Gaussian Process to Identify Hydrogel Constitutive Model

Jikun Wang, Tianjiao Li, Chung-Yuen Hui, Jingjie Yeo, Alan Zehnder

Mechanical and Aerospace Engineering
Cornell University
Ithaca NY 14853

ABSTRACT
Unlike traditional structural materials, soft solids can often sustain very large deformation before failure, and many exhibit nonlinear viscoelastic behavior. Modeling nonlinear viscoelasticity is a challenging problem for a number of reasons. In particular, a large number of material parameters are needed to capture material response and validation of models can be hindered by limited amounts of experimental data available. We have developed a Gaussian Process (GP) approach to determine the material parameters of a constitutive model describing the mechanical behavior of a soft, viscoelastic PVA hydrogel. A large number of stress histories generated by the constitutive model constitute the training sets. The low-rank representations of stress histories by Singular Value Decomposition (SVD) are taken to be random variables which can be modeled via Gaussian Processes with respect to the material parameters of the constitutive model. We obtain optimal material parameters by minimizing an objective function over the input set. We find that there are many good sets of parameters. Further the process reveals relationships between the model parameters. Results so far show that GP has great potential in fitting constitutive models.

Keywords: Viscoelastic, Tension Test, Finite Deformation, Large Strain, Stress Relaxation, Machine Learning

INTRODUCTION

Many materials, for example, soft solids such as rubber and gels, are viscoelastic or viscoplastic solids. These materials, besides being rate-sensitive, can sustain very large deformation before failure. To describe their mechanical properties, researchers have designed many complex constitutive models. However, most constitutive models contain many material parameters that cannot be directly determined by experimental data. In addition, validating the theoretical model is hindered by the limited amount of experimental data available. The process of fitting experimental data to theory can be extremely tedious and time-consuming. When the number of material parameters is large, poor fitting of data can occur even if the model captures the correct physics. To our best knowledge, it is still a great challenge to rapidly determine the parameters for complex viscoelastic constitutive models. Here we propose a method to find the parameters for constitutive models, which combines singular value decomposition (SVD) and machine learning tools, specifically, Gaussian process. Although our formulation is general, we demonstrate its usage and validate our algorithm by applying it to study the mechanical behavior of a nonlinear viscoelastic PVA hydrogel.

PVA CONSTITUTIVE MODEL

In our previous works [1], we have developed a 3D constitutive model which combines the finite strain elasticity of elastomers with the kinetics of bond breaking and reattachment. We have also demonstrated that our model accurately predicts results from uniaxial tension and torsion tests with complex loading histories. The constitutive model for the PVA gel is completely determined by four material parameters $\mu\rho, \alpha_B, t_B$ and $\mu\bar{\gamma}_\infty$. Hence a parameter set is specified by a four-component vector $\vec{x} = (\mu\rho, \alpha_B, t_B, \mu\bar{\gamma}_\infty)$. According to our constitutive model, in a uniaxial tension test where the stretch ratio $\lambda(t)$ is prescribed, the nominal stress $\sigma(t)$ corresponding to the parameter set $\vec{x} \equiv (\mu\rho, t_B, t_B, \mu\bar{\gamma}_\infty) \in \Omega \subset \mathbb{R}^4$ is

$$\sigma(t) = \left[\mu\rho + \mu\bar{\gamma}_\infty \frac{t_B}{2-\alpha_B}\left(1 + (\alpha_B - 1)\frac{t}{t_B}\right)^{\frac{2-\alpha_B}{1-\alpha_B}}\right]\left[\lambda(t) - \frac{1}{\lambda^2(t)}\right] + \mu\bar{\gamma}_\infty \int_0^t \phi_B\left(\frac{t-\tau}{t_B}\right)\left[\frac{\lambda(t)}{\lambda^2(\tau)} - \frac{\lambda(\tau)}{\lambda^2(t)}\right] d\tau \qquad (1a)$$

where

$$\phi_B\left(\frac{t}{t_B}\right) = \left(1 + (\alpha_B - 1)\frac{t}{t_B}\right)^{\frac{1}{1-\alpha_B}}. \tag{1b}$$

The units of parameters are $(\mu\rho, \alpha_B, t_B, \mu\bar{\gamma}_\infty) \sim (kPa,\ 1,\ sec,\ kPa)$

GAUSSIAN PROCESS MACHINE LEARNING

Singular value decomposition (SVD) and Gaussian process have been used by different groups of researchers to predict material behavior or to find crucial parameters for controlling system behavior [2, 3]. Here we use these tools to help us predict the output of our constitutive model, i.e., stress history. For a fixed strain history, we can use the constitutive model to calculate the stress history for different parameters. If we put all those stress histories together, we can get a stress matrix and apply singular value decomposition on it to get the basis and principal components of each stress history. Then we use the principal components of all those stress histories to train a Gaussian process. After training, under the same strain history, this Gaussian process can make predictions about the principal components of the stress history of any parameters it has not seen. As a result, we can use GP to evaluate the stress history over a large number of parameters then find the parameters that best fit the experimental data. The flowchart of Gaussian process machine learning is shown in Figure 1.

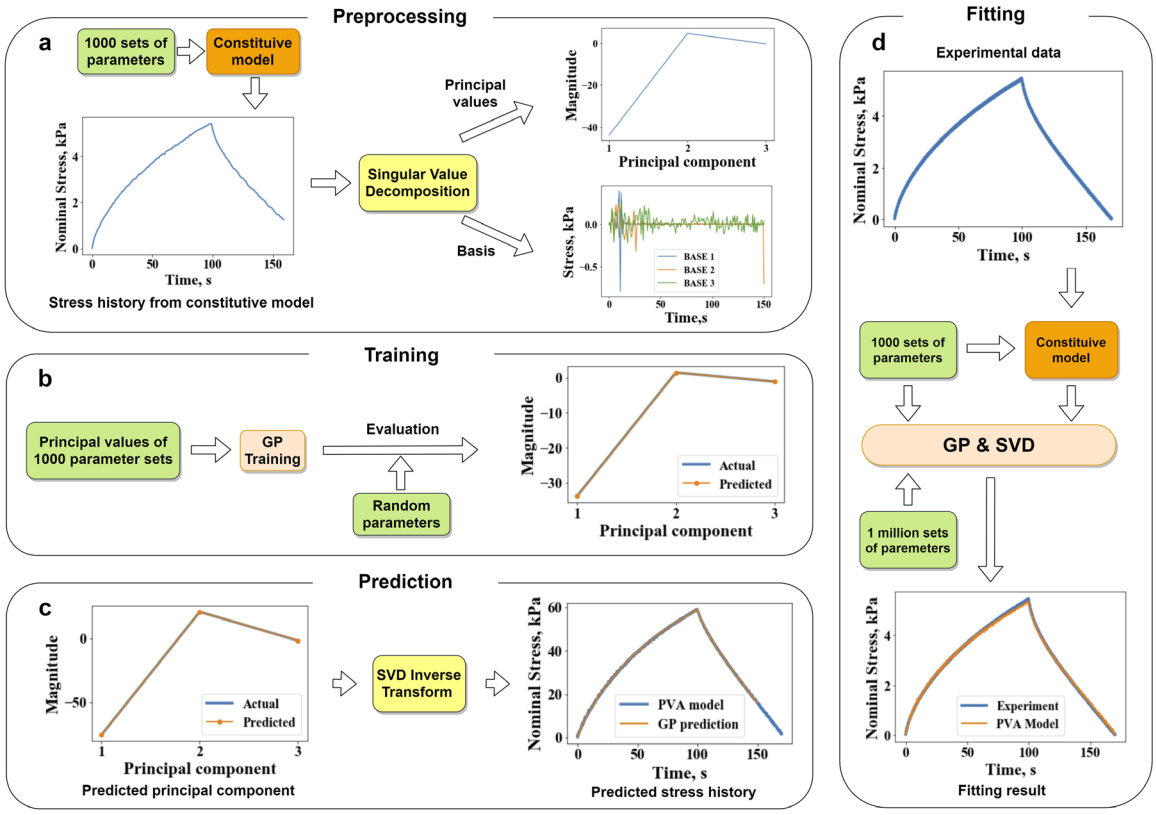

Figure 1: Workflow of Gaussian Process to identify constitutive model. (a) 1000 stress history curves calculated directly from the constitutive model are decomposed into basis and projections on this basis (i.e. principal components) using SVD; (b) the principal components of the 1000 parameter sets are taken as training set. After training, the GP can make predictions about the principal components of any parameter set not in the training set; (c) with the predicted principal components, the stress history of any parameter set can be predicted by applying SVD inverse transform to the predicted principal component without calculating the constitutive model; (d) the best parameters for the experimental data can be obtained by comparing the experimental stress history with the stress histories calculated for a large number of parameter sets.

EXPERIMENTAL METHODS

The dual-crosslink poly(vinyl alcohol) (PVA) hydrogels were prepared by incorporating ions in a chemically crosslinked PVA gel. This hydrogel system was first introduced by Mayumi et al [4]. First a 16 wt% PVA solution was made by dissolving PVA powder in distilled water. Then the PVA solution was chemically crosslinked by glutaraldehyde in an acidic environment. Then the chemical gel was washed several times to neutralize the pH. Finally, the gel was soaked in Borax/NaCl solution to form the physical bonds. The gel was soaked in the solution for at least 3 days to reach equilibrium prior to testing.

We performed uniaxial tension tests using a single PVA hydrogel specimen with four different strain histories. Some of the physical bonds will be broken after a test. Fortunately, the dual-crosslink PVA hydrogel has self-healing property; the bonds can reform and the gel's mechanical properties can totally recover to their original state after 30 minutes at room temperature.

The tests were performed using a custom-built tensile tester with the samples immersed in a mineral oil bath to prevent the specimens from drying. We conducted EXP 1, EXP 2, EXP 3 and EXP 4 sequentially, as shown in Figure 1. After each test, we kept the specimen in the oil bath 30 minutes to let it recover to the as-prepared state.

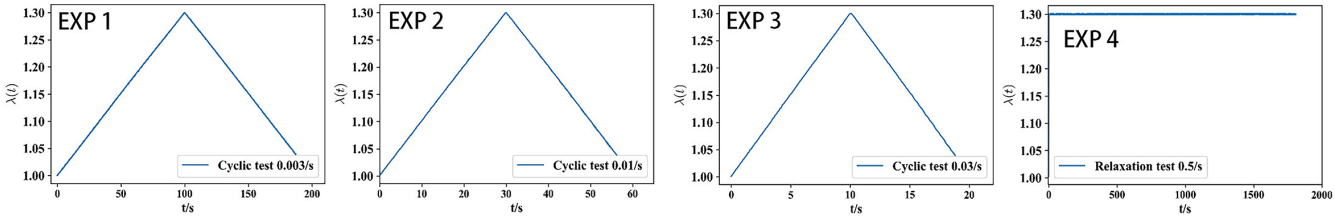

Figure 2: Four loading histories for PVA gels

RESULTS AND DISCUSSION

The best fitting result given by GP is illustrated in Figure 3. The agreement between experiments and theory is excellent. These results further demonstrate that our PVA constitutive model correctly captures the mechanical behavior of PVA gels. In addition, it shows that our machine learning algorithm is a powerful tool for determining material parameters in constitutive models.

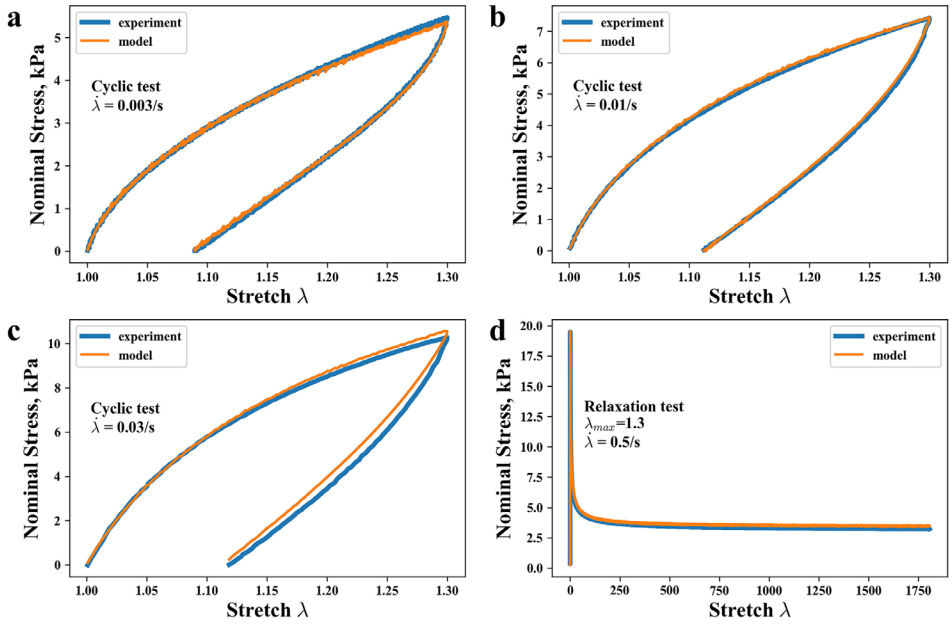

Figure 3: Comparison of model prediction to experiments using the parameters given by GP

Furthermore, using our method, we discovered that there are many sets of material parameters that fit the experiments well. The projections of those parameters on 2D subspaces are drawn in Figure 4, and these plots show that there are strong relationships between $\mu\rho$ and α_B, as well as t_B and $\mu\bar{\gamma}_\infty$. To understand the relationships, we use a result from our previous work [1], which demonstrates that when strains are small, that is, when $\lambda(t) \approx 1 + \epsilon(t)$, the constitutive model becomes

$$\sigma(t) = \int_{-\infty}^{t} Y\left(\frac{t-\tau}{t_B}\right)\frac{d\epsilon(\tau)}{d\tau}d\tau \tag{2a}$$

where

$$Y(t) \equiv \frac{3\mu\bar{\gamma}_\infty t_B}{2-\alpha_B}\left(1 + (\alpha_B - 1)\frac{t}{t_B}\right)^{\frac{2-\alpha_B}{1-\alpha_B}} + 3\mu\rho \tag{2b}$$

From this equation, it is clear that $\sigma(t)$ depends linearly on $\mu\bar{\gamma}_\infty$. Then $\mu\bar{\gamma}_\infty t_B$ must be a material constant C for a specific experiment, therefore $\mu\bar{\gamma}_\infty = C/t_B$. And it is clear that $\sigma(t)$ increases with $\mu\rho$ and α_B. Therefore, to produce the same stress history, a smaller α_B must be chosen if a larger value of $\mu\rho$ is already chosen. Figure 4 shows the machine learning predictions are consistent with those relationships, which further demonstrates the power of our method.

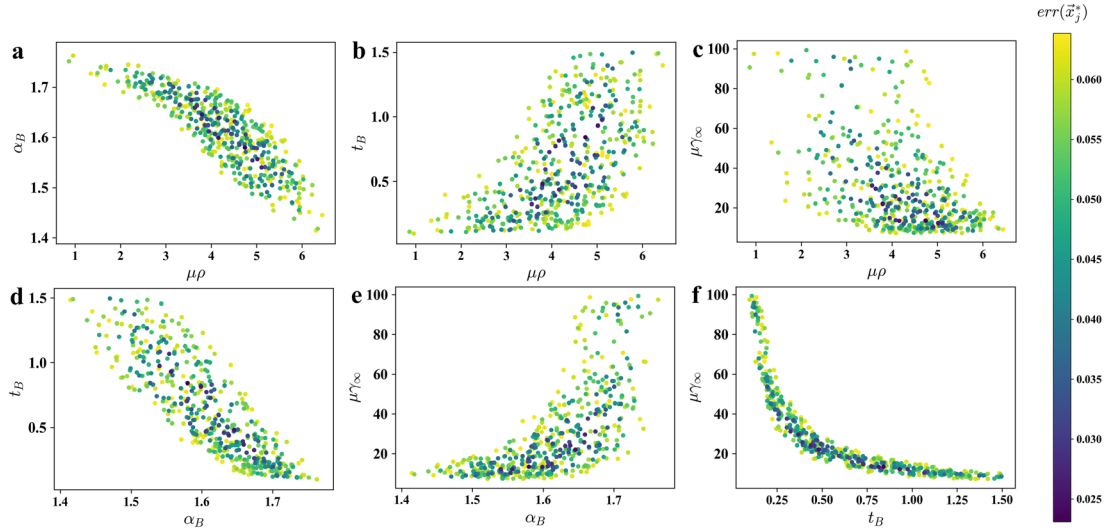

Figure 4: Distribution of 500 parameter sets which fit experiments very well.

Theoretically, all the results above can be obtained by calculating the constitutive model for 1 million parameter sets directly, however, a prediction that would take 1.6 minutes using SVD and Gaussian process metamodel for 1 million parameter vectors would take up to 322 hours to evaluate using the constitutive model. For more complicated constitutive models and longer strain histories, it is impractical to calculate the stress history for millions of parameter sets. This is the main advantage of our method over the traditional methods of data fitting.

ACKNOWLEDGEMENTS

This material is based upon work supported by the National Science Foundation, under Grant No. CMMI-1903308 and Grant No. CMMI-2038057.

REFERENCES

[1] J. Guo, R. Long, K. Mayumi and C.Y. Hui, "Mechanics of a dual cross-link gel with dynamic bonds: Steady state kinetics and large deformation effects, " **Macromolecules, vol. 49**, pp.3497-3507, (2016).

[2] M. Guo and J.S. Hesthaven, "Data-driven reduced order modeling for time-dependent problems," **Computer methods in applied mechanics and engineering, vol. 345,** pp.75-99, (2019).

[3] C. Yang, Y. Kim, S. Ryu, and G.X. Gu, 2020, " Prediction of composite microstructure stress-strain curves using convolutional neural networks," **Materials & Design, vol. 189,** p.108509, (2020).

[4] K. Mayumi, A. Marcellan, G. Ducouret, C. Creton, and T. Narita, "Stress strain relationship of of highly stretchable dual cross-link gels: separability of strain and time effect," **ACS Macro Letters, vol. 2**, pp. 1065-1069, (2013).