Prediction of rubber vulcanization using an artificial neural network

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Abstract

Determination of rubber rheological properties is indispensable in order to conduct efficient vulcanization process in rubber industry. The main goal of this study was development of an advanced artificial neural network (ANN) for quick and accurate vulcanization data prediction of commercially available rubber gum for tire production. The ANN was developed by using the platform for large-scale machine learning TensorFlow with the Sequential Keras-Dense layer model, in a Python framework. The ANN was trained and validated on previously determined experimental data of torque on time at five different temperatures, in the range from 140 to 180 °C, with a step of 10 °C. The activation functions, ReLU, Sigmoid and Softplus, were used to minimize error, where the ANN model with Softplus showed the most accurate predictions. Numbers of neurons and layers were varied, where the ANN with two layers and 20 neurons in each layer showed the most valid results. The proposed ANN was trained at temperatures of 140, 160 and 180 °C and used to predict the torque dependence on time for two test temperatures (150 and 170 °C). The obtained solutions were confirmed as accurate predictions, showing the mean absolute percentage error (MAPE) and mean squared error (MSE) values were less than 1.99 % and 0.032 $dN^2 m^2$, respectively.

keywords: rubber curing; machine learning; rubber rheological properties.

Available on-line at the Journal web address: http://www.ache.org.rs/HI/

TECHNICAL PAPER

UDC: 675.92.028.3-036.4: 004.032.26

Hem. Ind. **75 (5)** 277-283 (2021)

1. INTRODUCTION

Natural rubber (NR) is an elastomeric polymer, widely used in preparation of rubber products. Vulcanization process of rubber has a crucial influence on the final product quality [1]. Prediction of rubber vulcanization data is particularly complicated due to the induction and reversion phenomena, which are presented in Figure 1. [2].

In general, the crosslink density of rubber compounds starts increasing slowly during the induction period, after which the main vulcanization reaction takes place with a significant increase of the crosslink density. Furthermore, the rubber blend at high temperatures (above 140 °C) can reach the torque maximum and start to decrease, causing weaker mechanical properties at longer curing times [3]. Studies dedicated to prediction of products of rubber vulcanization, mostly, neglected the reversion phenomenon, whereas the induction period and the main vulcanization kinetics were analysed separately [4–8]. In order to avoid complicated cross-linking kinetics, rubber rheological data can be successfully predicted using artificial neural networks (ANN). Neural networks are composed of basic units, termed neurons or nodes, arranged in layers. A neuron collects information provided by other neurons to which it is connected by weighted connections, where synaptic weights multiply the input information. Each neuron transforms its input into output response, where the transformation includes two steps. The first step is neuron activation, where it is computed as the weighted sum of its inputs, and the second step, where the activation is transformed to response by using a transfer function. If input is denoted as x_i and weight as w_i, then the activation computation is the sum of $w_i x_i$, and the output is obtained by applying a transfer function f [9]. Functions, which domains are defined by real numbers, can be used as transfer functions, where the most common are linear, logarithmic sigmoid and hyperbolic tangent functions

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Paper received: 11 May 2021; Paper accepted: 11 October 2021; Paper published: 30 October 2021.
https://doi.org/10.2298/HEMIND210511026L

[10]. Nevertheless, by now there are limited ANN studies on rubber vulcanization and mostly, are aimed at tire design and tire curing equipment. In one study the ability of an ANN to evaluate variability of rheometric properties of rubber compounds from their formulations was investigated [11] and in another the optimum curing times of different rubber compounds were predicted [12].

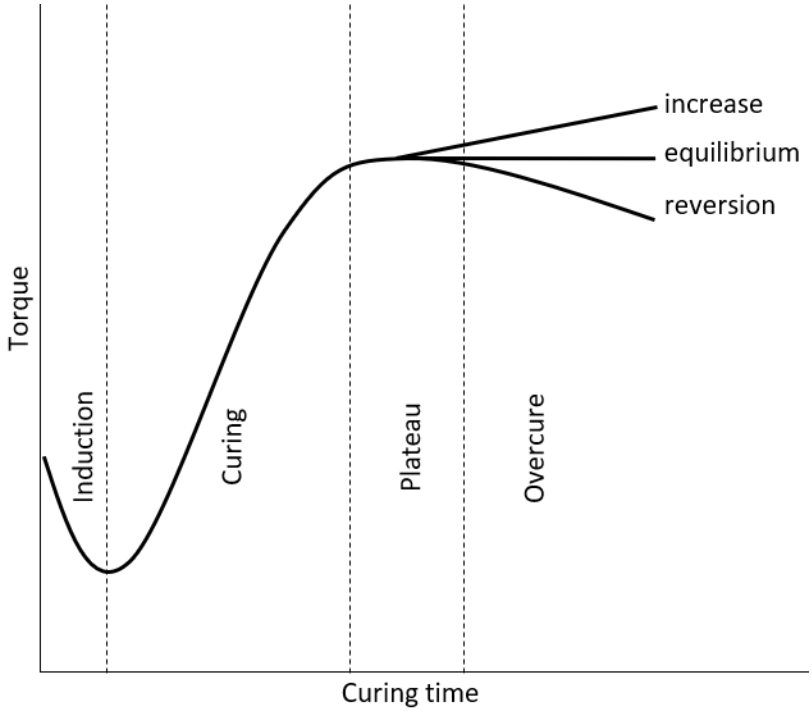

Figure 1. A typical vulcanization curve obtained from a rheometer

In this work, the focus was on ANN model development, able to predict the vulcanization data (torque versus time) for different temperatures. The significant contribution of this study is the built ANN model based on Python software that uses open-source license. Therefore, the rubber community (academic and industrial) can freely apply the introduced ANN code to predict vulcanization torque at different temperatures. The experimental rheometer data obtained previously [13] were used for training the ANN model. Furthermore, the number of neurons in the ANN architecture with one and two layers was varied to minimize the prediction error of the vulcanization torque. Additionally, three activation functions, ReLU, Sigmoid and Softplus, were examined to improve the ANN accuracy.

2. ARTIFICIAL NEURAL NETWORK DEVELOPMENT

Developing a predictive ANN model can be demanding due to nonlinear rubber vulcanization nature. The appropriate ANN architecture is the most challenging part in building the model and can be expected that ANN model with sufficient number of hidden layers may provide the adequate results, with the caution of overfitting [15]. Hidden layers consist of neurons, trained by a properly selected learning algorithm to acquire desired weight and activation function, which are considered as the key elements for the model. Regularly, three steps are followed in developing the ANN models: gathering large amounts of experimental vulcanization data, building and training the model, and model validation with testing [16].

The general ANN model, used in this study, consisted of input, hidden and output layers, as it is shown in Figure 2. [17,18]. The ANN has two neurons in the input layer (time and temperature), selected to predict the torque as a single neuron in the output layer.

The ANN model is built by using the Sequential Keras-Dense layer model of the TensorFlow [19]. Dense layer is commonly and frequently used, and it is a deeply connected neural network layer, in which the following operation is conducted [20]:

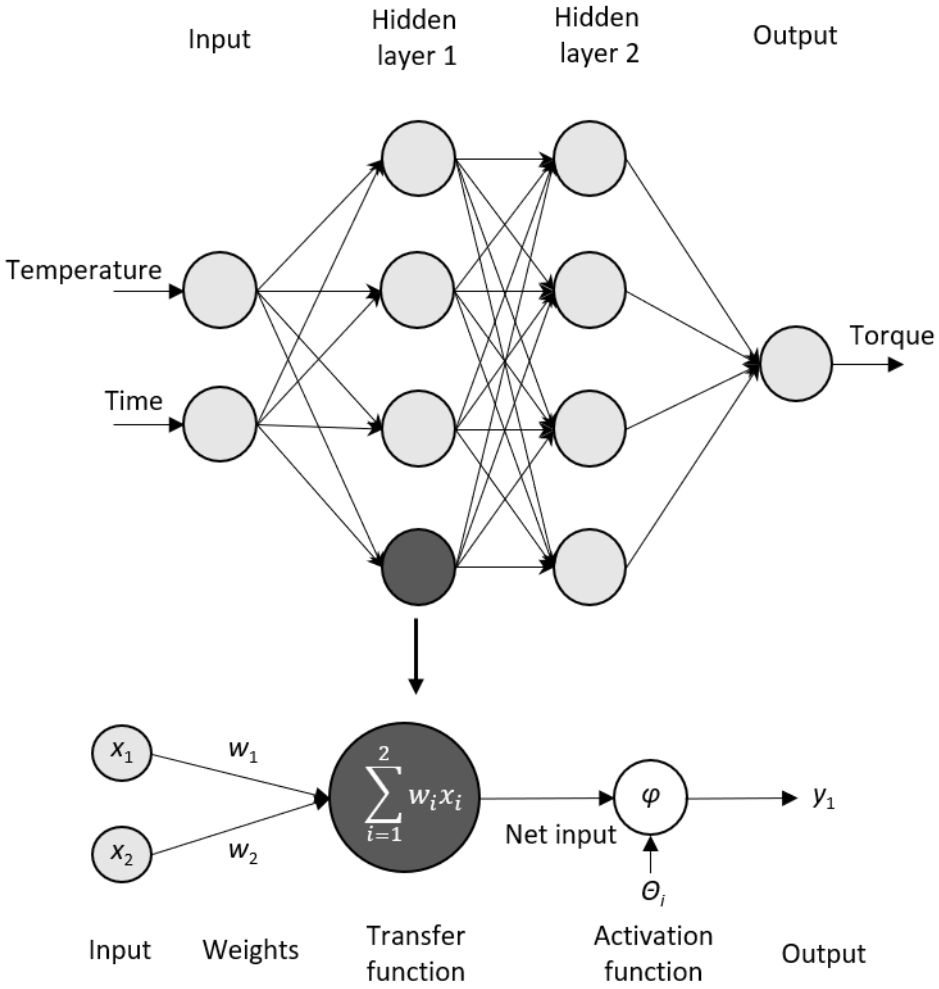

Figure 2. Example of the ANN architecture with two neurons in the input layer, two hidden layers with four neurons in each, and one neuron in the output layer

Output = activation (dot (input, kernel)) + bias (1)

where the *output* represents the output data, *activation* is the activation function, which is passed as the activation argument, *dot* is a numpy dot product of all inputs and their corresponding weights, *input* is the input data, *kernel* is the weight data created by the layer and *bias* is a bias vector created by the layer, which, in the Keras model, is assumed to learn its values [16].

In this research, three activation functions were used to obtain the model with minimal error: rectified linear unit (ReLU), Sigmoid and Softplus activation functions. The ReLU and Sigmoid functions are frequently used in the ANN models [21,22], while Softplus is a relatively novel differentiable function. The ReLU function is a linear function, and it is described as [23]:

$f(x) = 0$; for $x < 0$ $\quad f(x) = x$ for $x \geq 0$ (2)

The Sigmoid function is a non-linear, continuously differentiable, monotonic, S-shaped curve, and can be described as following [24]:

$$f(x) = \frac{1}{1+e^{-x}} \tag{3}$$

Softplus function is defined as [25]:

$f(x) = \ln (1+e^x)$ (4)

Significant parts of an ANN architecture are the loss function and optimizer. In this study, the mean squared error (MSE) was used as the loss function and for training, the model Adam optimizer in TensorFlow was tested. Adam

optimizer is commonly used since it is computationally efficient, well suited for problems that are large in terms of data and appropriate for non-stationary objectives [26,27].

Number of training epochs can cause a problem in training neural networks, where too many epochs may lead to overfitting of the training dataset, whereas epoch lack results in an underfitted model [16]. In the present work, 50, 100 and 150 epochs were tested, and, in order to avoid underfitting and overfitting, number of epochs was set to 100.

The last step in ANN model building is its validation, where the extracted trained model of the training phase is split and is going through the validation phase using the testing dataset. Validation is conducted by using random 20 % of the dataset as a test, while the rest of data is used for training. There is a requirement for the training and validation loss function values to be near zero compared to the respective initial values, representing the sum of errors for each data point.

Additional model validation was performed by comparing the ANN predictions with the experimentally obtained curves, where the MAPE and MSE were used as statistical methods.

MAPE and MSE are calculated by eqs. (5) and (6), respectively:

$$\mathrm{MAPE} = \frac{1}{n} \cdot \sum_{i=1}^{n} \left| \frac{A_i - F_i}{2} \right| \tag{5}$$

$$\mathrm{MSE} = \frac{1}{n} \sum_{i=1}^{n} (A_i - F_i)^2 \tag{6}$$

where n is the number of fitted points, A_i is the actual value and F_i is the forecast value.

In the prediction phase, the developed ANN model was tested for three different activation functions to predict torque data at 150 and 170 °C. Furthermore, numbers of hidden layers and neurons were varied to increase the model accuracy and reliability. Lastly, the predicted vulcanization data are validated by the experimentally obtained curves.

3. RESULTS AND DISCUSSION

3. 1. ANN model

Developed ANN model was trained with rheometric data obtained at 140, 160 and 180 °C [13], where three activation functions (Softplus, ReLU, Sigmoid) and the number of neurons in hidden layer were varied. Firstly, the tests were run with 2, 3, 4 and 5 neurons in one hidden layer for each activation function, and the results of statistical error, as an elimination criterion (MAPE and MSE), are shown in Table 1.

Table 1. Prediction results for rheometric curves determined at 150 and 170 °C by using three different activation functions (ANN with one hidden layer)

Activation function		Softplus		ReLU		Sigmoid	
		Temperature, °C					
No of neurons	Statistical method	150	170	150	170	150	170
2	MAPE,%	5.102	8.293	4.014	5.344	6.423	8.662
	MSE, dN² m²	0.234	0.524	0.4	0.522	0.37	0.556
3	MAPE, %	5.328	9.298	4.584	4.827	6.07	8.133
	MSE, dN² m²	0.306	0.627	0.406	0.489	0.323	0.502
4	MAPE, *%*	3.099	4.84	3.493	4.826	6.078	8.419
	MSE, dN² m²	0.111	0.216	0.341	0.489	0.324	0.523
5	MAPE, *%*	5.772	8.123	5.14	7.921	5.598	8.988
	MSE, dN² m²	0.322	0.515	0.238	0.472	0.288	0.667
10	MAPE, *%*	2.351	3.441	6.601	8.146	5.886	8.551
	MSE, dN² m²	0.152	0.221	0.506	0.646	0.295	0.567
15	MAPE, *%*	2.421	4.1	2.85	3.691	5.566	8.446
	MSE, dN² m²	0.101	0.149	0.169	0.236	0.269	0.516
20	MAPE, *%*	7.834	4.282	3.793	11.646	5.192	8.985
	MSE, dN² m²	0.491	0.18	0.189	0.971	0.237	0.671

It can be seen that ANN models using Softplus and Sigmoid activation functions enabled the adequate rheometric curve predictions at 150 °C, while, at 170 °C, the error was high. Furthermore, the ANN model with ReLU activation function and one hidden layer with 2, 3, 4 or 5 neurons cannot predict torque dependence on time, at 150 and 170 °C. These results led to increased number of neurons (10, 15 and 20) in the hidden layer (Table 1). More than 20 neurons were not tested since high number of neurons can cause overfitting. The ANN model with ReLU activation function with higher numbers of neurons in the hidden layer resulted in lower MAPE and MSE values. However, the ANN prediction of vulcanization process at 170 °C is not satisfactory.

In order to increase the prediction accuracy, ANN model with two hidden layers was developed, and the results are shown in Table 2.

Table 2. Prediction results of rheometric curves determined at 150 and 170 °C for three different activation functions (ANN with two hidden layers containing the same number of neurons)

Activation function		Softplus		ReLU		Sigmoid	
		Temperature, °C					
No of neurons	Statistical method	150	170	150	170	150	170
2	MAPE, %	8.556	5.318	6.21	7.673	6.35	8.573
	MSE, $dN^2 m^2$	0.65	0.32	0.568	0.626	0.366	0.55
3	MAPE, %	7.745	5.031	3.57	4.809	6.029	7.789
	MSE, $dN^2 m^2$	0.631	0.275	0.215	0.393	0.307	0.485
4	MAPE, %	4.788	8.563	3.395	7.111	6.328	8.467
	MSE, $dN^2 m^2$	0.208	0.552	0.169	0.395	0.34	0.562
5	MAPE, %	1.423	2.849	5.19	9.389	5.427	8.938
	MSE, $dN^2 m^2$	0.042	0.109	0.26	0.641	0.261	0.636
10	MAPE, %	2.167	2.118	6.335	7.111	5.712	7.907
	MSE, $dN^2 m^2$	0.068	0.04	0.332	0.395	0.295	0.556
15	MAPE, %	1.627	1.462	7.228	5.738	5.115	8.446
	MSE, $dN^2 m^2$	0.033	0.025	0.469	0.29	0.24	0.515
20	MAPE, %	0.574	1.989	4.918	8.562	5.974	0.515
	MSE, $dN^2 m^2$	0.006	0.032	0.2	0.537	0.305	0.529

It can be observed that the increase in the number of neurons in two hidden layers led to a decrease in MAPE and MSE values. Additionally, the ANN with two hidden layers provided superior results in comparison with the ANN with one hidden layer. As the ANN model using Softplus activation function resulted in the most accurate prediction, it is discussed here in more details. It can be seen that the ANN model with two hidden layers containing 20 neurons provides MAPE lower than 1.99 % and MSE lower than 0.032 $dN^2 m^2$. Comparison of the experimental data and prediction of the developed ANN model is shown in Figure 2.

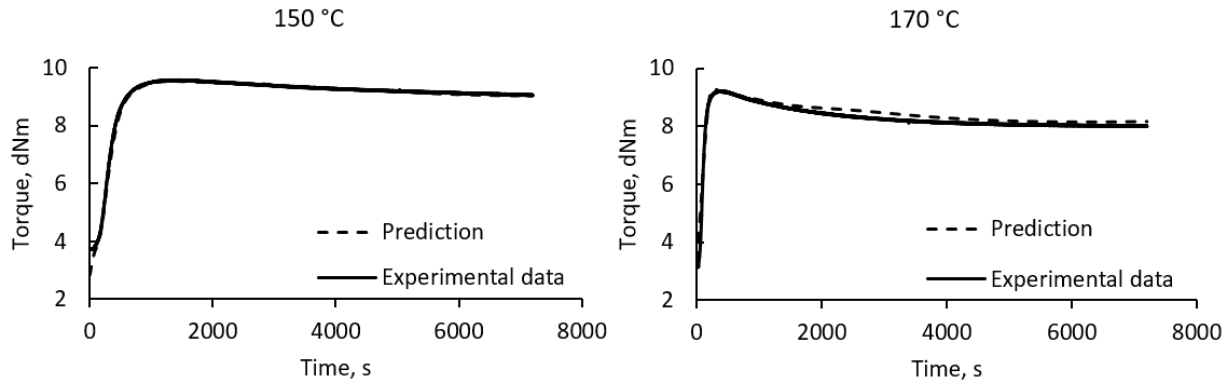

Figure 2. Experimental and predicted vulcanization data for 150 and 170 °C (experimental data are from [13])

The developed ANN model accurately predicted the torque dependences on time at test temperatures, enabling the fast and stable rheological prediction using only three rheological measurements, which is very important from the energy and cost savings aspects.

4. CONCLUSIONS

The aim of this work was to develop the fast and accurate ANN model for rheological data prediction of NR/SBR rubber blend, using free software (Python), facilitating the vulcanization kinetics estimation and leading to time and cost savings, significant for the rubber community. The number of hidden layers, neurons in hidden layers and activation function (Softplus, ReLU, Sigmoid) were optimized in order to minimize the vulcanization data prediction error. The ANN model using Softplus as the activation function, containing 20 neurons in each of two hidden layers, resulted in the most accurate curing curve foresight. The predicted and experimental data were compared, and MAPE and MSE values were less than 1.99 % and 0.032 dN^2 m^2, respectively.

Acknowledgements: The authors would like to acknowledge the Ministry of Education, Science and Technological Development of the Republic of Serbia for the financial support, Project No. 451-03-9/2021-14/200134.

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SAŽETAK

Predviđanje vulkanizacije gume korišćenjem veštačke neuronske mreže

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(Stručni rad)

Za sprovođenje efikasnog procesa vulkanizacije u gumarskoj industriji neophodni su pouzdani reološki rezultati umrežavanja kaučukovih smeša. Stoga, osnovni cilj ovog rada bio je razvoj napredne veštačke neuronske mreže (engl. *artificial neural network*, ANN) za brzo i tačno predviđanje vulkanizacije komercijalno dostupne elastomerne smeše za dobijanje gumenih proizvoda. ANN je trenirana na prethodno određenim eksperimentalnim podacima obrtnog momenta u zavisnosti od vremena na pet različitih temperatura, u temperaturnom opsegu od 140 do 180 °C, sa korakom od 10 °C. ANN model je razvijen pomoću platforme za mašinsko učenje *TensorFlow* primenom *Keras* modela i programskog jezika Pajton (*Python*), gde su model i optimizator bili sekvencijalni model i adam, redom. Sledeće aktivacione funkcije: *ReLU*, sigmoidna i *Softplus*, korišćene su za minimizaciju greške, gde je ANN model sa *Softplus* funkcijom pokazao najtačnije rezultate predviđanja. Varirani su brojevi neurona i slojeva, gde je ANN model sa dva sloja, i 20 neurona u svakom sloju, dao najbolje rezultate. Predloženi ANN model je treniran na podacima dobijenim na temperaturama od 140, 160 i 180 °C, a zatim je korišćen za predviđanje zavisnosti obrtnog momenta od vremena za dve preostale temperature vulkanizacije u datom eksperimentalnom setu (150 i 170 °C). Dobijena rešenja su potvrđena kao tačna predviđanja upotrebom različitih numeričkih metoda, za sve ispitivane temperature, gde su vrednosti srednje apsolutno procentualno odstupanje (*engl*. mean absolute percentage error MAPE) i srednje kvadratno odstupanje (*engl*. mean squared error MSE) manje od 1,99% i 0,032 $dN^2\,m^2$, redom.

Ključne reči: umrežavanje gume; mašinsko učenje; reološka svojstva gume