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A PHYSICS-INFORMED NEURAL NETWORK CONSTITUTIVE MODEL FOR CROSS-LINKED POLYMERS

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ABSTRACT

The behavior of Cross-linked Polymers in finite deformations is often characterized by nonlinear behaviour. In this paper, we propose to embed an artificial neural network (ANN) within a micro-mechanical platform and thus to enforce certain physical restrictions of an amorphous network such as directional dependency and history-dependency of the constitutive behavior of rubber-like materials during loading and unloading. Accordingly, a strain energy density function is assumed for a set of chains in each direction based on ANN and trained with experimental data set. Summation of the energies provided by ANNs in different directions can determine the strain energy density function of the matrix. Stress-strain relation is derived from strain energy density function. Polyconvexity is enforced to assure minimized potential energy, a global solution for an optimization problem, and thermodynamic consistency that show the model cannot generate energy. The model is validated against multiple sets of experimental data, e.g. uniaxial, shear, and biaxial deformation available in the literature. This model captures not only the loading and unloading paths but also the inelastic response of these materials, such as the Mullins effect and permanent set. The model can be generalized to other materials and other inelastic effects as well.

INTRODUCTION

Elastomers are widely meshed and cross-linked polymers that evince entropically elastic behavior and do not show reversible deformation. Their constitutive model plays a significant role in the determination of their mechanical behavior. Based on how these models are derived, elastomer models are categorized into phenomenological and micro-mechanical models [1]. Each of these categories comes with an inherent set of pros and cons. The first one is less complex, and usually, it is based on preset functions of strain invariants or principal stretches while disregarding the micro-structure of elastomers. On the contrary, micro-mechanical models are complex, but they consider the physical interpretation of elastomers. Many theoretical and experimental studies have been conducted on mechanical properties of them [2–4].

Among a large number of phenomenological models, few of the most formidable ones are introduces in the following. Og-

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den [5], in 1972, proposed a model to provide an adequate representation of the mechanical response of rubber-like materials. This model is simple and amenable to mathematical analysis. This is a stretch based model which ensures strain energy formulation as the summation of scaled power of principal stretch. Hereon, all mentioned phenomenological models in this paper are invariant based. In 1940, Mooney-Rivlin [6] model was proposed. This model is the summation of first and second invariant with their coefficients taken into consideration. In 1943, Neo-Hookean [7] model was proposed that is relatively simpler than Mooney-Rivlin model because it just considers first invariant. In order to add a curvature shape to last models that could not capture the "S" shape behavior of elastomers, Isihara [8] added the square of first invariant to Mooney-Rivlin model. This nonlinear term captures the data better than the mentioned models. After seven years, Gent and Thomas [9] proposed another model based on the summation of first invariant and logarithm of second invariant. This model, in fitting, is identical to Neo-Hookean model results. Caroll [10], in 2011, proposed a three-parameter strain energy function based on first and second invariants. This model shows an acceptable fitting but not as well as Swanson's model. In 2016, Zhao [11] proposed a model by deriving a partial differential equation that considers the balance between stored energy and stress work. As a result, the non-negligible problem of phenomenological models is that they are invariant based, and with a set of material parameters, they cannot describe some important mechanical behavior, such as softening, hardening, and deformation state dependency. In this state, we can see some advantages of micro-mechanical models compared to another category.

Micro-mechanical approaches are usually based on the deformation behavior of a single chain and the expansion of a certain number of chains in different directions. So, they can be directional dependent. Each chain consists of some rigid beams, and its movement in space is based on the random walk assumption Fig. 1.

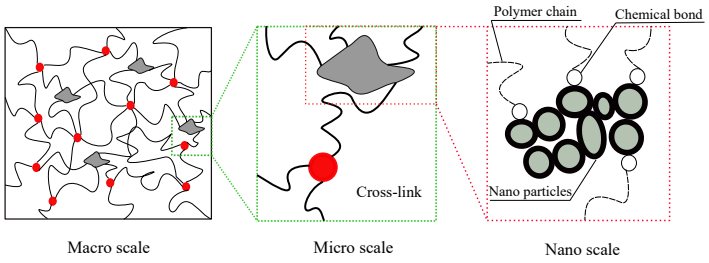

FIGURE 1. Schematic illustration for microstructure of filled elastomers which is depicted micro scale and nano scale

Three chain model [12] was proposed in 1943. It considers three chains along with eigenvectors of isochoric right Cauchy-Green tensor and create a free energy function for elastomers. Like three chain model, Arruda and Boyce proposed eight chain model [13]. The only difference is that there are eight chains along with half diagonals of a cube. In 2002, Miehe et al. [14] proposed a unit sphere model that chains are along with radius vectors from the center. These chains have a uniform distribution over the sphere. The complexity of micro-mechanical models is because of integration over the sphere. Although we simplify it with converting this integration to a summation, higher computational cost remains yet. Farhangi et al. [15, 16] investigated the effect of fiber. Hossain [7] showed in his paper that none of these models could predict different deformation states because they are directional based. Some researchers have reformed sphere model to solve these challenges. Miehe et al. [17] proposed a model, in 2004, that is non-affine micro-mechanical model to solve one of the challenges. They considered the motion of chains with assuming the contribution of topology constraints in their motion. In 2016, Khiem and Itskov [18] proposed a model based on network-averaging of the tube model. In 2002, Marckman et al. [19] proposed network alteration theory to show Mullins effect. They added damage to improve eight chain model in order to capture softening in rubbers. In 2013, Dargazany et al. [20] proposed a network evolution model in order to see anisotropic Mullins effect and permanent set. Their model is based on inverse Langevin function [21]. Mohammadi et al. [22,23] add thermo aging to this model, and Bahroululoumi et al. [24,25] added hydrolytic aging to the model. The advantage of micro-mechanical based models is that they can show inelastic behavior in materials. The recent rise of machine learning as a powerful technique for data analysis is impressive and practical [26, 27]. But, it does not work without combination with laws of physics. Some valuable researches have been conducted on the modeling of behavior of materials based on experimental data. Shojaeifard et al. [28,29] proposed a viscoelastic model and its framework in FEM. In 2004, Shen and Chandrashekhara [30] proposed a neural network based model for constitutive modeling. They showed that their results were acceptable compared to ABAQUS results. Oritz et al. [31], in 2016, proposed a data-driven model for elastic materials. They designed a constraint optimization problem while considering conservation laws and material laws. In 2019 [32], They extended their model to material history dependent. Their work is based on their last idea. However, they added time to their model to capture the effect of history in behavior of inelastic materials. A neural network based for the prediction of nonlinear viscoelastic materials was proposed by Al-Haik et al. [33]. They predicted stress relaxation of polymer matrix composites. Hashash et al. [34] by using a neural network constitutive model, to bypass stiffness matrix concept in finite element. However, they did not consider material laws and loading-unloading paths in their training.

This paper is organized as follows; in section 2, the main concepts of damage in constitutive model are introduced and de-

scribed in detail. Energy function estimation based on ANN, is presented in section 3. Section 4 explains the idea and formulation of the proposed model in detail. Model verification with experimental data is discussed in section 5. On resume, section 6 provides some concluding remarks and outlines some perspectives.

Damage in Elastomers

Elastomers exhibit changes in their mechanical behavior after first extension known as Mullins effect, which was first investigated by Mullins and his colleagues [35]. This phenomenon happens to both types of filled and unfilled elastomers. To provide a better micro-mechanical understanding of the damage resulting from Mullins effect, Fig. 2 shows some of the proposed physical interpretations of this effect in the literature i.e. chain breakage [36] in the filler interface, chain disentanglement [37], molecules slipping [38] and rupture in cluster of fillers [39]. After applying loading and unloading to filled elastomers, due to anisotropic damage in the material, a residual strain remains in the material known as permanent set. However, the permanent set in unfilled rubber is negligible.

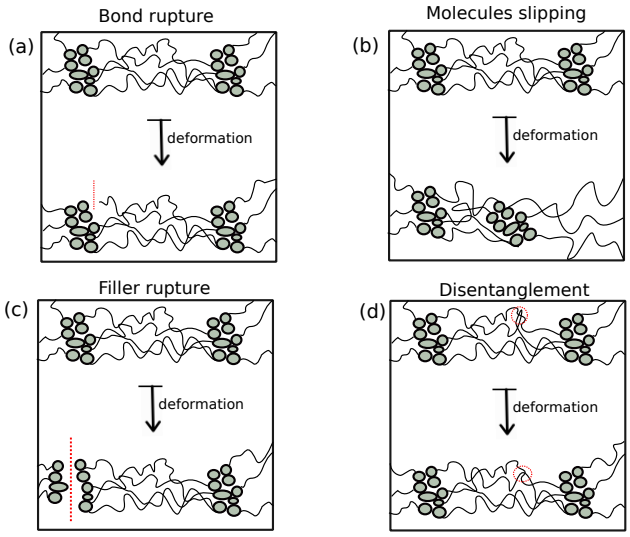

FIGURE 2. Physical explanation of Mullins effect

Energy Function Estimation

One of the concepts used in the statistical mechanics of soft material is the assumption of uniform distribution of polymer chain in diffident directions, which can be taken into account by averaging the response of the material over a micro-sphere. In these models, unit sphere act as a bridge between macro scale behavior of the material and micro scale (see Fig. 3). This approach can transfer information from micro structure behavior to the macroscopic behavior via homogenization over the unit sphere. In micro-sphere approach, the average response of the material over the sphere can be numerically calculated by n integration directions $[d_i]_{i=1...n}$ that are weighted by factors $[w_i]_{i=1...n}$. On the other hand, for cross-linked polymers, researchers use strain energy density function (Ψ) in terms of the deformation gradient to obtain stress by taking the derivative of strain energy. Hence, strain energy function over the sphere can be approximated by

$$\Psi = \frac{1}{4\pi}\int_S \psi^d dS^d \cong \sum_{i=1}^{n} w_i {\psi_i}^{d_i}, \tag{1}$$

which ${\psi_i}^{d_i}$ is strain energy in direction d_i, and 4π is normalization factor.

Proposed model

In this study, two parallel ANNs are considered as the strain energy of a set of available chains in each direction. The utilization of ANN concept for energy function makes it to be considered as a phenomenological approach. It adds advantages of phenomenological models to proposed model, which is mentioned in the introduction section. Subsequently, the summation of energies provided by ANNs in different directions of micro-sphere results in the strain energy function of the whole matrix. This assumption adds to the advantages of micro-mechanical category to our model. Fig. 3 shows a schematic design for the concept of this novelty in this research. In order to consider damage and history dependency of inelastic behavior of rubbers, we designed the proposed ANN model based on this. For the first ANN of the chains, one input is λ_{max}, one input is λ, and one input is one in order to remove over-fitting and bias problems in ANN training. Output of first ANN is $\overset{\vec{d}_i}{\psi_1}$ which $\vec{d}_i$ shows the direction of each chain in micro-sphere. For the second ANN of the chains, we have one as input for over-fitting challenge, β_{max} as input, and β as another input. Output of second ANN is $\overset{\vec{d}_i}{\psi_2}$. So, summation of $\overset{\vec{d}_i}{\psi_1}$ and $\overset{\vec{d}_i}{\psi_2}$ is the strain energy of a single chain.

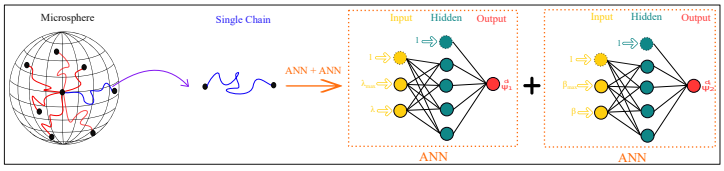

FIGURE 3. Schematic of proposed model for a microsphere

$\lambda^{\vec{d}_i}$ and $\beta^{\vec{d}_i}$ are defined as:

$$\lambda^{\vec{d}_i} = \sqrt{\vec{d}_i \mathbf{C} \vec{d}_i}, \qquad \beta^{\vec{d}_i} = \sqrt{\vec{d}_i \mathbf{C}^{-1} \vec{d}_i}, \tag{2}$$

which $\mathbf{C} = \mathbf{F}^T\mathbf{F}$ is right Cauchy-Green deformation tensor and $\mathbf{F}$ is deformation gradient tensor. We can write the total strain energy of microsphere as

$$\Psi = \Psi_1 + \Psi_2 = \frac{1}{A_s}\int_S \left({\psi_1}^d + {\psi_2}^d\right) d^d u \cong \sum_{i=1}^{k} \left({\psi_1}^{d_i} + {\psi_2}^{d_i}\right) w_i, \tag{3}$$

which A_s is the surface area of microsphere and w_i is weight factor corresponding to directions d_i. The strain energy of a single chain can be calculated as

$$\psi^d = {\psi_1}^d + {\psi_2}^d = ANN_1(\mathbf{W_1}, \lambda^{\vec{d}_i}, {\lambda_{max}}^{\vec{d}_i}) + ANN_2(\mathbf{W_2}, \beta^{\vec{d}_i}, {\beta_{max}}^{\vec{d}_i}), \tag{4}$$

which $\mathbf{W_1}$ and $\mathbf{W_2}$ are weight matrix of first and second ANN respectively. Moreover, the first Piola-Kirchhoff stress tensor $\mathbf{P}$ can be written as

$$\mathbf{P} = \frac{\partial \Psi}{\partial \mathbf{F}} - p\mathbf{F}^{-T} = \frac{\partial \Psi_1}{\partial \mathbf{F}} + \frac{\partial \Psi_2}{\partial \mathbf{F}} - p\mathbf{F}^{-T}. \tag{5}$$

For incompressible elastomers, $\det\mathbf{F} = 1$ and p denotes an arbitrary scalar parameter to guarantee incompressibility. We can express mentioned derivation as

$$\frac{\partial \Psi_1}{\partial \mathbf{F}} = \sum_{i=1}^{k} w_i \frac{\partial {\psi_1}^{d_i}}{\partial \lambda^{d_i}} \frac{\partial \lambda^{d_i}}{\partial \mathbf{F}}, \qquad \frac{\partial \Psi_2}{\partial \mathbf{F}} = \sum_{i=1}^{k} w_i \frac{\partial {\psi_2}^{d_i}}{\partial \beta^{d_i}} \frac{\partial \beta^{d_i}}{\partial \mathbf{F}}. \tag{6}$$

From now on, the only challenge is the design and training of two ANNs, which show the strain energy of a single chain by using the experimental data set. Depending on the complexity of the problem, a multi-layered ANN can be defined for a single chain. In this study, we used a costume structured feed-forward ANN with one hidden layer and four neurons, which is coded by our self, not employing any packages.

Verifying Proposed Model with Experimental Data

ANN models for materials, usually, are trained by stress-stretch experimental data set. During training, ANN model approximates the relation between stress and stretch. Unlike mathematical constitutive models, ANN constitutive models can be continuously retrained to adapt to new experimental data set as they get generated from new experiments. Besides, the selection of stress-stretch data set plays a vital role in the success of the model. This data set for training should cover the range for which this model will be used. Hence, in order to train our model with the data set, we need to define an error to minimize the difference between the approximated model and experimental data to calculate the weight matrix. The only difference with a simple (1D) ANN training is that here, we have the summation of 21 ANNs in 21 different directions such that each ANN in each direction has two parallel ANNs. The output of all of ANNs in different directions is strain energy that we should consider for training with our experimental data set. We can express the error as

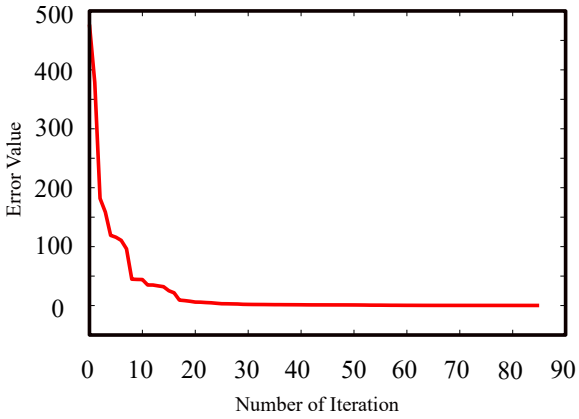

FIGURE 4. Convergency in the training of proposed ANN model with experimental data set

$$E(\mathbf{W}) = \frac{1}{2}\sum_j [\left(\sum_{i=1}^{k} w_i \frac{\partial {\psi_1}^{d_i}}{\partial \lambda^{d_i}} \frac{\partial \lambda^{d_i}}{\partial \mathbf{F}} + \sum_{i=1}^{k} w_i \frac{\partial {\psi_2}^{d_i}}{\partial \beta^{d_i}} \frac{\partial \beta^{d_i}}{\partial \mathbf{F}} - p\mathbf{F}^{-T}\right)_j - P_j]^2, \tag{7}$$

that i shows the number of directions in a microsphere, j shows the number of data in a set, $\mathbf{W}$ is weight matrix, and P_j is experimental stress j in data set. The training of the proposed model is conducted by a set of experimental data available in the literature [40]. We trained the proposed model by bi-axial data and predicted uni-axial extension and pure shear. Also, in order to guarantee that the strain energy function is polyconvex and ensure thermodynamics consistency, the weights that connect the input of λ and β to other neurons should be positive. In contrast, the weights that connect λ_{max} and β_{max} to other neurons should be negative. So, we have a constraint optimization problem to solve for training our model subjected to weights related to λ_{max} and $\beta_{max} \leq 0$; and weights related to λ and $\beta \geq 0$

Fig. 4 shows the error to the number of iterations after training.

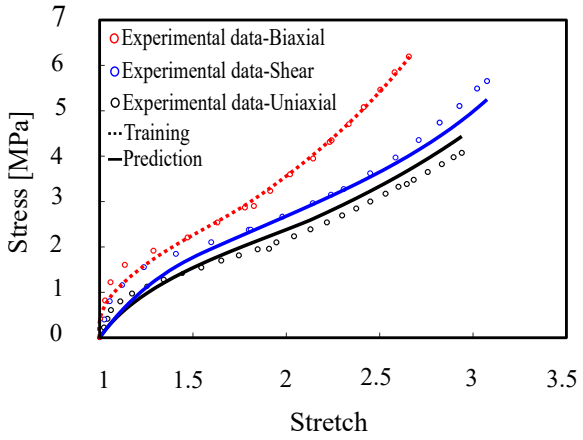

FIGURE 5. Model training and prediction of uni-axial, bi-axial and shear [40]

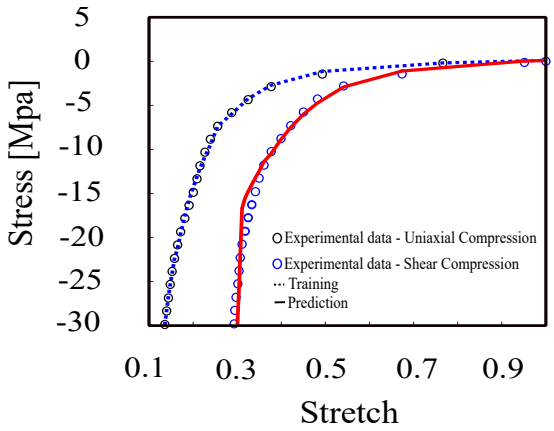

FIGURE 6. Model training with uni-axial compression and prediction of shear compression [41]

Loading Tension Response

In order to show the model can capture hyper-elastic behavior in different states of deformation, we use two experimental data sets from literature. In both of them, we train the model with bi-axial tension and predict uni-axial tension and pure shear. The only difference in ANN during loading deformation is that λ_{max} and β_{max} are equal to λ and β respectively. This limitation of the data leads to a model with only two inputs. Fig. 5 shows the results, respectively, for each data set. The results show the excellent performance of the proposed model for loading under different states of deformation.

Compression Response

Compression behavior of rubber like material is another one of their aspects that plays an essential role in industrial application. In order to show that the proposed model can train and predict their behavior, in this subsection, we trained data set of uni-axial compression experiments and predicted the behavior of shear compression. Fig. 6 shows the performance of the proposed model for compression tests.

Loading and Unloading Response in uni-axial Tension

The large strain behavior of filled rubbers is characterized by the strong Mullins effect, permanent set. Prediction of these inelastic features in elastomers is an important challenge with immense industrial and technological relevance. To consider that, unlike the last part, in this part, we consider λ_{max} and β_{max} as input in ANN model because modeling of inelastic effects is dependent on the history of material. On the other hand, training and fitting play a crucial role in the set of parameters in different constitutive models. Due to the simplicity of uni-axial extension test, usually, data of this state of deformation is available. Elastomerov et al. [42] showed in their paper that using one particular mode of deformation is not sufficient for fitting. Hereunder, Fig. 7 shows Mullins effect and permanent set for two data sets of uni-axial extension. We show that with the training of uni-axial data, we not only can interpolate Mullins effect but also extrapolate it during uni-axial deformation.

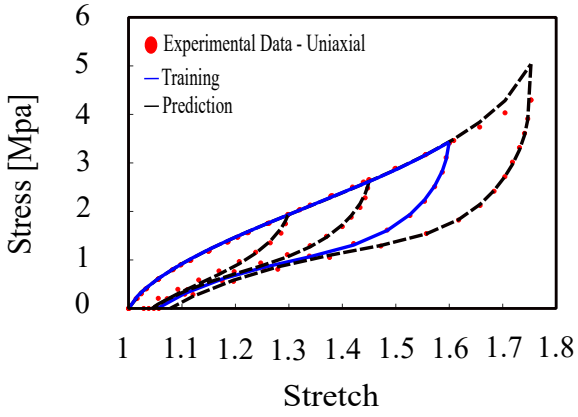

FIGURE 7. Model training and prediction of cyclic uni-axial tension with step-wise increasing of amplitude [43]

CONCLUSION

The motivation behind this work is the absence of a data-driven model that can describe the inelastic behavior of elastomeric materials for various states of deformation. In this study, we have proposed an ANN based constitutive model, which is a combination of the phenomenological and micro-mechanical categories to capture the inelastic behavior of elastomers. Stress-stretch experimental data sets used to train the ANN based constitutive model. This model has the potential to describe the behavior of any elastomeric materials along with their inelastic behavior, such as Mullins effect and permanent set. The model has been tested for various elastomers in different loading conditions (uni-axial extension and compression, bi-axial, pure shear, shear compression) and shows excellent performance to reveal inelastic behavior of elastomers. The model was trained by only a data set of a specific sate of deformation and predicted all other experimental curves. The ANN model has some weight constraints to guarantee polyconvexity and thermodynamics consistency that are necessary for proposing a constitutive model.

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