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PHYSICS-CONSTRAINED NN-BASED HYPERELASTIC CONSTITUTIVE MODELING OF TPVS

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Summary. *Understanding the thermo-mechanical behavior of thermoplastic vulcanizates (TPVs) is crucial to improving their design and development workflow for industrial applications. TPVs are aimed to replace traditional thermoset rubbers due to their processing ease, recyclability, and low density, but they face challenges in modeling strategies that accurately and efficiently capture their complex nonlinear mechanical behavior. Machine learning is increasingly integrated into various scientific and engineering fields, showing its greatest utility where conventional methods are inefficient. It is expected that machine learning, particularly neural networks, can circumvent the limitations of phenomenological and micromechanical constitutive modeling of TPVs. In this work, the capacity of physics-constrained neural network-based constitutive models in the literature to fit hyperelastic data is assessed, laying the groundwork to model more complex behaviors. To train the models, a cyclic loading-unloading test campaign was conducted for TPV-based test specimens, obtaining the necessary uniaxial tension, pure shear, and equibiaxial tension strain-stress data. Then, the models were benchmarked with the future aim of integrating them into a FEM package.*

1 INTRODUCTION

Elastomers play a pivotal role in various industrial applications due to their exceptional elasticity, resilience, and dynamical behavior [1]. They are widely used across multiple sectors, including automotive, aerospace, construction, and consumer goods. Among these, the automotive industry stands out as the primary consumer of elastomers, relying on them to manufacture tires, seals, gaskets, hoses, and various other components essential for vehicle performance and safety. Additionally, elastomers are integral to anti-vibration components, which are critical for minimizing noise, vibration, and harshness (NVH) in vehicles. These properties enhance passenger comfort and protect sensitive vehicle parts from dynamic stresses.

However, traditional thermoset rubbers are not recyclable, posing significant environmental challenges. This limitation drives the need for alternative materials that maintain the desirable properties of elastomers while offering better circularity. Thermoplastic elastomers (TPEs) are a special kind of elastomeric materials that merge the elastic characteristics of rubber with other properties of thermoplastics [2]. TPEs offer significant benefits, such as recyclability, ease of processing, and low density, making them increasingly popular in various applications. Among TPEs, thermoplastic vulcanizates (TPVs) are of particular interest. TPVs are composed of a high amount of crosslinked rubber particles dispersed in a thermoplastic matrix [3]. They are prepared by a complex reactive blending process characterized by the selective crosslinking of the rubber phase during the melt-blending with the thermoplastic under high shear conditions. This process is known as dynamic vulcanization.

The heterogeneous microstructure of TPVs generates a highly nonlinear stress-strain behavior, making the assessment of constitutive models that reproduce all the characteristics difficult and time-consuming. One of the most important characteristics of TPVs that needs to be predicted is their large nonlinear elastic deformation leading to their categorization as hyperelastic materials. The hyperelastic behavior is usually characterized by cyclic loading-unloading quasi-static tests. A TPV material subjected to cyclic loading-unloading tests exhibits strain softening or Mullins effect [4], permanent deformations [5], and hysteresis [6], which further complicates fitting their behavior using traditional constitutive models.

With the boom of "big data" and the emergence of the fourth paradigm of science [7]—which aims for data-driven scientific discovery— there is an increasing agreement that addressing complex science and engineering challenges demands innovative methodologies that integrate physics-based modeling with state-of-the-art machine learning (ML) techniques [8]. Artificial Neural Networks (ANNs) are the most popular ML models in the solid-mechanics research community. Since pioneers Ghaboussi et al. [9] proposed the first ANN as a mechanical constitutive model, their use has skyrocketed in the last years due to their ability to approximate complex nonlinear functions [10], their ease of use through advanced open-source frameworks, and the possibility to integrate them into finite element method (FEM) packages [11], among others.

ML, and particularly Deep Learning (DL) [12], benefited immensely from the establishment of benchmarks [13]. Benchmarking plays an indispensable role in the development and evolution of ML technologies by providing a standardized suite of tests for objective evaluation and comparison of various algorithms and models. Benchmarks set defined tasks and standards to achieve, driving innovation by challenging researchers to surpass these standards. For hyperelasticity, Treloar's data [14] has been extensively used to benchmark the accuracy of hyperelastic constitutive model fittings [15, 16], including data-driven models [17]. However, there is a lack of research on benchmarking data-driven hyperelastic constitutive models using real experimental data from TPEs, and particularly, TPVs.

In this work, the capacity of existing physics-constrained NN-based constitutive models in the literature [18, 19, 20] to fit hyperelastic data of TPVs was evaluated. The performed experimental campaign involved conducting cyclic loading-unloading tests on TPV-based specimens to obtain the necessary strain-stress data at different deformation modes, including uniaxial tension, pure shear, and equibiaxial tension. These data enabled to benchmark the ANN models with the future aim of integrating them into a FEM package, advancing the design and development workflow for industrial applications of TPVs.

2 HYPERELASTIC CONSTITUTIVE MODELING

2.1 Traditional constitutive models

Constitutive modeling in mechanics is the practice of representing the mechanical properties of materials through mathematical models. These models are formulated based on physical principles and supported by experimental data, combining mathematical problems with physical concepts and empirical evidence. In the most general form, constitutive equations in solid mechanics are tensor-valued tensor functions that define the relation between stress, such as the first Piola-Kirchoff or nominal stress $\mathbf{P}$, and a deformation measure, typically the deformation gradient $\mathbf{F}$. By capturing the relation between stress and strain in materials, constitutive models facilitate the prediction of material responses under various loading scenarios.

Hyperelastic constitutive modeling specifically addresses materials that exhibit significant elastic behavior when subjected to large deformations, allowing them to return to their original shape upon the removal of loads. Hyperelasticity extends linear elasticity to non-linear cases, making it suitable for predicting large strains [21]. These models are particularly relevant for elastomers and TPVs —known for their ability to undergo large, reversible deformations— and serve as a foundation for modeling more complex mechanical behaviors.

Traditional approaches to hyperelastic constitutive modeling can be broadly categorized into micromechanical and phenomenological models. Micromechanical models are inspired by the material's microstructure and attempt to link the macroscopic behavior to the microscopic properties of the material constituents. They often provide a detailed understanding of the material behavior but can be computationally intensive and require extensive knowledge of the material's microstructure.

Phenomenological models are based on observed material behavior and employ mathematical functions to describe the stress-strain relations. Common phenomenological models include the Yeoh [22], Mooney-Rivlin, Ogden, and Neo-Hookean models. While they are less computationally demanding and easier to implement, they may lack the precision in capturing complex material behaviors seen in micromechanical models, but they are still broadly used within the automotive industry for their simplicity. Most hyperelastic phenomenological models relate the invariants of the right Cauchy-Green tensor $\mathbf{C}$ (defined as the product of $\mathbf{F}$ and its transpose) to the strain energy density function Φ, from which consequently the stress response is derived. Expressing Φ in terms of the invariants of $\mathbf{C}$ satisfies objectivity by default [23].

2.2 Neural network-based constitutive models

Data-driven approaches have emerged as powerful tools for constitutive modeling in computational mechanics [24]. Traditional models are often limited by their reliance on predefined functional forms, which may not capture the full complexity of material behavior. In contrast, data-driven methods can learn directly from experimental data, providing more accurate and flexible models.

Due to their exceptional ability to approximate nonlinear functions, ANNs are naturally suited to model complex constitutive relations. However, standard ANNs, which lack knowledge of the underlying physics of the relations they model, face an immensely large function space. This vastness calls for extensive training data (hence they are more suited for big data regimes, often unattainable in science and engineering), limits their generalization and extrapolation ability, hinders the convergence of their algorithms, and results in black-box models that are often uninterpretable.

Physics-informed ML [25] is a burgeoning field of research that integrates physical knowledge into data-driven models to address the limitations of purely data-driven approaches. In the case of ANNs, they generally include physics in a weak form, where learning biases force the training to converge on physically consistent solutions by modifying the loss term, in the manner of Physics-Informed NNs (PINNs) [26]; or in a strong form, where inductive biases are encoded as model-based structures a priori. This second approach of injecting physical knowledge into the inner architecture of the NNs is being increasingly used for mechanical constitutive models in general [27] and for hyperelasticity in particular [17].

Hyperelastic materials present additional challenges for constitutive modeling. The fulfillment of multiple constitutive requirements within ANNs, including objectivity and polyconvexity, is particularly demanding. This challenge has been described as the main open problem of the material behavior theory [28]. Objectivity ensures that the constitutive model's predictions are frame-indifferent, while polyconvexity, a mathematical condition ensuring material stability and the existence of solutions to boundary value problems, is crucial for realistic simulations. Standard ANNs do not inherently satisfy these conditions, necessitating novel approaches.

To address these challenges, various advanced ANN architectures have been proposed. Constitutive ANNs (CANNs) [18] are designed to ensure polyconvexity by using activation functions that preserve convexity and pruning connections between nodes to result in a polyconvex strain energy density function. Input Convex NNs (ICNNs) [29, 20] achieve similar goals by building convex functions of invariant inputs using specific activation functions and non-negative weights, ensuring that the resulting model adheres to the necessary physical constraints. Neural Ordinary Differential Equations (NODEs) [19] take a different approach by leveraging the monotonicity of trajectories of ordinary differential equations to interpolate monotonic functions associated with the derivatives of strain energy rather than the energy itself. This method helps ensure that the models remain physically consistent and adhere to the principles of hyperelasticity.

3 METHODOLOGY

To obtain the necessary data to fit the NN-based constitutive models, experimental tests in three different deformation modes were conducted for a commercial TPV material. This approach ensures that the calibrated hyperelastic model accurately describes the material behavior during complex deformations. To take into account the Mullins effect, cyclic loading-unloading tests were conducted. Then, a backbone (or envelope) curve of the material's stabilized response was built, which considers the permanent set observed in the tests, according to standard procedures within the rubber industry.

3.1 Sample preparation

Specimens tailored to each deformation mode were prepared through an injection molding process, ensuring consistent material properties by using the same batch for all samples. The selected commercial TPV material was Santoprene$^{\mathrm{TM}}$ 101-55, provided by Celanese, a TPV known for its good dynamic performance. This material combines excellent physical properties and chemical resistance, making it suitable for a wide range of applications. Santoprene$^{\mathrm{TM}}$ 101-55 is shear-dependent and can be processed on conventional thermoplastics equipment for injection molding. It is recommended for applications requiring excellent flex fatigue resistance and offers outstanding ozone resistance.

Different specimens were injected for each of the deformation modes to be tested. For uniaxial tension (UT) tests, dumbbell-shaped specimens were used to concentrate stress in the gauge length, facilitating a clear failure point. Rectangular specimens were employed for pure shear (PS) tests to create uniform shear strain across the material. For compression tests, cylindrical specimens were utilized.

3.2 Cyclic loading-unloading tests

The structural properties of elastomers change significantly during the first several times the material experiences straining, a phenomenon commonly referred to as the Mullins effect. When an elastomer is loaded to a particular strain level and then completely unloaded to zero stress several times, the changes in structural properties from cycle to cycle, as measured by the stress-strain function, diminish. Once the stress-strain function no longer changes significantly, the material is considered stable for strain levels below that maximum. Since the Mullins effect typically stabilizes after five cycles, five load-unload cycles at each strain level were conducted. However, if the elastomer is subsequently subjected to a higher strain maximum, the structural properties will change significantly again. The loading patterns in the conducted cyclic tests allow us to observe the effects of Mullins softening on material stiffness and any plastic strain accumulation.

Specimens were subjected to cyclic loading using an MTS universal testing machine at a constant strain rate of 100%/min. For UT and PS tests, eight strain levels (10%, 20%, 30%, 50%, 70%, 100%, 150%, and 200%) were applied. For compression tests, six strain levels (5%, 10%, 15%, 20%, 30%, and 50%) were used.

Due to the lack of an equibiaxial testing machine, uniaxial compression tests were conducted to infer the equibiaxial (ET) deformation mode. For incompressible materials, uniaxial compression tests create a strain state equivalent to equibiaxial extension [30]. While equibiaxial tests are generally more complex and provide a more accurate material model by achieving a pure strain state, compression tests can serve as a proxy for equibiaxial tension due to our assumption of the incompressibility of TPV materials.

However, achieving pure compression strain is challenging because of friction-induced shearing strains and nonhomogeneous deformations like barreling, which can lead to inaccuracies in the test results. Additionally, the maximum strain level in compression tests is inherently lower than in uniaxial tension and pure shear tests, which limits the strain range that can be effectively analyzed for hyperelastic model development.

3.3 Backbone curve generation

Backbone curves representing the material's stabilized response were derived using data from the fifth cycle of each loading-unloading test. Due to the observed permanent set and given that hyperelastic models assume the stress-strain curve initiates at the (0, 0) point, Eqs. (1) and (2) were applied to adjust the curves before obtaining the backbone.

$$\sigma' = \sigma_0(1 + \varepsilon_{\mathrm{set}}) \tag{1}$$

$$\varepsilon' = \left(\frac{1 + \varepsilon_0}{1 + \varepsilon_{\mathrm{set}}}\right) - 1 \tag{2}$$

Where σ_0 is the nominal or engineering stress, ε_0 is the nominal or engineering strain, and ε_{set} is the plastic strain or permanent set.

As an example, Fig. 1 shows the fifth cyclic curves of uniaxial tension and the backbone curve obtained after adjusting the permanent set.

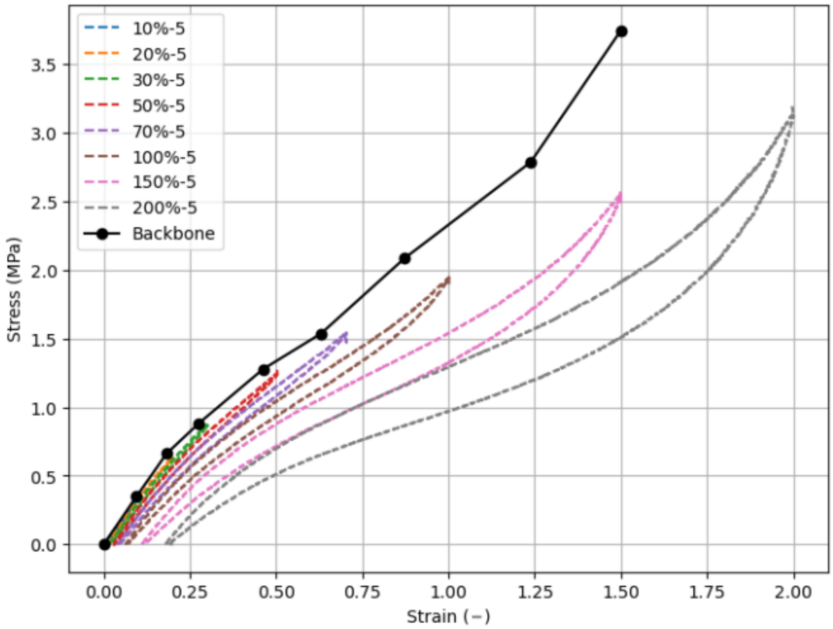

Figure 1: Fifth cyclic curves of uniaxial tension test of Santoprene$^{\text{TM}}$ 101-55 and processed backbone curve.

After adding one to the strain vectors to obtain stretch-stress curves, the backbone curves of each deformation mode were ready to serve as input for the selected hyperelastic constitutive models.

3.4 Constitutive modeling of backbone curves

Three different physics-constrained NN-based hyperelastic constitutive models were compared to study their applicability to fit backbone curves of TPVs: CANNs, ICNNs, and NODEs. Additionally, to compare the quality of the fittings to a traditional phenomenological constitutive model, the data was fitted to the Yeoh constitutive model too.

In addition to evaluating the applicability of selected data-driven models to the mechanical characterization methodology of TPVs, another objective was to study the models' capability to extrapolate from one deformation mode to another. By incorporating knowledge of hyperelastic material physics within the architecture of the models, it was intended to ascertain whether data from single-mode deformation tests could be used to fit curves for other deformation modes with adequate quality. If successful, this approach would be particularly valuable for deriving accurate material models of TPVs' hyperelastic behavior from the simplest test: the uniaxial tensile test.

4 RESULTS AND DISCUSSION

The results of the fittings of the different hyperelastic constitutive models are displayed in Fig. 2. Each column corresponds to the training data from different deformation modes: uniaxial tension (UT), equibiaxial tension (ET), pure shear (PS), and all data combined. Table 1 shows the R^2 scores of each fitting.

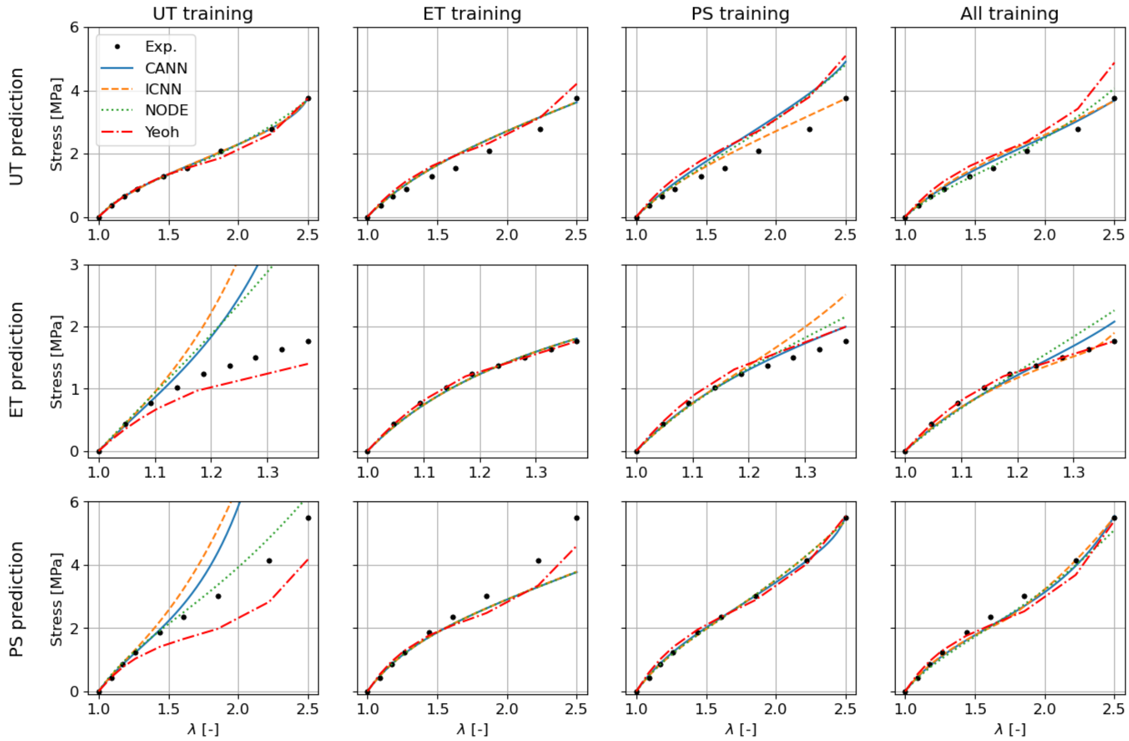

Figure 2: Fittings of hyperelastic constitutive models to the backbone data of Santoprene™ 101-55. The first three columns show the fittings to the three deformation modes using only training data from UT, ET, and PS, respectively. The column on the right displays the fittings using all data for training.

The NN-based models perform comparably across the different deformation modes. Each model shows a reasonable agreement with the experimental data, with slight variations in their accuracy depending on the mode of deformation used for training. When trained with all the data, ICNN obtains the most balanced fittings, with R^2 scores surpassing 0.975 in all deformation modes. CANN loses accuracy in fitting ET, and this drop is even greater for NODE. In contrast, Yeoh fails to accurately fit UT data at high strain levels.

Regarding the extrapolation ability of the NN-based models, ET and PS deformation modes extrapolate more accurately to other modes compared to the UT mode. When using UT data, predictions of the NN-based models align well with experimental data at low strains but then explode at higher strains, with the exception of NODE's prediction for the PS mode. On the other hand, Yeoh's predictions based on UT data tend to underestimate real values, while they are similar to the NN-based models using ET and PS data.

Table 1: R^2 scores of the fittings of hyperelastic constitutive models to the backbone data of Santoprene^{TM} 101-55.

		UT training	ET training	PS training	All training
UT prediction	CANN	0.999	0.954	0.694	0.985
	ICNN	0.999	0.953	0.958	0.974
	NODE	0.998	0.954	0.751	0.977
	Yeoh	0.993	0.936	0.657	0.820
ET prediction	CANN	-5.866	0.992	0.953	0.933
	ICNN	-13.771	0.992	0.621	0.975
	NODE	-2.053	0.992	0.880	0.826
	Yeoh	0.803	1.000	0.936	1.000
PS prediction	CANN	-6.622	0.849	1.000	0.989
	ICNN	-2.860	0.852	0.998	0.989
	NODE	0.957	0.849	0.998	0.981
	Yeoh	0.798	0.930	0.996	0.979

One notable limitation of the study is related to the fact that ET data was derived from compression tests. The low strain levels and frictional effects in the compression tests introduce concerns about the quality and reliability of the ET data. These factors might affect the accuracy of the models when fitting the data.

Additionally, to ensure the consistency and generalizability of these findings, it is crucial to replicate the process with other TPV materials. Doing so would verify whether the observed trends and model performances are specific to the analyzed Santoprene^{TM} material or if they can be generalized to other similar materials.

5 CONCLUSIONS

Overall, while the NN-based models show promising results in modeling the hyperelastic behavior of TPVs, further investigations and validations are necessary to confirm their applicability and reliability within the TPV-based part and product development workflow.

Physics-constrained NN-based hyperelastic constitutive models have great potential but they are still an ongoing area of research. Their extrapolation ability is promising, especially at low strains, which is particularly relevant since most TPV-based parts, such as antivibration components, typically do not experience large stretches in real use. However, actual ET tests are required for confirmation.

Future work includes validating the models by testing other TPV materials, integrating the NN-based constitutive models into a FEM package as material models, and addressing additional non-linear mechanical behaviors with the help of physics-informed ML techniques.

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