Modeling the concentration of carbonyl of ethylene propylene diene monomer during the thermal aging using artificial neural network

Hadjira Maouz, Asma Adda*, Salah Hanini

Laboratory of Biomaterials and Transport Phenomena (LBMPT), University of Médéa, Ain D'Heb 26000, Médéa, Algeria,
*Corresponding author; Email: docsolaire2017@gmail.com.

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ABSTRACT

The concentration of carbonyl is one of the most important properties contributing to the detection of the thermal aging of polymer ethylene propylene diene monomer (EPDM). In this publication, an artificial neural network (ANN) model was developed to predict concentration of carbenyl during the thermal aging of EPDM using a database consisting of seven input variables. The best fitting training data was obtained with the architecture of (7 inputs neurons, 10 hidden neurons and 1 output neuron). A Levenberg Marquardt learning (LM) algorithm, hyperbolic tangent transfer function were used at the hidden and output layer respectively. The optimal ANN was obtained with a high correlation coefficient R= 0.995 and a very low root mean square error RMSE = 0.0148 mol/l during the generalization phase. The comparison between the experimental and calculated results show that the ANN model is able of predicted the concentration of carbonyl during the thermal aging of ethylene propylene diene monomer.

I. Introduction

Polymer materials have been increasingly used in many areas, for example, aerospace, automobile, and construction industries [1]. Among the commodity polymer ethylene propylene diene monomer (EPDM) are predominantly ethylene-based terpolymers, but also propylene and diene, found in many industrial sectors such as automotive, building, nuclear, etc. These elastomers have many advantages because they have good chemical resistance to many environmental factors (oxygen, high temperature...) [2]. Aging, the EPDM degrades under the action of various environmental factors: radiation [3-5], the temperature [6,7], ozone [8], acidic or basic chemical agents [9,10], and oxygen [11-13], which leads to a loss of its functional properties (scission, cross-linking, loss of gloss, whitening, reduction of average molar mass, chalking…). There is a lot of work dealing with the thermal aging of EPDM matrices [14-21]. The oxidation of the polymers can be delayed or completely blocked by the addition of antioxidants. In the case of thermal aging, they are mainly radical inhibitors (which accelerate the termination) and non-radical radical hydroperoxide decomposers (which slow down the initiation) [2]. The very particular field of the thermal aging of ethylene propylene diene monomer (EPDM) does not escape the attempts of modeling and the industrial demand is strong for an approach that would allow, after short analysis periods, or even short, to predict the long term behavior of EPDM materials under conditions of actual use. Many researchers have adopted artificial intelligence approaches to predict material behaviors, characteristics and attributes under changing circumstances (e.g. aging over time, heat treatment due to temperature changes, etc) [22], these approaches, such as Artificial Neural Networks (ANN), indeed, the networks of formal or ANN always arouse a keen interest in most areas of engineering. They find their applications, not only in static modeling but also in dynamic modeling of evolutionary processes over time: form recognition, non-destructive testing, textual information filtering, and bioengineering, formulation of new materials, industrial process modeling, and control of the environment…etc. ANN which can be viewed as a universal approximation tool with an inherent ability to extract from experimental data the highly non linear and complex relationships between the variables of the problem handled [23].

In this study, the ANN approach was used to develop a model that uses aging conditions (temperature), percent concentrations of polymers and additives, aging times and pseudo-molecular weights of polymers and additives to accurately predict and reliably the concentration of carbenyl.

II. Methodology and modeling

Artificial neural networks (ANN) are a set of algorithms the design of which is at the origin very schematically inspired by the functioning of biological neurons. They are now used as a very powerful tool to model and analyze processes as well as to make the prediction of the behavior of a given system [24].

The ANN structure consists essentially of input layer (independent variables), a number of hidden layers and an output layer. Each of these layers consists of a number of interconnected processing units called neurons. These neurons interact by sending signals and they are connected to all the neurons on the previous layer, and the next layer by links called weights and links [25]. The architecture of the ANN model is showed in Figure 1.

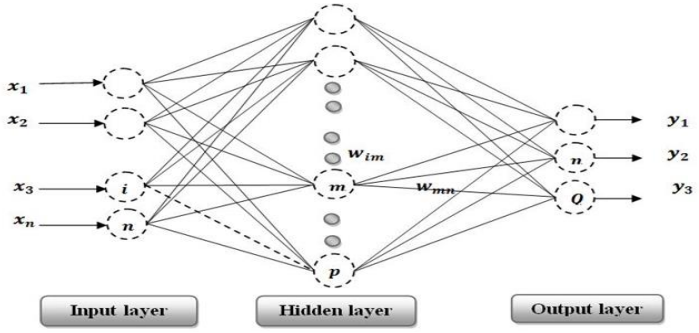


Figure 1. Structure of ANN.

The ANN are calculating techniques designed to simulate the way in which the human brain a carries out a specific task [26] through a large treatment distributed in parallel and constituted from simple treatment units [23, 27, 28]. Literature was used to determine the database [2]. In the current work, the size of the data was arranged as matrix with (462, 1) containing 07 inputs parameters and an output parameter is the concentration of carbonyl.

The statistical analysis (standard deviations (STD), minimum (min), maximum (max) and mean (mean)) for the inputs and output dataset are shown in Table 1.

Table 1. Statistical analysis of inputs and output.

	Min	**Mean**	**STD**	**Max**
t(h)	0	19.8839	19.0320	116.7330
T(°C)	140	149.1342	7.6889	160
X_1 (% w/w)	96.6000	96.7779	0.1123	97
X_2 (% w/w)	2.9000	3	2.9037	0.0188
X_3 (% w/w)	0	0.2432	0.1508	0.4800
X_4 (% w/w)	0	0.0635	0.0841	0.1900
M_w (kg/mol)	1.4551e+03	1.4603e+04	2.0816e+03	1.5715e+04
[CO] mol/l	7.0000e-04	0.0929	0.1483	1.2236

The ANN modeling of the concentration of carbonyl during thermal aging of EPDM was performed using MATLAB software. The experimental data were randomly divided into three subsets (training phase, validation phase and test phase). In order to obtain an optimal neural network giving approximate results to the experimental data, a hidden layer was used by changing the number of neurons from 1 to 30 30, the selection of neurons must generally be performed by trial and error. The tangent sigmoid transfer function was used for the hidden layer and

the output layer. The learning of the model must be done; each topology has been repeated three times. The optimal structure corresponds to the correlation coefficient R close to 1 and roots means squared error (RMSE), the mean absolute error (MAE) and the model predictive error (MPE) close to 0. The relations of RMSE, MAE and MPE are given by the following equations:

$$RMSE = \sqrt{\sum_{i=1}^{n} \frac{(Y_{exp} - Y_{cal})^2}{n}} \qquad (1)$$

$$\mathrm{MAE} = \frac{1}{\mathrm{n}} \sum_{\mathrm{i=1}}^{\mathrm{n}} \left| (\mathrm{Y_{exp}} - \mathrm{Y_{cal}}) \right| \qquad (2)$$

$$\mathrm{MPE}(\%) = \frac{100}{\mathrm{n}} \sum_{\mathrm{i=1}}^{\mathrm{n}} \left| \frac{(\mathrm{Y_{exp}} - \mathrm{Y_{cal}})}{\mathrm{Y_{exp}}} \right| \qquad (3)$$

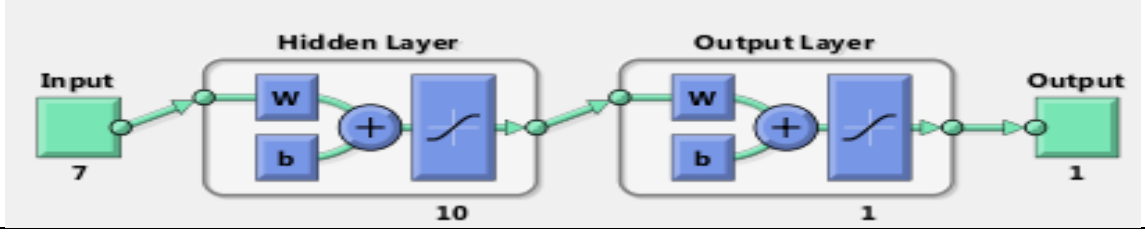
Where n is the total number of data points, Y_{exp} is the experimental value, and Y_{cal} represents the calculated value from the neural network model.

III. Results and discussion

According to the previous discussion, an artificial neural network is developed to predict the concentration of carbonyl during the thermal aging of EPDM. Ten neurons in the hidden layer were sufficient to achieve a better correlation coefficient (R=0.9940, R=0.995, R=0.9976 and R=0.9956 for training phase, validation phase, teste phase and all database respectively) with a very small roots mean squared error (RMSE=0.0148). The values of these parameters were obtained after performing several trial and error runs. The results obtained are presented in Table 2 and in Figure 2.

Table 2. Architecture of the optimized NN model.

Training algorithm	Input layer	Hidden layer		Output layer	
	Neurons number	Neurons number	Activation function	Neurons number	Activation function
BRBP - Levenberg-Marquardt. (Function ***trainbr*** de MATLAB)	7	10	Tangent	1	Tangent



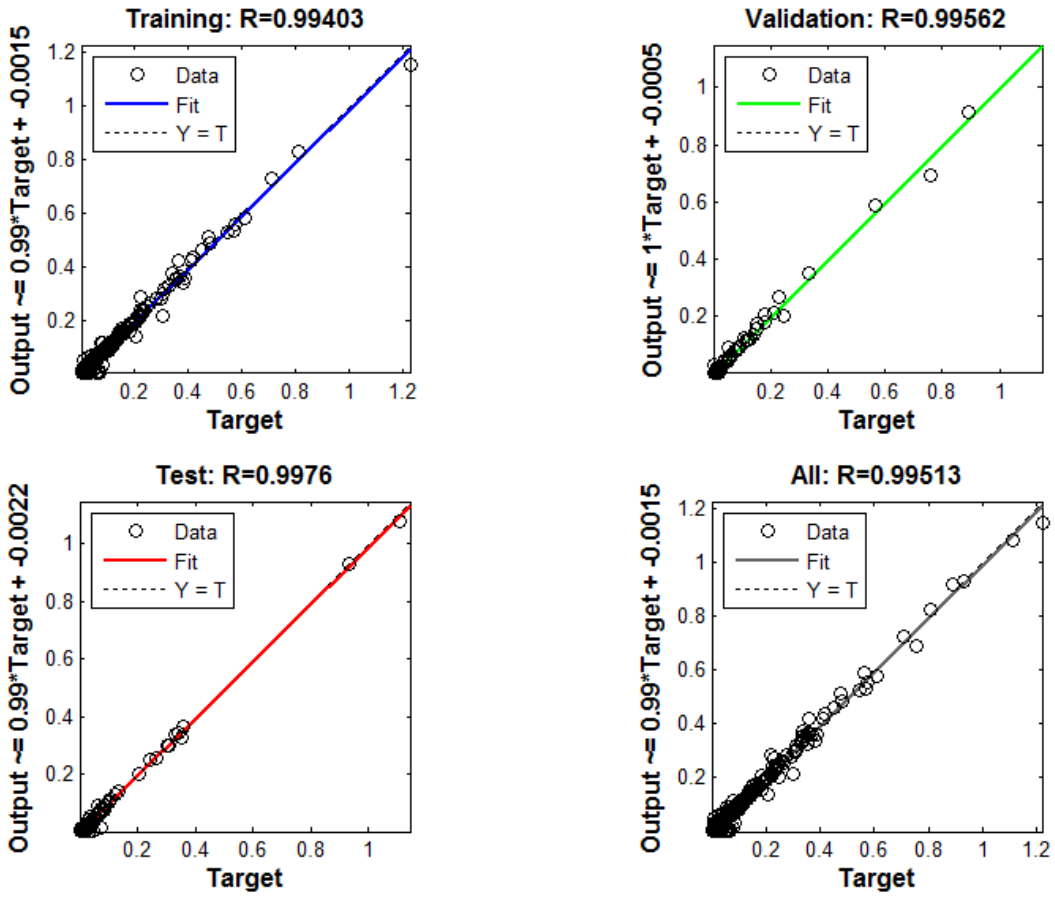


Figure 2. The regression curve represents the predicted concentration of carbonyl depending on the experimental concentration of carbonyl during thermal aging.

Table 3 represents the commonly used deviations calculated for the output (concentration of carboyl) of the ANN model for the whole database. From this table, we notice that the mean of RMSE is 0.0148, mean of MAE is 0.0084 and mean of MPE is 0.3616. Therefore, we can conclude that all values are close to zero.

Table 3. Statistical analyses of the error.

RMSE			MAE			MPE (%)		
min	**mean**	**max**	**min**	**mean**	**max**	**min**	**mean**	**max**
0	0.0148	0.0869	0	0.0084	0.0869	0	36.1658	755.2632

The weight matrices and bias vectors of the NN model are listed in Table 4. Where W_H [3,11] is the input and hidden layer connection weight matrix (10 rows x 7 columns), W_L [10,1] is the hidden and output layer connection weight matrix (10 rows x 1 column), b1 and b2 are biases of the hidden layer and the output layer respectively.

Table 4. Weights and bias of the optimized NN model.

Input-hidden layer connections								Hidden-output layer connections	
W_H							b_1	W_L	b_2
W1	W2	W3	W4	W5	W6	W7			
1.1362	0.0791	0.4043	1.2250	0.7262	-2.0289	-0.6626	-0.9483	0.1559	-0.1771
0.5889	-0.0673	-0.4726	-0.2911	1.3678	-1.6002	-1.5268	0.9923	0.7080	
0.1698	0.9602	-0.6758	1.1230	0.4780	1.6448	0.4760	0.7549	3.347	
-1.0632	0.6598	0.6758	1.123	0.4780	1.6448	0.4760	-0.2780	0.1378	
2.5253	1.0107	1.3051	-0.4789	-0.5071	1.0457	3.8888	-0.9328	2.4112	
-0.7138	-0.4915	0.7486	-0.6490	-1.203	0.4305	-1.2493	1.2225	-1.0225	
1.9406	-0.8526	0.3146	0.5711	-1.3168	1.6479	0.2273	1.6515	4.1543	
-3.2269	-2.5941	-0.1455	-0.0736	-0.8075	1.9229	0.4791	0.1673	4.3461	
3.7384	0.5313	0.2135	-0.7779	-0.5852	0.0863	-0.8666	3.6966	3.82	
0.8251	0.2768	1.1312	0.7670	-1.4215	1.3552	-1.2817	2.0021	2.2051	

The Figure 3 shows a comparison between the experimental data (indicated by empty circles) and the prediction results (indicated by black stars) of the concentration of carbonyl as function of time, it shows an acceptable agreement between experimental data from the literature and the results of predictions of the neuronal model for the time tested, which shows the reliability and robustness of our developed model.

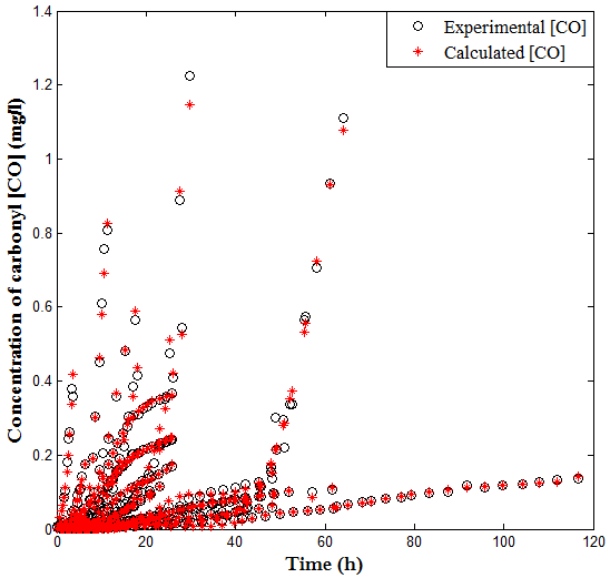


Figure 3. The comparison of experimental and calculated concentration of carbonyl [CO] as function of time.

III.1. Relative importance of the inputs

The relative importance of each input in the model was proposed by Garson (weight method) [29]. This method is based on the division of the connection weights to: Connection weights of input–hidden (W_H) and Connection weights of hidden–output (W_L). The contribution of the input variables for the neural network is given in Figure 4 where it can be seen that all selected inputs influence concentration of carbonyl. The most important factors are aging time (h) and X_4 (% w / w) with a high contribution of 22.1466% and 19.5693% for ANN model fitting.

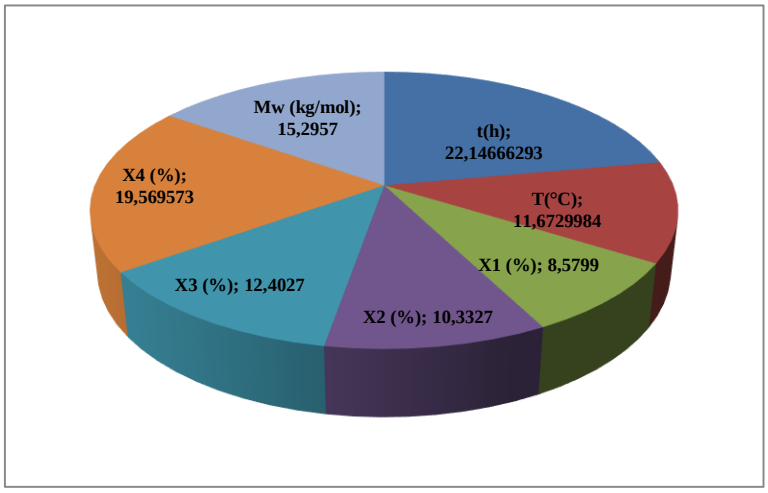


Figure 4. Relative importance (RI) of input variables

IV. Conclusion

In this paper, a suitable method for predicting concentration of carbonyl during the thermal aging of ethylene propylene diene monomer (EPDM) using an artificial neural network is described. In order to have an optimal ANN giving good modeling, we used: 7 neurons in input layer, 10 neurons in hidden layer with tangent sigmoïde activation function, and 1 neuron in output layer with tangent sigmoïde activation function. The network structure with [7"10"1] configuration trained with the Levenberg Marquardt learning (LM) algorithm. The ANN model was examined further, and both the correlation coefficient and the roots means squared error of the whole data set were R (0.9951) and RMSE (0.0148 mol/l) respectively, indicating the model accuracy and reliability. The sensitivity analysis identified that the most important variables that influence the carbonyl concentration are aging time (h) and X_4(% w / w)

List of nomenclature

[CO] –Concentration of Carbonyl

EPDM –Ethylene Propylene Diene Monomer

LM –Levenberg Marquardt learning

MAE –Mean Absolute Error

Max –Maximum

Min –Minimum

MLP-ANN –Multi Layered Perceptron Artificial Neural Network

MPE –Model Predictive Error

R –Correlation coefficient

RI –Relative Importance

RMSE –Root Mean Square Error

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