Tribological Behaviour Prediction and Optimisation for an Epoxy Clay System Based on Mechanical and Thermal Properties

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The prediction of polymer properties, based on its composition, it is a complex problem with no easy method to obtain directly and accurately results. Among the tribological properties, the friction coefficient and wear rate are the most interesting ones. The polymers based on epoxy resin, with clay as filler, show different properties depending on the clay concentration. This paper presents an analysis of the polymer properties variation with its filler concentration. Due to the tribological processes complexity, mechanical and thermal properties must be taken into account. The aim of this study is to find an optimal concentration value, with minimal influence on polymer properties, using neural network models. All value properties were used in order to optimize and predict the composite properties.

Keywords: polymer properties, tribology, filler concentration, neural network, prediction

The idea of using the fillers for reducing the price or for modifying the properties of epoxy resins is not new but still represents a challenge in establishing the optimal dispersion in composite matrix. The fillers can be categorized according to their composition, source, morphology and/or function. Some trussing agents have the tendency of agglomeration and, in order to avoid this, there are used in pre-polymeric mixture substances so-called dispersion agents. These substances can be either liquids or powder. The filler particle agglomeration in matrix phenomena leads to the decrease of the mechanical properties in this way creating faults into the final composite. The most used dispersion agents are: starch, clay and soapstone. The properties of the final composites are influenced by the properties of the components: shape filler phases, system morphology and polymer-filler interface interactions.

The dispersion problem can be solved by different methods, either physical or chemical. The physical blending methods use mechanical force while the chemical ones work by activating or modifying the additive's surface. It can be used physical, chemical or combination of these methods [1, 2].

The mixture of a filler with a resin results in a new material with changed properties. Some of these changes are advantageous while others are less favorable.

Usually, a Neural Network is described as an interconnected assembly of simple processing elements, units or nodes, whose functionality is loosely based on the animal neuron. The processing ability of the network is based on the unit connection strengths (weights) obtained by a process of adaptation to, or learning from, a set of training patterns [3-5].

In the present study two artificial neural models were used: first provides the filler concentration for known values of composite properties; and second allows the wear rate prediction based on filler concentration and sliding velocity.

Neural models also allow possibility to analyze the influences between composite properties and filler concentration. Based on this analyze, an optimization of composite recipe can be done.

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Experimental part

Materials and methods

The Epoxy system RE 4020 – DE 4020 was used as matrix to form particulate composites with Clay as filler, with a volume ratios starting from 1% to 30%. The epoxy resin has been obtained by reacting epichlorohydrin (propylene chloride) with bisphenol A. The reaction proceeds in two steps, first step - forming diglycidyl ether bisphenol A (DGEBA), which is called component A. The second one is strengthening the component A (DGEBA) with cycloaliphatic amine type nonylphenol - component B.

Today the polymer-clay composites are widely used in automobile industry. Clay, as layered structured material, gives to the composite the rise of hardness and the weight decrease, being used for trussing the composite in order to obtain cheap and weightless materials.

The clay powder is known as layered crystalline structures. Inside the matrix, these crystalline structures affect the mechanical properties, facilitating the stress transfer between the crystalline phase and the amorphous one [4, 6, 7].

In order to obtain an optimized polymer, it is necessary to find a clay concentration, which does not modify the base resin properties. With this aim, several mixtures were prepared, with clay concentrations in 1 - 11% range, stepping 1%; 11 - 15% range, stepping 2% and 15 - 30% range, stepping 5%.

In clay-epoxy composites, the compatibility between resin and powder must be taken into account. Clays are generally highly hydrophilic, while the majority of polymers are hydrophobic. Incompatibility between hydrophobic polymers and hydrophilic clays is the main reason of clay agglomeration formation in the polymer matrix. As consequence, clay surface modification is necessary in order to become compatible with the polymer [8, 9].

Before mixing process, clay powder was subjected to heat treatment for 6 h at 80°C, in order to remove most of the surface hydroxyl groups and decrease the powder's humidity content. Due to the treatment, the link between the additive and matrix treatment becomes better, thus

Table 1
TEST CONDITIONS

<table>
<tr><th colspan="5">To evaluate friction coefficient</th></tr>
<tr><td>Sliding time [s]</td><td>Normal force [N]</td><td>Distance [m]</td><td colspan="2">Sliding velocity [m/s]</td></tr>
<tr><td>1800</td><td>10</td><td>1700</td><td colspan="2">0.942</td></tr>
<tr><th colspan="5">To evaluate abrasive wear rate</th></tr>
<tr><td>Normal force [N]</td><td>Distance [m]</td><td colspan="3">Sliding velocity [m/s]</td></tr>
<tr><td>2</td><td>50</td><td>0.27</td><td>0.41</td><td>0.83</td></tr>
</table>

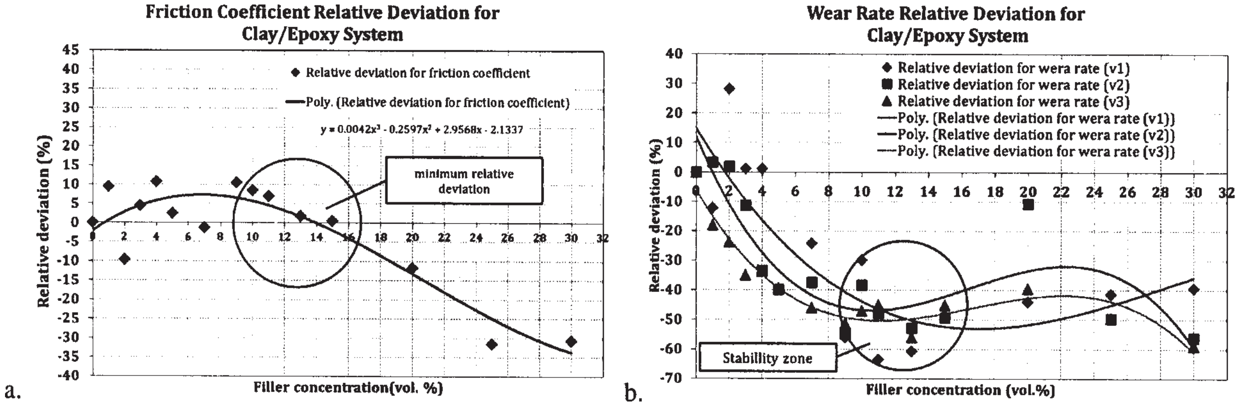

Fig.1 a) Friction coefficient relative deviation. b) Wear relative deviation

improving the mechanical properties of the composite [8 - 11].

This study tries to identify dispersant's concentration, which inside the composite does not change the properties of the resin.

Tribological Properties

For friction coefficient and wear rate assessment, a pin-on-disc fixture was used, on Universal Tribotester UMT2 (CETR®, SUA). The pin was made of filled material and the disk of solid steel - for friction coefficient; and covered with abrasive paper - for wear rate. Test conditions are presented in table 1.

After the pin-on-disk testing, the relative variation of friction coefficient was obtained (fig. 1 (a). The friction coefficient values: 20, 25 and 30% clay ratio are half than the pure resin, as it can be observed in figure 1 (a). A lower relative variation was observed in 9-15% domain, identified as minimal aberration area.

In abrasion wear tests, a lower wear rate of composites was obtained, comparing with pure resin. The stability area of wear rate (around 8-15 and 25%), indicate the homogeneous dispersion of clay powder into the matrix (fig. 1 b).

Mechanical Properties

For mechanical properties assessment, the three points bending method was chosen. The tests were performed according to ASTM D 790, ISO 178 and SR EN ISO 14125 standards, on a Testometric M 350 5K tester [8, 9]. Load application speed was 1 mm/min; the distance between the supports was of 22 mm. These were evaluated the breaking stress, Young modulus and breaking energy. The mechanical properties of the relative deviation, (fig. 2 a), has a minimum value around 10%, leading to the conclusion that this clay concentration does not produce big differences in mechanical properties comparing to pure resin and making the clay suitable as filler agent.

Thermal Properties

As thermal properties, thermal expansion and specific heat were chosen.

For measuring linear thermal expansion coefficient was used thermo-mechanical analyzer TMA-SDTA 840 from Mettler Toledo, the method is governed by ASTM E 831 [12]. For each type of material were tested three samples.

The DSC 1 from Mettler Toledo was used to determine specific heat. For each concentration were tested three samples, so the specific heat is the average of three values specified for temperature range. Due to the fact, that the clay shows a better thermal stability, the mixing with epoxy resin provide to the composite a lower value of thermal expansion coefficient comparing to the pure epoxy (fig. 2 b).

From each clay concentration, three samples were tested, looking for a minimal aberration domain. This was found around 9, 10, 11 and 13%, as it is presented in figure 2 b.

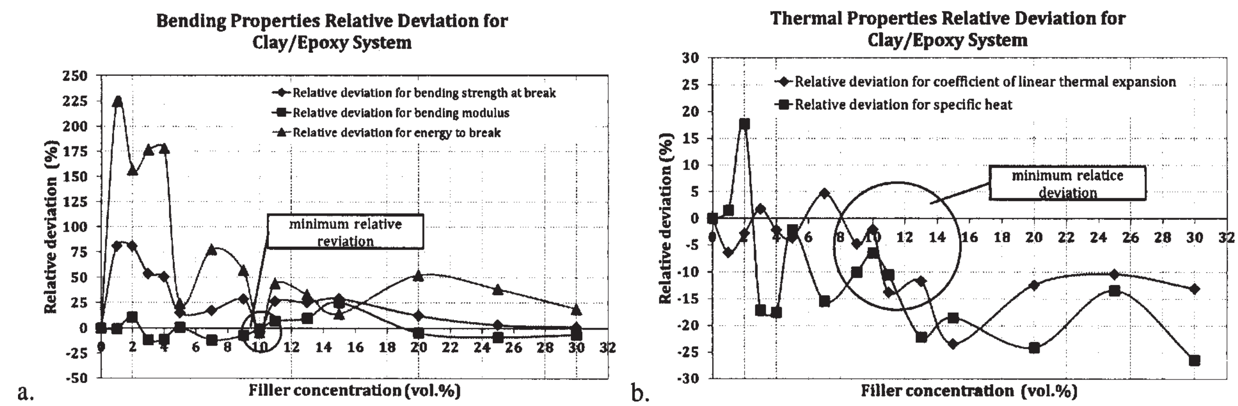

Fig. 2. a) Bending properties relative deviation. b) Thermal properties relative deviation

Parameter	Position network I
Filler Concentration [%]	Output
Friction Coefficient (μ)	Input
Wear Rate v1 [Nm/mm^3]	Input
Wear Rate v2 [Nm/mm^3]	Input
Wear Rate v3 [Nm/mm^3]	Input
Strength at Break [MPa]	Input
Young Modulus [MPa]	Input
Energy at Break [Nm]	Input
Coefficient of Thermal Expansion [ppm/°C]	Input
Specific Heat [J/g°C]	Input

Table 2
NEURAL NETWORK MODEL I INPUTS, OUTPUT AND OPTIMIZATION

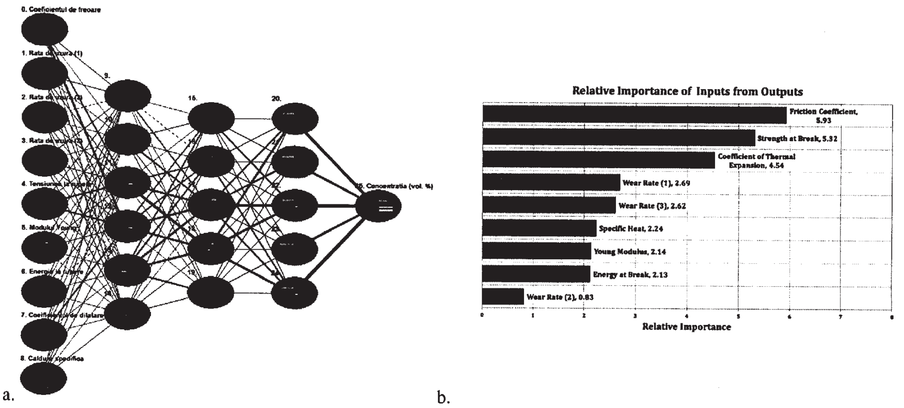

Fig. 3. a) Neural network architecture I; b) Input's importance for Network I.

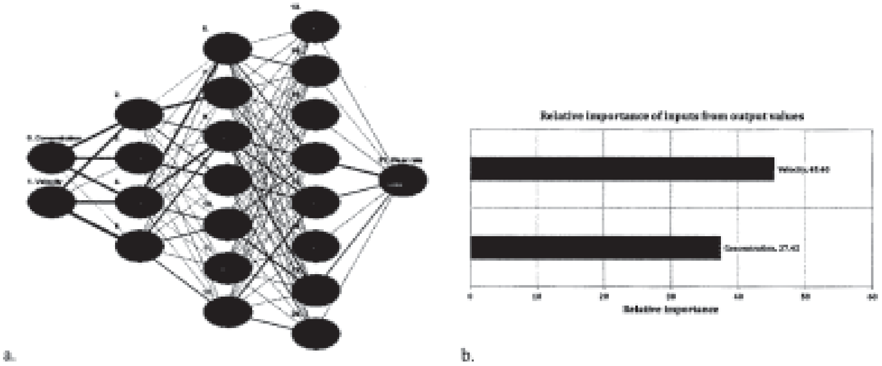

Fig. 4. a) Neural network architecture II; b) Input's importance for Network II.

The interval between 0-8% clay concentrations has been characterized by the instability of the evaluated thermal properties. The concentration from 10-30% clay powder has stabilized, resulting in lower values compared to resin without fillers.

Results and discussions

Artificial network based prediction for clay concentration

As it was mentioned before, two network models were built. First one was used to establish the optimal clay concentration, which does not significantly affect the mechanical properties of the composite by comparing it to the pure epoxy resin; a backpropagation type neural network was used. ANN was also used to optimize the tribological properties for clay/epoxy based composite. It was necessary to put in ANN model physical and mechanical properties, for making a complete characterization of the composite material, which is very important in tribological processes. It was created an artificial neural network by using the EasyNN simulator software.

The obtained network architecture, optimized by internal algorithms provided by the software package, has nine inputs - the values of previously measured properties. As output, the filler concentration was obtained (table 2). Three hidden layers complete the network, (fig. 3 a).

The learning target error was settled at 0.010% and this value was obtained after 181 training cycles.

After the learning stage, the network model can be used for the prediction of the optimal filler concentration. Using for inputs the experimental measured properties values for bulk resin, the network response was 9.7%, as filler concentration that not modify the composite properties comparing with bulk resin. Taking into account that during the experiments the 10% concentration value was established as optimal, the network can be considered valid.

The built network model can now be used for analyzing the filler concentration influence on tribological properties of the final composite. In table 2 are presented the composite properties values for optimal, maximum and respective minimal filler concentration. It is obvious that higher filler concentration leads to an improvement of tribological properties while lower concentration leads to worst properties.

Analyzing the inputs influence on output values, a hierarchy can be established for composite properties versus filler concentration. As can be observed in figure 3 (a), the most influencing inputs are: friction coefficient, breaking stress and thermal expansion coefficient.

In figure 3 (b) are presented the hierarchy of inputs-output links strength, as Importance influences in neural

Parameter	Position network II	Optimization with Network II	
Filler Concentration [%]	Input	6.6	21.9
Velocity [m/s]	Input	0.594	0.3672
Wear Resistance [mm^3/Nm]	Output	4.8229	20.4022

Table 3
NEURAL NETWORK MODEL II INPUTS, OUTPUT AND OPTIMIZATION

network model. It is obvious that the most powerful links are established between the friction coefficients, the strength at break, the coefficient of thermal expansion, and the wear rates, to output – the filler concentration. This leads to the conclusion that these properties are the most affected by little variations of filler concentration.

Second ANN model was created with the aim to predict the material's recipe with the best wear resistance properties. The corresponding obtained network II architecture has two inputs - the values wear resistance measured properties and one output – filler concentration. Three hidden layers complete the network, (fig. 4 a).

The training target error was settled at 0.010% and this value was obtained after 60123 training cycles. In figure 4 (b) are presented the analyses of relative importance.

As can be observed in figure 4b, the most influencing parameter on wear rate is sliding velocity.

Another use of the neural model is the optimization of filler concentration, for a specific velocity, in order to obtain a maximum wear resistance (table 3).

Conclusions

After testing several combinations of epoxy resin - clay filler, some conclusions are to be drawn as it follows.

All studied properties of clay composites show a strong dependence on powders concentration. Friction coefficient shows an improvement for a clay concentration of 20, 25 and 30%, comparing with the pure resin. A lower relative variation was observed for 9-15%. During wear tests, a lower wear rate is obtained (comparing to the pure resin) for all the tested composites. The minimum for mechanical properties was observed at 10% clay concentration as it shows relative variation. For the thermal properties, minimum results were for 5, 9, 10 and 13% clay concentrations, as it can be observed studying relative deviation.

Based on neural networks capability to create links between input-output data couples, two models were created. The aim was to analyze and optimize the filler concentration in epoxy matrix based composites, from tribological properties point of view.

The neural network models are efficient tools for optimal clay concentration establishing with the aim to get better composites without non-significant mechanical properties modifications. The value 9.7% was obtained as neural model response while 10% was the experimental established value.

It is important to observe that both neural models presented above show the same behavior of tribological properties for the analyzed polymer, leading to the conclusion that the phenomenology of filler influence on composite properties is correctly implemented by the artificial networks.

Another conclusion can be the fact that the neural network models are suitable tools for mechanical and tribological properties optimization, for desired application and working conditions.

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