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Data-driven macro-micro-macro modelling of rubber-like materials

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Abstract

The characterization of the multiaxial mechanical behaviour of polymers has been challenging, as the vast number of proposed models demonstrates. These models are based on analytical expressions of sate variables (invariants or principal stretches). Machine learning brings new tools to characterize polymers from macroscopic experiments. However, classical machine learning modelling as classical Neural Networks have several drawbacks, as the need for extensive data, the lack of robustness, and the lack of compliance with physical principles. In polymers, physics-based machine learning brings the best of both worlds by performing data-driven characterization considering physical principles and reducing the number of needed tests. In this work, using a simple procedure for crossing scales, we present a new data-driven procedure to characterize the entropic behaviour of a representative macromolecule directly from any single macroscopic test on the polymer by solving a linear system of equations. This single test may be homogeneous like a tensile test or a biaxial test, or it can also be a nonhomogeneous test where the deformation map is measured through digital image correlation and the cell load is recorded. The resulting macromolecule behaviour fully characterizes the reversible behaviour of the polymer and can be used in an efficient manner in finite elements to perform accurate simulations of polymers.

Keywords: hyperelasticity, B-splines, machine learning, polymers

1 Introduction

The forces in a polymer are a result of a change of the entropy of the macromolecules (chains) [1]. This observation brought the classical statistical theory of polymers. This theory resulted in the Neo-Hookean model and in the appearance of the first invariant $I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$ of the right Green-Cauchy deformation tensor $\boldsymbol{C}$, where λ_i are the principal stretches. The Neo-Hookean statistical model results in a stored energy as

$$\Psi(I_1) = \frac{\mu}{2}(I_1 - 3) \tag{1}$$

where μ is the shear modulus. Since polymers are considered almost incompressible, we have $\lambda_1\lambda_2\lambda_3 = 1$, and for an in-plane test (any biaxial test, including uniaxial, equibiaxial and pure shear tests as particular cases), the first Piola (nominal) stress for a polymer is

$$P_i = \frac{\partial\Psi(\lambda_1,\lambda_2,\lambda_3)}{\partial\lambda_i} - \frac{\lambda_3}{\lambda_i}\frac{\partial\Psi(\lambda_1,\lambda_2,\lambda_3)}{\partial\lambda_3};\ \ i = 1{,}2 \tag{2}$$

It can be easily shown that the Neo-Hookean model is equivalent to a full network model in which each representative chain has the stored energy

$$\psi(\Lambda_{ch}) = \frac{3\mu}{2}(\Lambda_{ch} - 1)\ \ with\ \ \Lambda_{ch} = \boldsymbol{C}{:}(\boldsymbol{r}_{ch} \otimes \boldsymbol{r}_{ch}) \tag{3}$$

where $\boldsymbol{r}_{ch}$ is the direction of the chain at hand and $\Lambda_{ch} \equiv (\lambda_{ch}^{C})^2$ is the affine squared stretch of the chain. The total energy is obtained by integration in the microsphere Ω (i.e. integrating in all directions) as $\Psi = \frac{1}{\Omega}\int \psi d\Omega$. Then, by simply noting that

$$\frac{1}{\Omega}\int_{\Omega} \Lambda_{ch} d\Omega = \frac{I_1}{3} \tag{4}$$

we get the result in Eq. (1). Statistical models that account for the limited extensibility of chains, which include the inverse Langevin function, are written also in terms of the first invariant only, the same as the 8-chain (Arruda-Boyce) model. In this case, we can write a general form of the type $\Psi(I_1)$, and Eq. (2) is also obtained through the chain rule by

$$\frac{\partial\Psi(I_1(\lambda_1,\lambda_2,\lambda_3))}{\partial\lambda_i} = \frac{\partial\Psi(I_1)}{\partial I_1}\frac{\partial I_1}{\partial\lambda_i} \tag{5}$$

Remarkably, if we assume a stored energy of the form $\Psi(I_1)$, we can uniquely determine the behaviour of this polymer from a single test using our WYPiWYG (What-you-prescribe is what-you-get) approach [2]. That technique has been used to model even anisotropic materials [3, 4]. However, since Mooney's works, it has repeatedly noted that functions of the form $\Psi(I_1)$ fail to represent the polymer behaviour under a general deformation mode. Rivlin introduced the more general functional form $\Psi(I_1, I_2)$. Characterizing the general form $\Psi(I_1, I_2)$ from a full set of experiments is a quite simple task, because in essence, this is a simple problem of

determining two manifolds of two variables that fully represent the behaviour of the polymer under any deformation mode, namely $P_1(\lambda_1, \lambda_2)$ and $P_2(\lambda_1, \lambda_2)$. However, the amount of experimental data needed for this task is quite large, since different biaxial tests with different ratios would need to be performed, and the model gives no much more information than the raw experimental data. Indeed, model-free techniques may be used in this case.

The purpose of the rest of the paper is to show that, learning the proper state variable, from a single macroscopic test, as a tensile test or a nonhomogeneous test, the behaviour of the macromolecule can be determined and then used to predict the behaviour of the polymer in a finite element program under any deformation type.

2 Methods

The first ingredient of the proposed approach is to use the proper state variable. With the procedures used in this work, it is possible to determine that the micro-macro state variable link for determining the chain stretch, which in the classical statistical theory is $\Lambda_{ch} = \boldsymbol{C}: (\boldsymbol{r}_{ch} \otimes \boldsymbol{r}_{ch})$, and which resulted in the first invariant for phenomenological models, is not the best choice, please refer to [5]. Indeed, it can be shown and learned from experimental data and theoretical grounds that a much better proposal is $\lambda_{ch} = \boldsymbol{U}: (\boldsymbol{r}_{ch} \otimes \boldsymbol{r}_{ch})$, where $\boldsymbol{U} = +\sqrt{\boldsymbol{C}}$ is the right stretch tensor and is the chain stretch. If the statistical theory is applied with this micro-macro link, the "Neo-Hookean" model (note that in this case is not a Neo-Hookean model because the shear stress is not linear in terms of the amount of shear) would be –cf. Eq.(3)

$$\psi(\lambda_{ch}) = \frac{3\mu^*}{2}\left(\lambda_{ch}^2 - 1\right) \; with \; \lambda_{ch}^2 = \boldsymbol{U}: (\boldsymbol{r}_{ch} \otimes \boldsymbol{r}_{ch}) \tag{6}$$

Unfortunately, in this case the integral in the micro-sphere must be performed numerically, for example employing the Bazant-Oh integration rule. The terms in Eq. (2) are

$$\frac{\partial \Psi(\boldsymbol{U})}{\partial \lambda_i} = \frac{1}{\Omega}\int_{\Omega} \frac{\partial \psi\left(\lambda_{ch}(\boldsymbol{r}_{ch}, \boldsymbol{U})\right)}{\partial \lambda_{ch}(\boldsymbol{r}_{ch}, \boldsymbol{U})} \frac{\partial \lambda_{ch}(\boldsymbol{r}_{ch}, \boldsymbol{U})}{\partial \lambda_i} d\Omega \tag{7}$$

Remarkably, this model, using only one constant, namely μ^* (which value can be shown to be $\mu^* = 0.7\mu$), brings the correct nonvanishing slope in Mooney plots and the correct slope in the transverse axis in biaxial tests. The predictive power of this "Neo-Hookean" model for multiaxial loading, is far better than the classical Neo-Hookean model, despite also having a single constant with an equivalent physical meaning.

Motivated on this theory, one may propose a general chain function of the type $\psi(\lambda_{ch})$, to be later integrated numerically during finite element simulations, given the deformation gradient (in practice the principal stretches) at the material routine level. Machine Learning algorithms just need to learn this function from any single macroscopic test. We use B-splines as a representation basis for this function because derivatives are immediate; indeed, we determine the derivatives. If we assume that this function is known, we can write

$$P_{ch}(\lambda_{ch}) := \frac{\partial \psi(\lambda_{ch})}{\partial \lambda_{ch}} = \sum_{i=1}^{nv} N_i(\lambda_{ch}) \hat{P}_{chi} = [N_1(\lambda_{ch}), \dots, N_{nv}(\lambda_{ch})] \begin{bmatrix} \hat{P}_{ch1} \\ \vdots \\ \hat{P}_{chnv} \end{bmatrix} \quad (8)$$

where *nv* is the number of vertices in the B-spline, $\{\lambda_{chi}, \hat{P}_{chi}(\lambda_{chi})\}$ are the vertices to be determined and $N_i(\lambda_{ch})$ are the B-spline interpolation functions. Then, for a general biaxial test, after a little algebra, we get (see [7]) the following expression for the nominal stress in each *i=1,2* axis

$$P_i = \sum_{j=1}^{nq} \left(r_{chji}^2 - r_{chj3}^2 \frac{\lambda_3}{\lambda_1} \right) w_j [N_1(\lambda_{chj}, \dots, N_{nv}(\lambda_{chj})] \begin{bmatrix} \hat{P}_{ch1} \\ \vdots \\ \hat{P}_{chnv} \end{bmatrix} \quad (9)$$

where *nq* is the number of integration points in the sphere and w_j are the weights, and r_{chji} are the *i*-direction components of the chain at integration point *j*. Then, establishing a loss function as the error between the predicted P_i and the measured value for any given test, and using least squares, we can obtain the vertices $\{\lambda_{chi}, \hat{P}_{chi}(\lambda_{chi})\}$ of the B-spline representing the chain behaviour, by simply solving a linear system of equations, i.e.

$$\boldsymbol{A}\hat{\boldsymbol{P}}_{ch} = \boldsymbol{b} \quad (10)$$

where

$$\boldsymbol{A} = \bar{\boldsymbol{N}}^{\boldsymbol{T}} \boldsymbol{W} \bar{\boldsymbol{N}} + \boldsymbol{D}^{\boldsymbol{T}} \boldsymbol{\Omega} \boldsymbol{D} \;\; and \;\; \boldsymbol{b} = \bar{\boldsymbol{N}}^{\boldsymbol{T}} \boldsymbol{W} \boldsymbol{P} \quad (11)$$

and $\bar{\boldsymbol{N}}$ has components $\bar{N}_{i,j} = \sum_{j=1}^{nq} \left(r_{chji}^2 - r_{chj3}^2 \frac{\lambda_3}{\lambda_1} \right) w_j [N_1(\lambda_{chj}, \dots, N_{nv}(\lambda_{chj})]$ for the specific case of uniaxial test (other cases are similar). The matrix $\boldsymbol{W}$ is a diagonal matrix to assign different weights if needed to different parts of the domain. The terms $\boldsymbol{D}^{\boldsymbol{T}} \boldsymbol{\Omega} \boldsymbol{D}$ are smoothing terms to account for possible noisy experimental data. See [7] and [8] for further details. The procedure may be easily extended to nonhomogeneous tests, where the data needed is the map of strains obtained from digital image correlation and the force in the load cell. In this case, the algebra is more elaborate, see [8], but the concept remains as simple. Indeed, the problem is also solved through a linear system of equations which has the same form as in Eq. (11), but which coefficients account also for the finite element formulation. See [8] for further details.

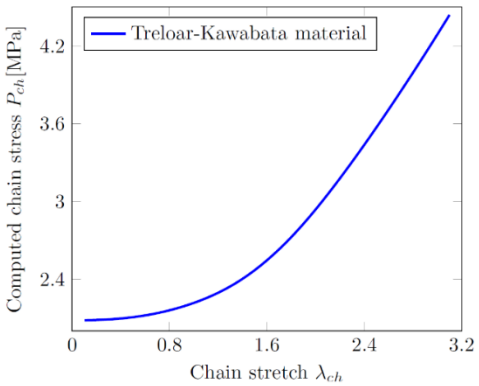

Figure 1: Reverse-engineered representative polymer chain behavior for Treloar-Kawabata material

3 Results

We have applied the above-mentioned data-driven procedure to predict the behaviour of the widely-known Treloar polymer, which is almost identical to the behaviour of the Kawabata et al polymer; experimental multiaxial data is given in [9]. We use any single test curve from Treloar or Kawabata's experiments to obtain the chain behaviour $P_{ch}(\lambda_{ch})$ given in Eq. (8). This behaviour, using a Kawabata test is depicted in Fig. 1. With this reverse-engineered behaviour, we can perform predictions for the biaxial tests from Kawabata [9]. Figure 2 shows the excellent predictive power of the method.

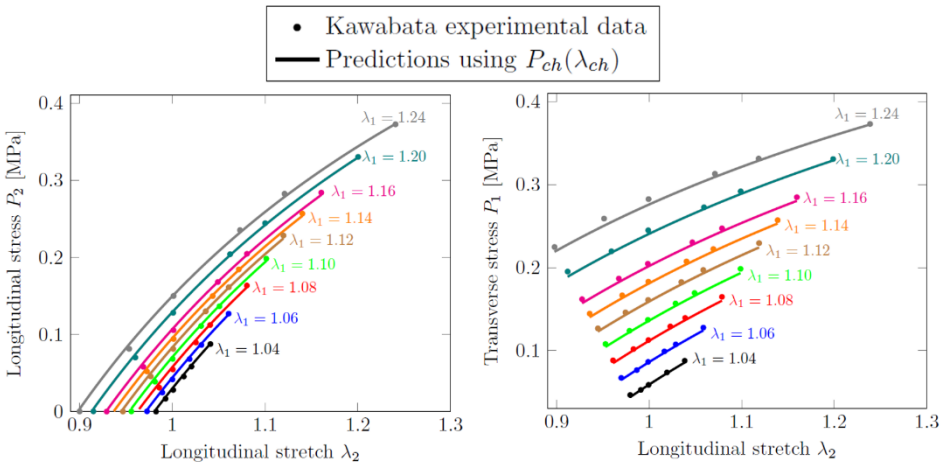

Figure 2: Predicted biaxial experiments for the Kawabata et al material using the reverse-engineered chain behaviour of Figure 1.

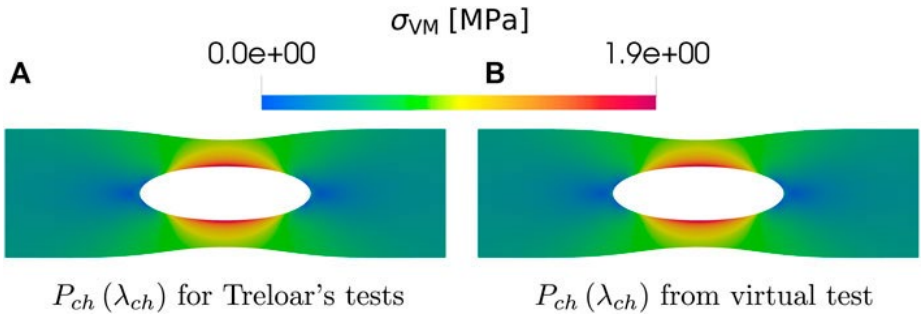

Figure 3: Simulation of a perforated plate of Treloar's material with $P_{ch}(\lambda_{ch})$ obtained either from Treloar's homogeneous test or reverse-engineered from the nonhomogeneous experiments. Details for this simulation are given in Ref. [8]

The same approach may be applied to a non-homogeneous experiment. In this case, we have performed a virtual test using the previous $P_{ch}(\lambda_{ch})$. Then, from that simulation we reversed engineered the $P_{ch}(\lambda_{ch})$ and repeated the simulation with the new $P_{ch}(\lambda_{ch})$. The comparison of both simulation is reproduced in Figure 3. It is shown that no appreciable difference is obtained; see details in Ref. [8].

4 Conclusions and Contributions

In this work we present a novel non-parametric data-driven formulation to accurately model the polymer behaviour under 3D loading. The procedure is based on B-spline representation of the behaviour of the representative macromolecule, which is obtained from a single load-displacement curve any macroscopic homogeneous experiment. Alternatively, it can also be obtained from the hon-homogeneous deformation map and the load-displacement curve obtained from digital image correlation of any specimen. Remarkably, we simply solve a linear system of equations in any case to obtain the characteristic chain behaviour.

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