/026007
- [21] Kim S., Laschi C., Trimmer B., Soft Robotics: A Bioinspired Evolution in Robotics, Trends in Biotechnology, 31(5), 2013, 287–94, 10.1016/j.tibtech.2013.03.002
- [22] Bird R.B., Armstrong R., Hassager O., Dynamics of Polymeric Liquids: Volume 1 Fluid Mechanics, John Wiley and Sons, Inc., New York, 2 edition, 1987

- [23] Middleman S., *Fundamentals of Polymer Processing*, McGraw-Hill, New York, 1977
- [24] Tadmore Z., *Principles of Polymer Processing*, Wiley, New York, 1979
- [25] Denn M.M., Fifty Years of Non-Newtonian Fluid Dynamics, *AIChE Journal*, 50(10), 2004, 2335–2345, 10.1002/aic.10357
- [26] Fischer C., Braun S.A., Bourban P.E., Michaud V., Plummer C.J.G., Manson J.A.E., Dynamic Properties of Sandwich Structures with Integrated Shear-Thickening Fluids, *Smart Materials and Structures*, 15(5), 2006, 1467–1475, 10.1088/0964-1726/15/5/036
- [27] Laun H.M., Rheology of Extremely Shear Thickening Polymer Dispersions (Passively Viscosity Switching Fluids), *Journal of Rheology*, 35(6), 1991, 999–1034, 10.1122/1.550257
- [28] Lee Y.S., Wetzel E.D., Wagner N.J., The Ballistic Impact Characteristics of Kevlar® Woven Fabrics Impregnated with A Colloidal Shear Thickening Fluid, *Journal of Materials Science*, 38(13), 2003, 2825–2833, 10.1023/A:1024424200221
- [29] Smay J.E., Cesarano J., Lewis J.A., Colloidal Inks for Directed Assembly of 3-D Periodic Structures, *Langmuir*, 18(14), 2002, 5429–5437, 10.1021/LA0257135
- [30] O'Bryan C.S., Bhattacharjee T., Niemi S.R., Balachandrar S., Baldwin N., Ellison S.T., Taylor C.R., Sawyer W.G., Angelini T.E., Three-Dimensional Printing with Sacrificial Materials for Soft Matter Manufacturing, *MRS Bulletin*, 42(08), 2017, 571–577, 10.1557/mrs.2017.167
- [31] M'Barki A., Bocquet L., Stevenson A., Linking Rheology and Printability for Dense and Strong Ceramics by Direct Ink Writing, *Scientific Reports*, 7(1), 2017, 6017, 10.1038/s41598-017-06115-0
- [32] Nelson A.Z., Schweizer K.S., Rauzan B.M., Nuzzo R.G., Vermant J., Ewoldt R.H., Designing and Transforming Yield-Stress Fluids, *Current Opinion in Solid State and Materials Science*, 23(5), 2019, 100758, 10.1016/j.cossms.2019.06.002
- [33] Aguado B.A., Mulyasmita W., Su J., Lampe K.J., Heilshorn S.C., Improving Viability of Stem Cells During Syringe Needle Flow Through the Design of Hydrogel Cell Carriers, *Tissue Engineering Part A*, 18(7-8), 2012, 806–815, 10.1089/ten.tea.2011.0391
- [34] Lopez Hernandez H., Grosskopf A.K., Stapleton L.M., Agmon G., Appel E.A., Non-Newtonian Polymer-Nanoparticle Hydrogels Enhance Cell Viability during Injection, *Macromolecular Bioscience*, 19(1), 2019, 1800275, 10.1002/mabi.201800275
- [35] Glassman M.J., Chan J., Olsen B.D., Reinforcement of Shear Thinning Protein Hydrogels by Responsive Block Copolymer Self-Assembly, *Advanced Functional Materials*, 23(9), 2013, 1182–1193, 10.1002/adfm.201202034
- [36] Yan C., Mackay M.E., Czymmek K., Nagarkar R.P., Schneider J.P., Pochan D.J., Injectable Solid Peptide Hydrogel as a Cell Carrier: Effects of Shear Flow on Hydrogels and Cell Payload, *Langmuir*, 28(14), 2012, 6076–6087, 10.1021/la2041746
- [37] Cai L., Dewi R.E., Heilshorn S.C., Injectable Hydrogels with In Situ Double Network Formation Enhance Retention of Transplanted Stem Cells, *Advanced Functional Materials*, 25(9), 2015, 1344–1351, 10.1002/adfm.201403631
- [38] Ewoldt R.H., Clasen C., Hosoi A.E., McKinley G.H., Rheological Fingerprinting of Gastropod Pedal Mucus and Synthetic Complex Fluids for Biomimicking Adhesive Locomotion, *Soft Matter*, 3(5), 2007, 634–643, 10.1039/b615546d
- [39] Nelson A.Z., Ewoldt R.H., Design of Yield-Stress Fluids: A Rheology-to-Structure Inverse Problem, *Soft Matter*, 13(41), 2017, 7578–7594, 10.1039/C7SM00758B
- [40] Nelson A.Z., Bras R.E., Liu J., Ewoldt R.H., Extending Yield-Stress Fluid Paradigms, *Journal of Rheology*, 62(1), 2018, 357–369, 10.1122/1.5003841
- [41] Corman R.E., Rao L., Bharadwaj N.A., Allison J.T., Ewoldt R.H., Setting Material Function Design Targets for Linear Viscoelastic Materials and Structures, *Journal of Mechanical Design*, 138(5), 2016, 051402, 10.1115/1.4032698
- [42] Xu H., Li Y., Brinson C., Chen W., Brinson L.C., Chen W., Brinson C., Chen W., A Descriptor-Based Design Methodology for Developing Heterogeneous Microstructural Materials System, *Journal of Mechanical Design*, 136(5), 2014, 051007, 10.1115/1.4026649
- [43] Olson G.B., Designing a New Material World, *Science*, 288(5468), 2000, 993–998
- [44] Scott Blair G., Psychorheology: Links Between The Past and The Present, *Journal of Texture Studies*, 5(1), 1974, 3–12, 10.1111/j.1745-4603.1974.tb01083.x
- [45] Moskowitz H.R., Psychorheology — Its Foundations and Current Outlook, *Journal of Texture Studies*, 8(2), 1977, 229–246
- [46] Moskowitz H.R., On Fitting Equations to Sensory Data: A Point of View and a Paradox in Modeling and Optimizing, *Journal of Sensory Studies*, 15(1), 2000, 1–19, 10.1111/j.1745-459X.2000.tb00403.x
- [47] Szczesniak A.S., Correlating Sensory with Instrumental Texture Measurements — An Overview of Recent Developments, *Journal of Texture Studies*, 18(1), 1987, 1–15, 10.1111/j.1745-4603.1987.tb00566.x
- [48] Kokini J.L., Kadane J.B., Cussler E.L., Liquid Texture Perceived in the Mouth, *Journal of Texture Studies*, 8(2), 1977, 195–218, 10.1111/j.1745-4603.1977.tb01175.x
- [49] Voisey P.W., Modernization of Texture Instrumentation, *Journal of Texture Studies*, 2(2), 1971, 129–195, 10.1111/j.1745-4603.1971.tb00580.x
- [50] Elejalde C.C., Kokini J.L., The Psychophysics of Pouring, Spreading and In-Mouth Viscosity, *Journal of Texture Studies*, 23(3), 1992, 315–336, 10.1111/j.1745-4603.1992.tb00528.x
- [51] Henry W.F., Katz M.H., Pilgrim F.J., May A., Texture of Semi-Solid Foods: Sensory and Physical Correlates, *Journal of Food Science*, 36(1), 1971, 155–161, 10.1111/j.1365-2621.1971.tb02059.x
- [52] Dahl D.W., Moreau P., The Influence and Value of Analogical Thinking During New Product Ideation, *Journal of Marketing Research*, 39(1), 2002, 47–60, 10.1509/jmkr.39.1.47.18930
- [53] Needham D.R., Begg I.M., Problem-Oriented Training Promotes Spontaneous Analogical Transfer: Memory-Oriented Training Promotes Memory for Training, *Memory & Cognition*, 19(6), 1991, 543–557, 10.3758/BF03197150
- [54] Chan J., Fu K., Schunn C., Cagan J., Wood K., Kotovsky K., On the Benefits and Pitfalls of Analogies for Innovative Design: Ideation Performance Based on Analogical Distance, Commonness, and Modality of Examples, *Journal of Mechanical Design*, 133(8), 2011, 081004, 10.1115/1.4004396
- [55] Atman C.J., Cardella M.E., Turns J., Adams R., Comparing Freshman and Senior Engineering Design Processes: An In-Depth Follow-Up Study, *Design Studies*, 26(4), 2005, 325–357, 10.1016/j.destud.2004.09.005
- [56] Barbe W.B., Swassing R.H., Milone M.N., Teaching Through Modality Strengths: Concepts and Practices, Zaner-Bloser, Columbus, Ohio, 1979
- [57] Granta: CES Selector Materials Selection Software, available at <http://www.grantadesign.com/products/ces/>

- [58] Davis J.G., The Rheology of Cheese, Butter and Other Milk Products. (The Measurement of “Body” and “Texture”), *Journal of Dairy Research*, 8(02), 1937, 245, 10.1017/S0022029900002090
- [59] Ferry J.D., *Viscoelastic Properties of Polymers*, John Wiley & Sons, Inc, New York, 3 edition, 1980
- [60] Tschoegl N.W., *The Phenomenological Theory of Linear Viscoelastic Behavior*, Springer-Verlag, Berlin, 1989
- [61] Martinetti L., Soulages J.M., Ewoldt R.H., Continuous Relaxation Spectra for Constitutive Models in Medium-Amplitude Oscillatory Shear, *Journal of Rheology*, 62(5), 2018, 1271–1298, 10.1122/1.5025080
- [62] Armitage D., Hughes M., Sinden A., The Preparation of “Bouncing Putty”: An Undergraduate Experiment in Silicone Chemistry, *Journal of Chemical Education*, 50(6), 1973, 434
- [63] Minuto M.A., Method of Making Bouncing Silicone Putty-Like Compositions, 1983, US Patent 4,371,493 A
- [64] Wright J.G.E., Process for Making Puttylike Elastic Plastic, Siloxane Derivative Composition Containing Zinc Hydroxide, 1951, US Patent 2,541,851 A
- [65] McGregor R.R., Warrick E.L., Treating Dimethyl Silicone Polymer with Boric Oxide, 1967, US Patent 2,431,878 A
- [66] Linear viscoelasticity of complex coacervates, *Advances in Colloid and Interface Science*, 239, 2017, 46–60, 10.1016/j.cis.2016.08.010
- [67] Muderick A., Therapeutic Putty with Increasing or Decreasing Stiffness, 2009, US Patent 7,618,349 B1
- [68] Gibbon R.M., Monitor Putty with Increasing Stiffness, 1997, US Patent 5,693,689
- [69] Ewoldt R.H., Bharadwaj N.A., Low-Dimensional Intrinsic Material Functions for Nonlinear Viscoelasticity, *Rheologica Acta*, 52(3), 2013, 201–219, 10.1007/s00397-013-0686-6
- [70] Kokini J.L., Cussler E.L., Predicting the Texture of Liquid and Melting Semi-Solid Foods, *Journal of Food Science*, 48(4), 1983, 1221–1225, 10.1111/j.1365-2621.1983.tb09196.x
- [71] Cook D.J., Oral Shear Stress Predicts Flavour Perception in Viscous Solutions, *Chemical Senses*, 28(1), 2003, 11–23, 10.1093/chemse/28.1.11
- [72] Bharadwaj N.A., Ewoldt R.H., The General Low-Frequency Prediction for Asymptotically Nonlinear Material Functions in Oscillatory Shear, *Journal of Rheology*, 58(4), 2014, 891–910, 10.1122/1.4874344
- [73] Pipkin A.C., *Lectures on Viscoelasticity Theory*, Springer, New York, 1972
- [74] Ewoldt R.H., McKinley G.H., Mapping Thixo-Elasto-Visco-Plastic Behavior, *Rheologica Acta*, 56, 2017, 195–210, 10.1007/s00397-017-1001-8
- [75] Dealy J.M., Weissenberg and Deborah Numbers — Their Definition and Use, *Rheology Bulletin*, 79(2), 2010
- [76] Ewoldt R.H., Gurnon A., Lopez-Barron C., McKinley G.H., Swan J., Wagner N.J., A Report on “LAOS Rheology Day” Held Friday the 13th at the Colburn Laboratory, University of Delaware, *Rheology Bulletin*, 81(2), 2012
- [77] Dimitriou C.J., Ewoldt R.H., McKinley G.H., Describing and Prescribing the Constitutive Response of Yield Stress Fluids Using Large Amplitude Oscillatory Shear Stress (LAOS_{stress}), *Journal of Rheology*, 57(1), 2013, 27–70, 10.1122/1.4754023
- [78] Skedung L., Danerlöv K., Olofsson U., Michael Johannessson C., Aikala M., Kettle J., Arvidsson M., Berglund B., Rutland M.W., Tactile Perception: Finger Friction, Surface Roughness and Perceived Coarseness, *Tribology International*, 44(5), 2011, 505–512, 10.1016/j.triboint.2010.04.010
- [79] Skedung L., Arvidsson M., Chung J.Y., Stafford C.M., Berglund B., Rutland M.W., Feeling Small: Exploring The Tactile Perception Limits, *Scientific Reports*, 3, 2013, 2617, 10.1038/srep02617
- [80] Crosby C.A., Wehbe M.A., Hand Strength: Normative Values, *The Journal of Hand Surgery*, 19(4), 1994, 665–670, 10.1016/0363-5023(94)90280-1
- [81] Hand Anthropometry, accessed at https://usability.gtri.gatech.edu/eou_info/hand_anthro.php
- [82] Ashendorf L., Vanderslice-Barr J.L., McCaffrey R.J., Motor Tests and Cognition in Healthy Older Adults, *Applied Neuropsychology*, 16(3), 2009, 171–176, 10.1080/09084280903098562
- [83] Hubel K.A., Yund E.W., Herron T.J., Woods D.L., Computerized Measures of Finger Tapping: Reliability, Malingering and Traumatic Brain Injury, *Journal of Clinical and Experimental Neuropsychology*, 35(7), 2013, 745–758, 10.1080/13803395.2013.824070
- [84] Potter M.C., Wyble B., Hagmann C.E., McCourt E.S., Detecting Meaning in RSVP at 13 ms per Picture, *Attention, perception & psychophysics*, 2013, 10.3758/s13414-013-0605-z
- [85] Cambridge Polymer Group, The Cambridge Polymer Group Silly Putty(TM) “Egg”, Technical report, 2002, available at <http://www.campoly.com/files/6713/5216/6040/sillyputty.pdf>
- [86] Edgeworth R., Dalton B.J., Parnell T., The Pitch Drop Experiment, *European Journal of Physics*, 5(4), 1984, 198–200, 10.1088/0143-0807/5/4/003
- [87] Here’s What Happens When You Drop 100 Pounds of Silly Putty Off of a 141-Foot Building, 2017, available at <https://www.wral.com/here-s-what-happens-when-you-drop-100-pounds-of-silly-putty-off-of-a-141-foot-building/16760896/>
- [88] Morioka M., Griffin M.J., Thresholds for the Perception of Hand-Transmitted Vibration: Dependence on Contact Area and Contact Location, *Somatosensory & Motor Research*, 22(4), 2005, 281–97, 10.1080/08990220500420400
- [89] Brisben A.J., Hsiao S.S., Johnson K.O., Detection of Vibration Transmitted Through an Object Grasped in the Hand, *Journal of Neurophysiology*, 81(4), 1999, 1548–1558, 10.1152/jn.1999.81.4.1548
- [90] Astin A.D., Finger Force Capability: Measurement and Prediction Using Anthropometric and Myoelectric Measures, Ms thesis, Virginia Polytechnic Institute and State University, 1999
- [91] Ewoldt R.H., Johnston M.T., Caretta L.M., Experimental challenges of shear rheology: how to avoid bad data, in S. Spagnoli, ed., *Complex Fluids in Biological Systems*, Springer, 2015, 207–241
- [92] Carey-De La Torre O., Elastic Stiffening in PVA-Borax Studied with Experimental Medium Amplitude Oscillatory Shear, Ms thesis, Department of Mechanical Science and Engineering, University of Illinois at Urbana-Champaign, 2017
- [93] Martinetti L., Carey-De La Torre O., Schweizer K.S., Ewoldt R.H., Inferring the Nonlinear Mechanisms of a Reversible Network, *Macromolecules*, 51(21), 2018, 8772–8789, 10.1021/acs.macromol.8b01295
- [94] Carey-De La Torre O., Ewoldt R.H., First-Harmonic Nonlinearities Can Predict Unseen Third-Harmonics in Medium-Amplitude Oscillatory Shear (MAOS), *Korea-Australia Rheology Journal*, 30(1), 2018, 1–10, 10.1007/s13367-018-0001-2
- [95] Honerkamp J., Weese J., Determination of the relaxation spectrum by a regularization method, *Macromolecules*, 22(11), 1989, 4372–4377, 10.1021/ma00201a036
- [96] Honerkamp J., Weese J., A Nonlinear Regularization Method for the Calculation of Relaxation Spectra, *Rheologica Acta*, 32(1),

- 1993, 65–73, 10.1007/BF00396678
- [97] Winter H., Analysis of Dynamic Mechanical Data: Inversion into a Relaxation Time Spectrum and Consistency Check, *Journal of Non-Newtonian Fluid Mechanics*, 68(2-3), 1997, 225–239, 10.1016/S0377-0257(96)01512-1
- [98] Stadler F.J., Bailly C., A New Method for the Calculation of Continuous Relaxation Spectra From Dynamic-Mechanical Data, *Rheologica Acta*, 48(1), 2009, 33–49, 10.1007/s00397-008-0303-2
- [99] Baumgaertel M., Winter H., Interrelation Between Continuous and Discrete Relaxation Time Spectra, *Journal of Non-Newtonian Fluid Mechanics*, 44, 1992, 15–36, 10.1016/0377-0257(92)80043-W
- [100] Friedrich C., Braun H., Weese J., Determination of Relaxation Time Spectra by Analytical Inversion Using a Linear Viscoelastic Model with Fractional Derivatives, *Polymer Engineering and Science*, 35(21), 1995, 1661–1669, 10.1002/pen.760352102
- [101] McDougall I., Orbey N., Dealy J.M., Inferring Meaningful Relaxation Spectra from Experimental Data, *Journal of Rheology*, 58(3), 2014, 779–797, 10.1122/1.4870967
- [102] Davies A., Anderssen R., Sampling Localization in Determining the Relaxation Spectrum, *Journal of Non-Newtonian Fluid Mechanics*, 73(1-2), 1997, 163–179, 10.1016/S0377-0257(97)00056-6
- [103] Cole K.S., Cole R.H., Dispersion and Absorption in Dielectrics I. Alternating Current Characteristics, *The Journal of Chemical Physics*, 9(4), 1941, 341–351, 10.1063/1.1750906
- [104] Larson R.G., *The Structure and Rheology of Complex Fluids*, Oxford University Press, Oxford, 1999
- [105] Berret J.F., Appell J., Porte G., Linear Rheology of Entangled Wormlike Micelles, *Langmuir*, 9(11), 1993, 2851–2854, 10.1021/la00035a021
- [106] Georgieva G.S., Anachkov S.E., Lieberwirth I., Koynov K., Kralchevsky P.A., Synergistic Growth of Giant Wormlike Micelles in Ternary Mixed Surfactant Solutions: Effect of Octanoic Acid, *Langmuir*, 32(48), 2016, 12885–12893, 10.1021/acs.langmuir.6b03955
- [107] Nakaya-Yaegashi K., Ramos L., Tabuteau H., Ligoure C., Linear Viscoelasticity of Entangled Wormlike Micelles Bridged by Telechelic Polymers: An Experimental Model for a Double Transient Network, *Journal of Rheology*, 52(2), 2008, 359–377, 10.1122/1.2828645
- [108] Chambon F., Petrovic Z.S., MacKnight W.J., Winter H.H., Rheology of Model Polyurethanes at the Gel Point, *Macromolecules*, 19(8), 1986, 2146–2149, 10.1021/ma00162a007
- [109] Winter H.H., Chambon F., Analysis of Linear Viscoelasticity of a Crosslinking Polymer at the Gel Point, *Journal of Rheology*, 30(2), 1986, 367–382, 10.1122/1.549853
- [110] Chambon F., Winter H.H., Linear Viscoelasticity at the Gel Point of a Crosslinking PDMS with Imbalanced Stoichiometry, *Journal of Rheology*, 31(8), 1987, 683–697, 10.1122/1.549955
- [111] Grindy S.C., Learsch R., Mozdhehi D., Cheng J., Barrett D.G., Guan Z., Messersmith P.B., Holten-Andersen N., Control of Hierarchical Polymer Mechanics with Bioinspired Metal-Coordination Dynamics, *Nature Materials*, 14, 2015, 1210–1216, 10.1038/nmat4401
- [112] Grindy S.C., Holten-Andersen N., Weber W., Anseth K.S., Spatz J.P., Bowman C.N., Messersmith P.B., Holten-Andersen N., Bio-Inspired Metal-Coordinate Hydrogels with Programmable Viscoelastic Material Functions Controlled by Longwave UV Light, *Soft Matter*, 13(22), 2017, 4057–4065, 10.1039/C7SM00617A
- [113] Rubinstein M., Colby R.H., *Polymer Physics*, Oxford University Press, Oxford, 2003
- [114] Koike A., Nemoto N., Inoue T., Osaki K., Dynamic Light Scattering and Dynamic Viscoelasticity of Poly(vinyl alcohol) in Aqueous Borax Solutions. 1. Concentration Effect, *Macromolecules*, 28(7), 1995, 2339–2344, 10.1021/ma00111a029
- [115] Nemoto N., Koike A., Osaki K., Dynamic Light Scattering and Dynamic Viscoelasticity of Poly(vinyl alcohol) in Aqueous Borax Solutions. 2. Polymer Concentration and Molecular Weight Effects, *Macromolecules*, 29(5), 1996, 1445–1451, 10.1021/ma951101y
- [116] Lee Y.H., Corman R.E., Ewoldt R.H., Allison J.T., A Multiobjective Adaptive Surrogate Modeling-Based Optimization (MO-ASMO) Framework Using Efficient Sampling Strategies, *Proceedings of the ASME 2017 International Design Engineering Technical Conferences and Computers and Information in Engineering Conference*, 2017, 10.1115/DETC2017-67541, paper DETC2017-67541, V02BT03A023

A Appendix: Rheology short course module for hands-on linear viscoelasticity

The following figures specifically compare two of the putties used for teaching hands-on perception of linear viscoelasticity: green (firm) and yellow (soft). Participants are

Figure A1: Soft (yellow) and firm (green) putties are subjected to a step-strain at 1% strain. The stress σ relaxation behavior is normalized by the input strain γ_0 and reported as a relaxation modulus $G(t)$. Experimental limits (short time and minimum torque) are shown as shaded regions.

Figure A2: Soft (yellow) and firm (green) putties are subjected to a step-stress of 100 Pa to mimic loading during use for physical therapy applications, which are more readily thought of as stress-controlled loading scenarios. The resulting material strain γ is normalized by the input stress σ_0 and reported as a creep compliance $J(t)$. Intuitively, the softer putty (yellow) is more compliant than the firmer putty.

given samples of each putty and before seeing any rheological data are invited to consider the following prompts:

1. What is different (rheologically) between the putties?
 - Push, pull, bounce, and play.
 - Is the difference you feel and observe viscous? elastic? relaxation timescale?
2. What rheological test would measure this?
 - What deformation? (shear, extension,...)
 - What is controlled input? (strain, strain-rate, stress,...)
 - What scheduling of input? (step, oscillation,...)

After discussing these questions, the answers are revealed in Figures A1–A4, in addition to other figures for the entire range of putties reported in our work here.

This module has been used in lectures at the home university of the authors, in addition to several rheology short courses with significant participation from industry. These courses have been hosted in Houston, TX (July 2015), Buenos Aires, Argentina (April 2016), Minneapolis, MN (June 2016, August 2018), Istanbul, Turkey (April 2017), Irvine, CA (May 2017), Stanford, CA (June 2019), and at KU Leuven, Belgium (September 2017, September 2019).

Finally, a simple summary of the putties is shown in Figure A5 which places all the putties on number lines of viscosity and elastic modulus, in a format similar to Figure 1.

Figure A3: Small amplitude oscillatory shear experiments for the soft (yellow) and firm (green) putties with an input stress amplitude of $\sigma_0 = 100$ Pa. Filled symbols represent the elastic or storage modulus G' ; open symbols represent the viscous or loss modulus G'' .

Figure A4: The same data shown in Figure A3 is fit to a single-model Maxwell model, represented with the solid G' and dashed G'' black lines. Fit parameters are shown for each putty. There is disagreement between the model and the experimental data at low frequencies in the elastic modulus G' and high frequencies in the viscous modulus G'' .

Figure A5: Low-dimensional descriptions of complex materials can help ground these materials within the frameworks for which intuition already exists. Figure adapted from [6, 7], and [8].