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A PHYSICS-BASED DATA-DRIVEN APPROACH FOR MODELING OF ENVIRONMENTAL DEGRADATION IN ELASTOMERS

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ABSTRACT

Elastomers are now commonly used in a number of industries, including aerospace, structure, transportation, shipbuilding, and automotive, due to their excellent workability, formability, and flexibility. During their activity, elastomers are subjected to harsh environmental conditions, which decreases their resilience. False predictions made early in their lives can have major financial and environmental implications. Elastomers' performance and properties, such as strength, durability, and density, are influenced by chemical changes in these materials, known as degradation, which occurs over time. This process can alter the morphology of a polymer matrix as well as cause chain scission and cross-linking, resulting in different behaviors than that of the unaged material. To demonstrate the effect of thermal-oxidative aging on the mechanical behavior of elastomers, several experimental and theoretical models have been proposed. In view of the large volume of experimental data available on micro-structural evolution in the course of aging, we propose a physics-based data-driven approach to overcome the shortcomings of both phenomenological and micro-mechanical models. This work presents a novel thermodynamically consistent, multi-agent machine-learned model for predicting the constitutive behavior of cross-linked elastomers during environmental aging, such as thermo-oxidative and hydrolytic aging for various states of deformation. Single mechanism degradation changes the polymer matrix over time where it is causing chain scission, reduction of cross-links, and morphology change. To capture the idealized Mullins effect and permanent set due to the effect of single aging mechanisms on nonlinear mechanical responses of elastomers, we propose a data-driven model for simulating inelastic elements in a polymer matrix. By using a sequential order reduction, we were able to reduce the 3D stress-strain tensor mapping problem to a small number of super-constrained 1D mapping problems. To systematically classify such mapping problems into a few categories, an assembly of multiple replicated conditional neural network learning agents (L-agents) is used based on our recent work. Each category is represented by a different type of agent. The effect of deformation history, aging time, and aging temperature is captured by this model. The model is validated using a broad collection of data, ranging from our experimental results to data from the literature. In addition, thermodynamic consistency and frame independence are investigated. The most significant achievements of this model are its precision, simplicity, and prediction of inelasticity under various states of deformation. The model's accuracy and simplicity make it a good option for commercial and industrial applications. Conveniently, due to the model modular nature, it can be expanded in the future to include viscoelasticity and non-isotropic formation for better precision.

INTRODUCTION

Nowadays, cross-linked elastomers, due to excellent workability, form-ability, and versatility, play a significant role in several industries such as aerospace, structure, transportation,

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shipbuilding, and automotive [1–3]. Cross-linked elastomers encounter aggressive environmental conditions during their operation, which affects their durability [4–6]. Overestimation of their life-time can have high financial and environmental costs. Chemical changes in these materials are known as degradation, that occurs during a period referred as aging. Degradation affects cross-linked elastomers' performance and properties, including their strength, toughness, and density.

In theoretical approaches, researchers usually combine hyper-elastic constitutive models, which describe the hyperelastic behavior of materials irrespective of the effects of aging time and temperature, with degradation models, which demonstrate the decay of materials during aging. Hyper-elastic models fall into three main categorizes: the phenomenological approach, the micro-mechanical approach, and the data-driven approach. In the first one, a mathematical model is considered without any physical interpretation, and model parameters are fitted by experimental data set. These phenomenological models, with a set of material parameters, do not employ the micro-physics behind materials [7]. The micro-mechanical approach is based on the statistical mechanics of polymers' structure and employs physical meaning during analysis. Thus, some of them can show inelastic behaviors of cross-linked elastomers, such as the Mullins effect and permanent set. Three-chain model [8], eight-chain model [9], full network model [10], extended tube model [11], non-affine micro-sphere model [12], network alteration [13], and network evolution model [14] are the most famous models in this category. Micro-mechanical models, with functions and material parameters related to micro-structure, consider micro-structural information. However, given the complexity of these models, their practical application is limited. The data-driven approach tries to address the limitations of both phenomenological and micro-mechanical models by obtaining micro-structural behavior from the macroscopic experimental data set. Montans et al. [15] proposed an inverse engineering data-driven model that captures hyper-elastic behavior of materials by solving a piece-wise set of equations. Oritz et al. [16] proposed a data-driven model by designing an optimization problem subject to constraints related to conservation and physical laws. In addition, we recently proposed a neural network approach by the concept of network decomposition, which not only predicts different states of deformation but also indicates the inelastic behavior of cross-linked elastomers [17–19].

In recent decades, the phenomenon of degradation has attracted wide attention; Scholars have proposed different mathematical degradation models based on their applications. Arrhenius model [20], Archard model [21], and Paris model [22] fall in single mechanism degradation, which is continuous and without any fluctuation. Several models have been proposed from a combination of constitutive models and degradation models. Ha-Anh and Vu-Khanh [23] employed an Arrhenius function and combined it with the Mooney-Rivilin model to predict the hyperelastic behavior of aged polychloprene. Lion et al. [24] proposed a phenomenological model by splitting Helmholtz free energy into three parts. In 2013, Johlitz [25] proposed a phenomenological model that considers both mechanical and chemical degradation. Meanwhile, in the micro-mechanics category, Mohammadi et al. [4, 26] proposed a model for thermo-oxidative aging that combined the Author's network evolution model with the Arrhenius function and different decay rates. This study [27] investigated the geometry dependency of aging between substrate and adhesive by employing finite element simulation based on chemo-mechanical modeling. There are a few models that theoretically investigate hydrolysis [28]. Viera et al. [29] proposed a model for hydrolysis using Bergstrom and Boyce's constitutive model by decomposing mechanical behavior into time-dependent and time-independent parts. In another study, the author investigated the mechanical behavior of biodegradable materials during hydrolysis aging by employing a quasi-linear viscoelastic model [30]. In another micromechanical study, Bahrololoumi et al. [31,32] proposed a model for hydrolysis that combined Author's network evolution model and Arrhenius decay function. All the mentioned models have several advantages and disadvantages.

Phenomenological approaches are empirical, simple, and less interpretable; however, micromechanical approaches are highly interpretable but complex because they consider the readjustment of kinks, the rearrangement of convolutions, reorientation, and the uncoiling of molecular chains. Meanwhile, the emergence of machine-learned (ML) models has attracted much attention as a way to address the mentioned challenges of the phenomenological and micromechanical approaches.

The main objective of this work is to propose a simple data-driven model to not only predict thermo-oxidative aging and hydrolysis but also can show inelasticity which does not show a fixed trend of deformation versus applied force. The proposed model is based on the concept of a cooperative multi-agent system $\mathscr{A}_j^i$, $i \in \{1,n\}$, $j \in \{1,m\}$ to describe different features in the material behaviour with $n \times m$ different Neural Network learning agents (L-agents), which is responsible for learning from experimental data sets. We have simplified the 3D stress-strain tensor mapping problem into a small number of super-constrained 1D mapping problems by means of a sequential order reduction. We assume that during aging, the polymer matrix is changed by degradation, and due to the lack of exact knowledge on the behavior and interaction of polymer microstructures, we model it using an L-agent system. The model is validated by our set of experimental data. In addition, relaxation and intermittent experimental data available in the literature are used to show the proposed model's accuracy in different conditions.

Model and Method

Current phenomenological, micromechanical, and data-driven methodologies for demonstrating stress-strain recorded in cross-linked elastomers experience the effects of the absence of information on 3D structures. Then, we are only ready to quantify the strain recorded of the straightforward structure by utilizing digital image correlation (DIC) techniques.

- Continuum Mechanics In the first step, we use the 3D mapping of second order stress/strain tensors from continuum mechanics understanding. Here, by introducing strain energy Ψ_m as the middle agent in mapping, where $\mathbf{F} \rightarrow \Psi_m \rightarrow \mathbf{P}$, we can use Finite strain theory to simplify $\mathbf{F} \rightarrow \mathbf{P}$ mapping because it needs complex fourth order tensor mapping in hyperelastic materials. Accordingly, strain energy can be replaced as the part of the framework which needs to derive stress tensor $\mathbf{P}(\mathbf{F})$.

One specific benefit of using Ψ_m as the middle agent is that it guarantees the material objectivity and thermodynamic consistency on all the derived constitutive models (see Truesdell et al. [33]). There are several conditions on strain energy, which must be authorized in a data-driven model to be specific

$$\begin{aligned} \Psi_m(\mathbf{F}) \geq 0 \quad & when \quad \mathbf{F} \neq 0 && Increase\ energy, \\ \Psi_m(\mathbf{F}) = 0 \quad & when \quad \mathbf{F} = \mathbf{I} && Normalization, \\ \Psi_m(\mathbf{F}) \rightarrow \infty \quad & when \quad det\mathbf{F} \rightarrow \infty / 0 && Growth\ condition. \end{aligned} \tag{1}$$

Meanwhile, the ellipticity constraint is a significant challenge for hyperelastic materials. In the absence of traction forces, it can be applied by using strain energy. **So, in our model, the first constraint we implement is to compel agents to infer $\Psi_m(\mathbf{F})$ so that it fulfills Eq. 1 and polyconvexity condition.**

- Microsphere In the second step, we implement a concept from polymer physics for the cross-linked amorphous network. For isotropic polymers, polymer chains are considered uniformly distributed in every single spatial direction at the virgin state. This homogenized spatial arrangement lets us use the microsphere concept, which helps one to represent the 3D matrix as a homogeneous assembly of 1D elements spread over a microsphere in different spatial directions. This methodology can move knowledge from super-simplified 1D components to create a complex 3D polymer matrix by means of homogenization over the unitsphere. Besides by discretizing the sphere into limited areas, the integration can be taken out mathematically over N_d integration directions $[d_i]_{i=1...N_d}$ with various weight factors $[w_i]_{i=1...n}$ [34]. The strain energy of polymer matrix Ψ_m in relation to its elements can therefore be written as

$$\Psi_m = \frac{1}{4\pi} \int_S {\Psi_m}^d dS^d \cong \sum_{i=1}^{N_d} w_i {\Psi_m}^{d_i}, \qquad where \qquad {\Psi_m}^{d_i} = \mathscr{B}^{d_i} \tag{2}$$

where ${\Psi_m}^{d_i}$ is the sub-matrix element energy in direction d_i represented by one L-agent team $\mathscr{B}^{d_i}$ reflecting an additive cooperation between multiple L-agents $\mathscr{A}^i_\bullet$. Eq2 represents the integral $S(\theta,\phi) = \int_0^\theta \int_0^\phi \sin(\theta) d\theta d\phi$ with the unit vector $r = \sin(\theta)\cos(\phi)e_x + \sin(\theta)\sin(\phi)e_y + \cos(\phi)e_z$ over the unitsphere. Assuming an identical team in all directions in the virgin state, namely $\mathscr{B}^{d_i} = \mathscr{B}^{d_j}$, initial isotropy is guaranteed, but due to different loading in different directions, the material may easily become anisotropic. In addition, because L-agents respond in each direction to varying loading, the model may consider the onset of damage, deterioration, and propagation of cascading failure in directionally sensitive materials.

- Network Decomposition Network decomposition is the third concept we are using from statistical mechanics which helps us by infusing simple patterns on top of each other to predict complex patterns. This concept represents energy of an element ${\Psi_m}^{d_i}$ by superposing the energy of multiple sub-elements, ${\Psi_m}^{d_i} = \sum_{j=1}^{N_s} {\Psi_j}^{d_i}$, where each sub-element is responsible for a single inelastic feature. A team of cooperative L-agents $\mathscr{B}^{d_i} = [\mathscr{A}^i_j]$ will calculate the energy of one element, representing each sub-element by one L-agent, and then repeat this cooperative team in different directions to provide us with the matrix energy. Consequently, by substituting Eq. (2) we can extract the energy of the matrix directly with regard to sub-elements and the L-agents as given here that serve them.

$$\begin{aligned} \Psi_m &= \frac{1}{4\pi} \int_S {\Psi_m}^d dS^d \cong \sum_{i=1}^{N_d} \sum_{j=1}^{N_s} w_i {\psi_j}^{d_i} \\ \Psi_m &\approx \sum_{i=1}^{N_d} \sum_{j=1}^{N_s} w_i \mathscr{A}^i_j \qquad where \qquad {\Psi_m}^{d_i} = \sum_{j=1}^{N_s} \mathscr{A}^i_j. \end{aligned} \tag{3}$$

where, for each element, *Ns* is the number of sub-elements considered. For each sub-elements, super-simplified scalar-to-scalar mapping behavior has been derived and can be represented by a simplified feed-forward L-agent neural network. The first Piola—Kirchhoff stress tensor $\mathbf{P}$ can be derived on the basis of Eq. 3, as

$$\mathbf{P} = \frac{\partial \Psi_m}{\partial \mathbf{F}} - p\mathbf{F}^{-T} = \sum_{i=1}^{N_d} \sum_{j=1}^{N_s} w_i \frac{\partial \mathscr{A}^i_j}{\partial \mathbf{F}} - p\mathbf{F}^{-T}, \tag{4}$$

where p signifies the Lagrange multiplier to ensure incompressibility of the material.

Validation

Here, the model was validated by predicting the inelastic behavior of cross-linked elastomers for different states of deformation in two cases; (i) thermo-oxidative aging and (ii) hydrolysis. Several experimental data sets from relaxation tests and intermittent tests were used. Data sets were designed to capture the effect of (i) deformation, (ii) deformation history, (iii) aging time (t) and (iv) aging temperature (θ). In both of the case studies, we represented the matrix by 21 teams, each with two agents, which is a very small number (21 integration-points [34]). Note that the number of teams and their related agents can be chosen based on the trade-off between accuracy and computational cost.

The inputs and internal parameters of L-agents were built via λ_{j-max} parameters to capture the deformation of the rubbers with full memory. In order to allow teams to predict various deformation states, the first and second deformation invariants were supplied to each team [35]. The condition was satisfied by providing input set $S_1^{d_i} = [{\lambda_1}^{d_i}; \lambda_{1-max}^{d_i}, t, \theta]$ to L-agent 1 and $S_2^{d_i} = [{\lambda_2}^{d_i}; \lambda_{2-max}^{d_i}, t, \theta]$ to L-agent 2

$$ {\lambda_1}^{d_i} = \sqrt{d_i \mathbf{C} d_i}, \qquad {\lambda_2}^{d_i} = \sqrt{d_i \mathbf{C}^{-1} d_i}, \qquad \mathbf{C} = \mathbf{F}^T \mathbf{F} \tag{5} $$

where ${\lambda_1}^{d_i}$ and ${\lambda_2}^{d_i}$ were intended to lead to the first and second sub-elements and to reflect, respectively, I_1 and I_2. For the ANN structure of L-agents, we considered one input layer, one hidden layer with four neurons and three activation functions, softplus ($\psi(\bullet) = ln(1+e^{\bullet})$), sinusoid ($\psi(\bullet) = sin(\bullet)$) and hyperbolic tangent ($\psi(\bullet) = tanh(\bullet)$). In short, the rubber matrix was represented by a cooperative game of 21 teams of 2 agents through $\mathscr{A}_j^i$, $i \in \{1,21\}$, $j \in \{1,2\}$. After agent fusion, the final cost function is given by

$$ E(\mathbf{W}_1, \mathbf{W}_2, \mathbf{W}_3) = \frac{1}{2} \sum_{n=1} [g_1 (\sum_{i=1}^{21} \sum_{j=1}^{2} w_i \frac{\partial \mathscr{A}_j^i}{\partial {\lambda_j}^{d_i}} \frac{\partial {\lambda_j}^{d_i}}{\partial \mathbf{F}} - p\mathbf{F}^{-T}) g_1 - P_n]^2, \tag{6} $$

subjected to weights related to λ_{1-max} and $\lambda_{2-max} < 0$, and weights related to λ_1 and $\lambda_2 > 0$ to satisfy thermodynamic consistency and polyconvexity, respectively. We optimize the cost function using optimization algorithms [36]. Eqs. (7) and (8) demonstrate the derivation of the energy of each sub-element with respect to the gradient of deformation.

$$ \sum_{i=1}^{21} w_i \frac{\partial \mathscr{A}_1^i}{\partial {\lambda_1}^{d_i}} \frac{\partial {\lambda_1}^{d_i}}{\partial \mathbf{F}} = \sum_{i=1}^{21} w_i \frac{\partial \mathscr{A}_1^i}{\partial {\lambda_1}^{d_i}} \frac{1}{{\lambda_1}^{d_i}} \mathbf{F} (d_i \otimes d_i). \tag{7} $$

$$ \sum_{i=1}^{21} w_i \frac{\partial \mathscr{A}_2^i}{\partial {\lambda_2}^{d_i}} \frac{\partial {\lambda_2}^{d_i}}{\partial \mathbf{F}} = -\sum_{i=1}^{21} w_i \frac{\partial \mathscr{A}_2^i}{\partial {\lambda_2}^{d_i}} \frac{1}{{\lambda_2}^{d_i}} \mathbf{F}^{-1} \mathbf{F}^{-T} \mathbf{F}^{-1} (d_i \otimes d_i). \tag{8} $$

Case Study 1: Thermo-Oxidation + Mechanical Damage

Natural Rubber: Thermal degradation was predicted at various times and temperatures to validate model prediction performance. Firstly, experimental data from a natural rubber relaxation test filled with 60 phr carbon-black was used to demonstrate model results. Fig. 1 shows the results.

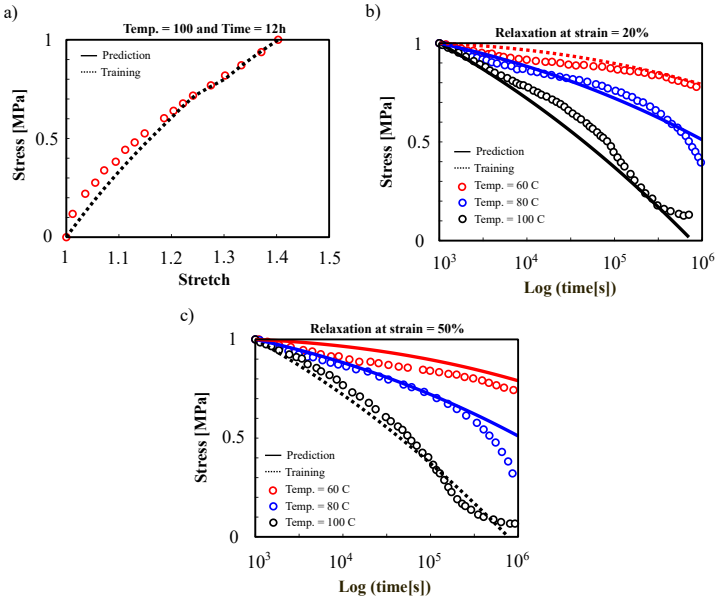

FIGURE 1. Training and model predictions for NR against; (a) intermittent test, (b) relaxation test at $\varepsilon = 20\%$, (c) relaxation test at $\varepsilon = 50\%$.

Black SBR: We performed an experimental study to investigate the impact of aging on the constitutive behavior of cross-linked elastomers to further evaluate the efficiency of the model. Black SBR sheets with a thickness of $\frac{1}{800}$ were therefore purchased from the company Rubbercal. Next, samples were punched in accordance with the standard ASTM D412 using a bone shape punch. We aged specimens at four different temperatures in zero humidity. namely $45°C$, $60°C$, $80°C$ and $95°C$. We applied various sets of tensile and cyclic tests on aged samples using a TESTRESOURCES tensile testing unit. Fig. 2 indicates the performance of the model in the prediction of inelastic behavior such as the Mullins effect and permanent set.

Case Study 2:Hydrolysis + Mechanical Damage

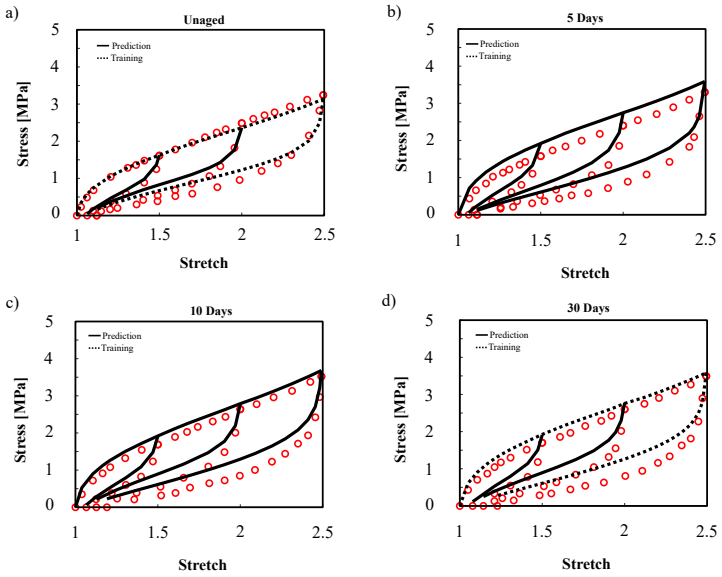

FIGURE 2. Training and model predictions for SBR against loading-unloading responses at 60°C; (a) unaged, (b) aged for 5 days, (c) aged for 10 days, (d) aged for 30 days.

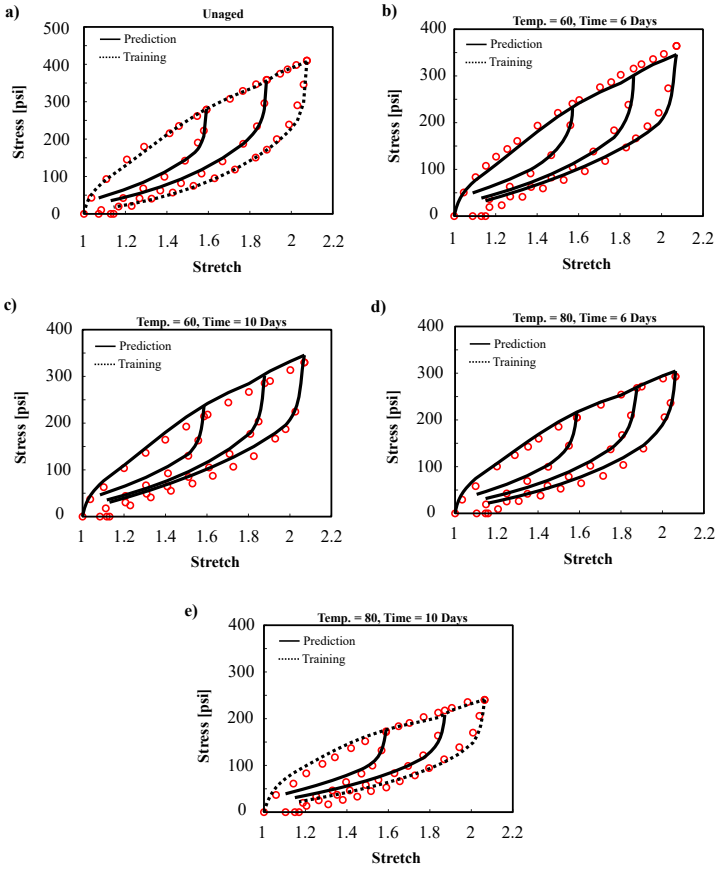

FIGURE 3. Multiple SBR training and model predictions; (a) unaged, (b) constitutive behavior for 6 days of age at temperature study of 60°C, (c) constitutive behavior for 10 days of age at temperature study of 60°C, (d) constitutive behavior for 6 days of age at temperature study of 80°C, (e) constitutive behavior for 10 days of age at temperature study of 80°C.

Styrene-butadiene rubber (SBR): SBR procured sheet with dimensions of $24" * 12" * 0.125"$ was used from one batch-one supplier with a punch die for validation of the invented model (Die c from the ASTM-D412). Samples were placed at temperatures of 60°C and 80°C at constant pressure in sealed containers filled with distilled water. After that, ten days before characterization, they were removed and dried at room temperature. We used an SFM-20 united testing machine with a 1000 lb load cell for quasi-static tensile tests. A strain rate of $43.29 \frac{\%}{min}$ was used at room temperature for extension. The samples were stretched until breakage in monotonic failure experiments, but the samples were expanded to preset amplitudes of 1.3, 1.6, 1.9, and 2.1 in the cyclic test. The central zone extension was measured using an external extensometer. Fig. 3 shows the effect of time and temperature on damages induced by deformation.

CONCLUSION

At present, there is no constitutive model that can cover all single degradation mechanisms simultaneously for elastomers (i.e. thermo-oxidative and hydrolytic aging). Single mechanisms degradation changes the polymer matrix over time, consisting of chain scission due to the presence of temperature, reduction of cross-links attributed to the attendance of water, and reduction of cross-links as a result of the oxygen existence. The mechanical behavior of elastomers can be influenced directly by this alteration in the polymer matrix. Here, the idealized Mullins effect and permanent set have been modeled on the effect of single aging mechanisms on nonlinear mechanical responses of rubber-like materials. We derived a CNN based constitutive model for single mechanisms aging in quasi-static deformation for different states of deformation. The method only requires a macroscopic experimental data set, which is available easily. The performance and predictive capabilities of the model are illustrated by comparing it to different sets of experimental data, such as relaxation and intermittent tests. Also, thermodynamic consistency and frame independency of the model was investigated. The proposed model is based on the homogeneous diffusion assumption and is primarily suitable for thin samples. The accuracy and simplicity of the model make it a proper choice for commercial and industrial applications because we do not need to know the exact behavior and interaction of micro-structures; however, in the future, the model can be extended to consider viscoelasticity and non-isotropic formation for better precision due to platform of the model.

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