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Modeling the Young Modulus of Nanocomposites: A Neural Network Approach

Leandro F. Cupertino, Omar P. Vilela Neto, Marco Aurelio C. Pacheco, Marley B. R. Vellasco and Jose Roberto M. d'Almeida

Abstract—Composite materials have changed the way of using polymers, as the strength was favored by the incorporation of fibers and particles. This new class of materials allowed a larger number of applications. The insertion of nanometric sized particles has enhanced the variation of properties with a smaller load of fillers. In this paper, we attempt to a better understanding of nanocomposites by using an artificial intelligence's technique, known as artificial neural networks. This technique allowed the modeling of Young's modulus of nanocomposites. A good approximation was obtained, as the correlation between the data and the response of the network was high, and the error percentage was low.

I. INTRODUCTION

I N materials science, a composite is a material with two or more distinct phases; a continuous matrix phase and a dispersed filler or reinforcement phase. Nanocomposites are a special class of composites in which the dispersed phase has one or more dimensions below 100 nm. When these constituents, matrix and dispersed filler, are put together, one can achieve properties better than the two phases independently. Therefore, the use of reinforcements to improve materials resistance and lightness has been extensively done. In the past few years, nanoparticles have gained great attention and have been studied as a new successfully approach, which enables the creation of materials with low percentage of filler and greatly improved properties [1] [2] [3].

Numerical and analytical models are essential tools for studying the controlling parameters of composites as well as nanocomposites' properties. Many existent models [4] [5] [6] show that the particle's modulus, aspect ratio and volume fraction are key factors influencing the mechanical properties of composites and nanocomposites. For clay reinforced resin matrix nanocomposites, for example, these are key parameters governing the material's stiffness [7]. Besides, the exfoliation of the clays plays also an important role [8]. Particularly, exfoliation is greater with less silicate's layers, or when the inter-lamellar spacing is bigger. Both factors imply in a consequent properties enhancement of the composite.

However, despite many works on nanocomposite's properties, the understanding of the relationship between the

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macroscopic behavior and nanostructure properties is still an open question. Currently, several analytical models are based on the rule of mixtures [9], supposing a perfect load transfer between matrix and filler. These approaches work for a specific combinations of matrixes and fillers and are not so confident when overloading concentration of fillers. However, it's known that the relationship between filler volume fraction and mechanical properties of a nanocomposite can also be non linear.

Nowadays, the process used to create these new improved materials is made by exhaustive experimental work based on the specialist's knowledge, implying in a lot of physical experiments and high cost.

This work aims to find a function which can be used as a proxy of the natural behavior of matrix/nanofiller mixture, modeling the relationship between the Young's modulus and the concentration and characteristics of each filler, by means of an Artificial Neural Network (ANN). Young's modulus is a mechanic property and can be defined as the ratio of the uniaxial stress over the uniaxial strain. The proposed proxy shows good result when compared with previously developed models and can be used to reduce the number of experiments needed to develop nanocomposites, helping material specialists to decide what combination of parameters and materials should be made experimentally. This feature reduces cost and time to develop nanocomposites with useful properties.

This paper is structured as follow: section II briefly explains the nanocomposites; section III makes a quick summary about the ANN used here; section IV describes the computational approach of the developed model, section V presents the results and the comparison with other models and finally, section VI presents the conclusions.

II. NANOCOMPOSITES

The properties of composites depend on both the characteristics of the reinforcement (quantity, size, shape and distribution) and matrix. Thus, several classification schemes are available for such materials. The taxonomy defines them due to the type of matrix and the morphology of its reinforcing agents. Regarding the type of matrix it may be polymeric, metallic or ceramic. As for the reinforcement, it may be particulates (large or dispersed particles), fibrous (long or short fibers) or structural (laminated). Recently, with the inclusion of nanoparticles as reinforcing agents, a new class of materials was created and it is called nanocomposites. Nanocomposites are materials in which the dispersed phase consists of particles at the nanometer scale. Unlike the traditional approach, in which the use of particles is often used to fill volume and reduce the cost of final product, the interaction of nanoparticles with the matrix always provides an improvement in mechanical, electrical, optical and/or thermals properties. By maintaining the same concentration (mass or volume), these materials have a surface area larger than the microcomposites and, unlike them, need little reinforcement volume to significantly alter the properties. While microcomposites need reinforcements volume in the range of 30% by volume (v/v) in a nanocomposite composition up to a maximum of 10% v/v is sufficient. Above this value often present decay in performance.

In taxonomic terms the nanocomposites retain the same logic, but they are slightly different and may be classified as spherical, baculiforms or lamellar. For each of these classifications, one can cite the example of silica, carbon nanotube and montmorillonite, respectively. The nanoclays can be treated in various ways, seeking to improve the interface between the matrix and the load, such as carbon nanotubes aminofunctionalized [1].

It should be borne in mind that the intent of the nanocomposite, as regards the mechanical properties, is not to replace fiber composites, since the increase in properties when using the nanoparticle is not superior to those obtained with fibers. One of the current proposals is the hybridization between the two approaches, obtaining a material with two charges, but lighter and with superior properties.

III. ARTIFICIAL NEURAL NETWORK

Computational intelligence is a branch of computer science that develops algorithms and techniques to imitate some cognitive abilities, like recognition, learning and evolution. ANN is one of its techniques, which mimics the behavior of biological neurons and has been widely used in problems of series prediction, pattern recognition and function approximation [10]. It can be a non-linear mathematical model used to find complex relationships between inputs and targets of a function.

The neuron representation is illustrated in Fig. 1. The artificial neuron k, typically called processor element, has a set of inputs x_m (dendrites) and an output y_k (axon). The inputs are pounded by synaptic weights w_{km} (synapses), which determines the effect of entry x_m on processor k. These weighted inputs are summed, providing the internal potential of the processor (v_k) . The output or activation state y_k of the element processor k is finally calculated using an activation function $\varphi(.)$, typically a sigmoid function.

An ANN is composed by interconnected neurons and may have different types of topology. In this paper, we used multiple layers of neurons where the outputs of each neuron of a layer are used as inputs of the next one. This topology in known as Multi-Layer Perceptron (MLP) [10] (Fig. 2).

ANN with only one hidden layer is able to approach, with a specific precision, any continuous function [11] [12]. The fundaments for this statement is that any continuous



Fig. 1. Representation of an artificial neuron.



Fig. 2. ANN topology presented with four layers: an input, two hidden and an output. Each neuron is represented as a circle.

function, limited within an interval, can be approximated as a linear superposition of sines, which can be mapped by the neurons of the hidden layer, since the activation function of the neurons is the log-sigmoid one. To approach a nonlinear function, two hidden layers are needed.

In order to achieve a good input/output relationship, a training algorithm adjusts the ANN's weights through an error minimization between the network output and the target. Many such input/target samples, known as patterns, are needed to train a network. This data is divided into three sets: training, validation and test. The first one are the patterns presented to ANN in order to optimize the network's weights, minimizing the error. The validation set intends to give the generalization ability to the model, avoiding overfitting. Finally, the test set, with a database never presented before to the ANN, is used to measure the effective quality of data fitting. Fig. 3 shows the optimization procedure during the training stage.

The main advantage of an ANN over other interpolation methods is its capability of modeling systems with a very strong non-linear behavior. Even though, for composites and nanocomposites, it's not possible to extract deep physical insights on the interfacial interaction between matrix and filler from the ANN. However, one can obtain trends that can help in constructing new physical models or in understanding



Fig. 3. Training algorithm schema.

the composite manufacturing process, as already made for the growth of other nanostructures, such as quantum dots [13].

IV. COMPUTATIONAL APPROACH

A. Database

The study of polymeric nanocomposites is being extensively done in a lot of domains like physics, chemistry, materials science, engineering and even dentistry. Properties like hardness, drying time, stress and thermal and electrical conductivities are analyzed, and researches sought to tailor these properties. Although the number of data published in the literature is high, the different goals cause a heterogeneous availability of properties' data. In this project we decided to initially work with the Young's modulus, since this property is one of the most studied and that presents a greater number of available data. It is also our interest to work with other properties, but this requires a partnership with other experimental groups, in order to obtain the necessary data.

The data used in this work was collected from various articles published in the recent years. These data include the use of thirteen matrices and eighteen nanofillers. Table I lists the components supported by the ANN and their respective references. Some of the data were taken from graphs which, although includes a noisier measure, enable the study of a greater range of materials.

Five parameters were chosen as inputs of the network to represent the behavior of the elastic modulus (Young's Modulus): the matrix material, the filler material, filler's weight percentage (%wt), filler's diameter and filler's aspect ratio. The aspect ratio can be defined as the ratio of its longer dimension to its shorter dimension. Each of the matrices and fillers were referenced as a class. When necessary, new classes of matrices and fillers can be introduced on the creation of the neural network.

The data collected was filtered to remove some outliers that could difficult the network learning. Then, the inputs and outputs were normalized as shown in table II. The filler concentration and relative Young's modulus where normalized in a different way in order to make possible a little extrapolation of database's values.

B. Artificial Neural Network

The total number of data used in the development of the network was 151. On the data selection for the training set,

TABLE I MATRICES AND FILLERS USED ON THE ANN.

Matrix	Filler	Ref.
UP	MMT (Cloisite 30B)	[2]
UP	MMT (Cloisite 25A)	[2]
UP	MMT	[2]
UP	MMT Na+	[2]
PMMA	MMT (Cloisite 20A)	[16]
PMMA	CNF (PR-21-PS)	[3]
PMMA	CNF (PR-24-PS)	[3]
PP	CaCO ₃	[22]
PP	CaCO ₃	[19]
PP	SiO ₂	[7]
iPP	CaCO ₃	[14]
E-glass-PP	MMT (Nanomer 1.28E)	[17]
PLLA (moulded)	HAP	[18]
PLLA (moulded)	g-HAP	[18]
PLLA (annealed)	HAP	[18]
PLLA (annealed)	g-HAP	[18]
PA6	SiO ₂	[7]
PA6	MMT	[4]
PU	CNF	[20]
SBR	Clay	[21]
NBR	Clay	[21]
CNBR	Clay	[21]
Epoxy	TiO ₂	[15]
Epoxy	Carbon Black	[1]
Epoxy	DWCNT	[1]
Epoxy	DWCNT–NH ₂	[1]
Epoxy	SWCNT	[23]
Epoxy	DWCNT	[23]
Epoxy	MWCNT	[23]
Epoxy	MWCNT-NH ₂	[23]
Epoxy	Carbon Black	[23]
Epoxy	DWCNT-NH ₂	[23]

TABLE II NORMALIZATION OF THE ANN DATA.

Property	Normalization	
Matrix class	Real [0, 1]	
Filler class	Real [0, 1]	
Filler diameter	Real [0, 1]	
Filler aspect ratio	Real [0, 1]	
Filler concentration	Real [0, 0.9]	
Relative Young modulus	Real [0.1, 0.9]	

all matrices and fillers were fixed as members, summing up 113 (75% of data). For the validation set, 23 (15%) patterns were randomly selected. The test set had 15 (10%) different patterns (not used in the training or validation sets), 7 of which refer to a special combination of matrix/filler. In the latter one, a new combination of matrix and filler was proposed, i.e., on the training and validation sets we had samples of PP/CaCO₃ and PA₆/SiO₂, so on the test set we evaluated a different composite (PP/SiO₂) that was not used neither in the training nor in the validation sets. These were chosen in order to evaluate the real predictive power of the developed system.

The training algorithm chosen was based on the Levenberg-Marquardt minimization algorithm [24], since it has a fast convergence, which contributes to the learning speed of the ANN. The error metric used was the root mean

squared error (RMSE), so the best network configuration was chosen based on which configuration gave the lowest RMSE on the validation set, avoiding over fitting and allowing generalization.

Once it's known that the relationship between filler volume fraction and mechanical properties of a nanocomposite can be non linear, we used a two layer MLP network. In order to select the best network architecture, the number of neurons on each layer was varied from 3 to 12 on the first hidden layer and from none to 4 on the second one. As the initial weights are randomly selected, we used the average RMSE of 100 runs to evaluate each network architecture. Besides, the activation function of the hidden and output layers was varied between hyperbolic tangent, linear and saturating linear transfer functions.

V. RESULTS AND DISCUSSION

In order to evaluate the results of the ANN two different metrics were used: the correlation coefficient (R value) and the mean average percentage error (MAPE). The first one indicates the strength and direction of a linear relationship between two random variables. In other words, it measures how well the output data variation is explained by the neural model. The second metric measures how close the predicted results are from the actual data.

The configuration of the best network found has the activation function of the hidden layer as the hyperbolic tangent and for the output layer, linear. The numbers of neurons on the first and second hidden layers are 10 and 2, respectively.

The results of the neural network showed a great correlation in all the sets used. Values of 0.994, 0.999 and 0.999 were obtained for the training, validation and test sets, respectively. Values above 0.9 indicate a good agreement between experimental and predicted values. Fig. 4 shows a linear regression between the network outputs and the targets. Showing that, as the target and output values tend to be the same, the model is well adapted to the used data.

In terms of MAPE, the results showed values of 4.17%, 3.63% and 5.06% for the training, validation and test sets, respectively. Once the experimental measured error is commonly around 10%, and the network error is below this range, the results obtained are acceptable. Fig. 5 shows the output of the data used on the training, validation and test sets (\circ) and the achieved response of the ANN (.). It can be clearly seen that the predicted behavior of the network is quite close to the experimental data. It's important to notice that the first seven data of the test set consists of a mixture of matrix and filler which has never been presented during the training and validation of the network. This indicates that the network can predict the Young's modulus of an unknown mixture, once these matrix and filler were used separately on the training set. This feature enables the researcher to predict how the mixture of two different materials, not done experimentally, should work.

In order to compare the ANN with different models studied [9], the filler fraction was converted from weight to volume fraction and it's concentration's scale is shown in terms of both, weight and volume fractions (see Fig. 6). In [9], the author uses four different composites sets to discuss six distinct models. In each of these sets the matrix and filler remained the same, being the weight fraction of the filler, the only variable changed. Two of these models are the rules of mixtures of Voigt and Reuss, 1 and 2, respectively, where E, E_d and E_c are the Young's modulus of the composite, disperse and continuous phase respectively, and ϕ is the dispersed particle volume fraction. Although these two models don't have a very high accuracy, the Reuss' model is the lower limit of the Young's modulus, as the Voigt's model is the upper limit.

$$E = \phi E_d + (1 - \phi)E_c \tag{1}$$

$$\frac{1}{E} = \frac{\phi}{E_d} + \frac{(1-\phi)}{E_c}$$
(2)

The other equations are described in terms of the bulk (K) and shear (G) modulus of the composite, and its dispersed and continuous phase (3, 4 and 5). These equations are differed by the f function as shown in 6-9.

$$E_r = \frac{E}{E_c} = \left[\frac{9G_r K_r}{6(1+\nu_c)K_r + 3(1-2\nu_c)G_r}\right]$$
(3)

$$K = \left[\frac{4G_d(K_c - K_d) + fK_d(3K_c + 4G_d)}{3(K_d - K_c) + f(3K_c + 4G_d)}\right]$$
(4)

$$G = \left[\frac{G_g(G_c - G_d) + fG_d(G_c + G_g)}{(G_d - G_c) + f(G_c + G_g)}\right]$$
(5)

where K_r and G_r are defined as K/K_c and G/G_c , respectively. The four different equations used for f are given below.

$$f = \exp(\phi) \tag{6}$$

$$f = (1 - \phi)^{-1} \tag{7}$$



Fig. 4. Correlation between target and network outputs of the training, validation and test sets together

$$f = \exp\left[\frac{\phi}{1 - \frac{\phi}{\phi_m}}\right] \tag{8}$$

$$f = \exp\left(1 - \frac{\phi}{\phi_m}\right) \tag{9}$$

where ϕ_m is the maximum packing volume fraction or percolation-threshold of particles.

Figures 6-9, plot the rule of mixture equations as well as the relative modulus approach. For the relative modulus approach all the four proposed equations for f were used. The resultant plots using f from 6, 7, 8 and 9 are referred to as models I, II, III and IV, respectively.

In Fig. 6, a nanocomposite of organoclay (bis(hydroxyl ethyl)-(methyl)-rapeseed quaternary ammonium organoclay) matrix wiah a nylon-6 filler [4] is used to compare the existing models and the ANN. In this case, a low concentration of filler is used (maximum volume fraction of 3%). As can be seen, in this concentration range, the variability of the data is almost linear, and model III gives the best fitting.

Fig. 7 shows a montmorillonite/SBR (Styrene Butadiene Rubber) [21] composite set, where a different result is noticed. With greater volume fraction and a nonlinear behavior, the ANN is able to follow the trend of the experimental data where the other models fail.

In Fig. 8 and 9, composites of montmorillonite clay with NBR (Nitrile Rubber) [21] and CNBR (Carboxylated Acrylonitrile Butadiene Rubber) [21] matrices are respectively shown. Once again a nonlinear behavior is observed and the ANN's ability to follow the trend of the experimental data is the best one for both sets. For these cases, a limitation on models III and IV is noticed. As the particle volume is higher than the percolation-threshold volume fraction (ϕ_m , defined as 0.12 and 0.10 to models III and IV, respectively), the models can't correctly predict the composite's behavior anymore. This is easily observed on 8 and 9 and evinced in Fig. 8 and 9. Moreover, by using a maximum of 0.55 wt% in the training set of the neural network, this network is able to



Fig. 5. Output results of the ANN: target (0) and network (.) outputs.



Fig. 6. Comparison of models for the organoclay (bis(hydroxyl ethyl)-(methyl)-rapeseed quaternary ammonium organoclay)/nylon-6 set.



Fig. 7. Comparison of models for the montmorillonite/SBR set.

predict any value at concentrations till the maximum training value.

From the above results it can be said that, in general, the ANN has a better fitting to the experimental points and thus better represents the Young's modulus variation than the analytical models I-IV, which present a good behavior just for composites with low volume fractions of filler. The poor result achieved by the ANN for the composite organoclay (bis(hydroxyl ethyl)-(methyl)-rapeseed quaternary ammonium organoclay)/nylon-6 set may have been caused by a scarce quantity of data with low weight fraction of filler used on the development of the network.

Finally, having a well developed ANN one can get an idea of the variation of the relative modulus (E/E_c) as function of



Fig. 8. Comparison of models for the montmorillonite/NBR set.

any of the synthesis parameters presented to the network. As an example we have varied the concentration and diameter of a particle. Fig. 10 shows the level set output of the network corresponding to a material composed by a matrix of poly(methyl methacrylate) (PMMA) with carbon nanofibers (CNF) with an aspect ratio of 100. It can be observed that smaller the particle diameter, faster the modulus increases, for example, a composite having a diameter of 100 nm, needs 30 wt% to achieve a relative modulus of 4, while another one, with a diameter of 600 nm needs 40wt% to achieve the same result. In this case, with a small filler concentration a good response of Young's module is obtained. On the other hand, larger particles need a higher concentration to obtain a similar result. Besides, when the smaller particles achieve high concentrations the composite modulus decreases. This can be explained by clustering of these particles, which will act as if they were larger. Therefore, a micro-reinforced composite is formed, instead of a nanocomposite.

VI. CONCLUSIONS

The process of creation of new composite materials is considered to be very expensive. It's necessary to perform several experiments in order to select a suitable combination of matrix/filler to achieve some determined objective. Normally this issue is done by trial and error.

The use of computational techniques to predict the behavior of composites, given their synthesis input parameters, was proposed in this paper. The network developed presents high correlations values of 0.994, 0.999 and 0.999 for the training, validation and test sets, respectively. Besides, MAPE values of 4.17%, 3.63% and 5.06% for the training, validation and test sets, are low, confirming the good correlation of the network with the experimental data.

The network developed was compared with analytical models available on the literature. The ANN's results show



Fig. 9. Comparison of models for the montmorillonite/CNBR set.

a better agreement with the data set than the other models. These results prove that through the use of ANN; it is possible to predict if a certain matrix/filler pair can give the desired property, preventing the researcher to spend materials, time and money. With the procedure here developed, one can gather enough data to cover a larger amount of matrix and filler. Moreover, other properties can be inferred too. The future goal is the construction of a simulator able of predict several nanocomposites properties with decreasing time and cost.

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Fig. 10. Inference of PMMA/CNF composite.

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