Study on the prediction of the mechanical properties of polymeric composites with polypropylene and graphene using artificial neural networks

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ABSTRACT

Nanocomposites are promising new materials capable of achieving good physical properties with low weights and easy processing. Understanding how each component and its properties affect the final product's mechanical capability is essential to improve performance and defining the optimal production route. In this context, mathematical modeling can be a valuable tool to increase efficiency and reduce the number of experiments and resources used. The present work proposes the development of an Artificial Neural Network (ANN) to predict the tensile strength of nanocomposites composed of polypropylene (PP) and graphene. The database was built with literature data published in research articles, using the melt flow rate, graphene content, compatibilizer content, initial tensile strength, and graphene's surface area as input variables. Different architectures were tested, changing the number of hidden layers, the number of neurons in these layers, and the activation functions. The best network architecture consisted of a single hidden layer with 5 neurons using the sigmoid activation function. The prediction of the final tensile strength had satisfactory results, reaching an R^2 value of 0.83 and a mean square error of 8.47.

INTRODUCTION

Due to new demands and regulations, the automotive industry's requirements for reducing energy consumption and gas emissions continue to increase. One possible solution may be substituting the standard heavy materials for lighter ones that can withstand the same stress.[1], [2] Different studies have shown that in the case of a 10 % in car weight reduction, fuel consumption, and emissions may be decreased by 6 % to 8 % and 5 % to 6 %, respectively.[3]

Using polymer help to reduce the vehicle weight, impacting directly in the reduction of fuel consumption and gas emissions.[4] Polyolefins are polymer materials that are vastly used in the automotive industry, both in interior and exterior applications. Among this group, polypropylene (PP) is probably the most recognized due to its affordability and easy processing.[5] However, PP has mechanical and thermal properties inferior to the demands that are required for some applications. Therefore, it can be reinforced with synthetic fibers, natural fibers, carbon nanotubes, and graphene and derivates.[6], [7]

In this context, polymeric composites are materials that can be used in various applications as they have good mechanical performance, are easy to process, and are lightweight.[8] The performance of these materials can be increased in different ways, such as by adding hard materials (ceramics, glass fiber, or silica), using chemical additives, and making coating layers.[9] Moreover, their performance may be further improved by adding nanomaterials to the polymeric matrix. Due to their unique structure and nano size, various nanoparticles are widely used as reinforcing fillers for advanced applications.[10]

Among the different filler nanomaterials, graphene has raised much attention in recent years due to its exceptional mechanical, thermal, and electrical properties. This material is composed of carbon atoms bonded in a 2D honeycomb structure. Incorporating this nanomaterial in polymer matrices may enhance their properties even using low contents, thus, not increasing the weight.[11], [12]

However, adding a graphene material to the formulation does not imply that the properties are easily increased.[13] One of the main issues regarding the use of graphene in polymeric composites is its dispersion in the polymer matrix.[14] Due to its lack of surface functional groups, graphene has only a limited ability to disperse in solvents. A good dispersion in polymeric composites only occurs when the graphene content is low.[15] If the content is continuously increased, it may lead to the formation of graphene aggregates, which do not add the desired properties.

Trying to predict the polymer composites' properties is not easy. Many factors can impact the final characteristic of a material, its initial state and properties, and the conditions used in manufacturing. Artificial neural networks (ANN) are models with a great capacity to adapt to the data received. Therefore, this modeling technique may be applied to predict polymer composites with PP and graphene. Zakaulla and contributors [16], [17] used ANN to predict the properties of polycarbonate and graphene polymer composites and polyetheretherketone (PEEK) and graphene, reaching excellent results in both cases. Khanam et al. [18] used a similar model to predict the thermal and mechanical properties of linear low-density polyethylene and graphene but using also processing conditions to predict the final characteristics of the composite. Kazi et al. [19] used an ANN model to predict the optimal fiber concentration for PP and cotton fiber composites using the desired characteristics as inputs for their model.

In the present study, a neural network model was proposed to predict the mechanical properties of PP and graphene composites using the initial characteristics of the materials and their ratio in the mixture (melt flow rate, initial tensile strength, surface area, graphene content, compatibilizer content). Different ANN architectures were tested, changing the number of hidden layers, the number of neurons, and the activation functions in each hidden layer. The best network was selected considering performance metrics, mean squared error (MSE), and determination coefficient (R^2).

METHODOLOGY

DATA ACQUISITION

The database was built using research articles that evaluated the mechanical performance of PP and graphene nanocomposites. The macro search was performed using "polypropylene", "graphene", and "nanocomposite" as keywords in different repositories. After that, only articles containing information regarding the composition and mechanical properties of the nanocomposites were considered, resulting in 17 research articles. The data was extracted from each article (composition and tensile strength) to train and test the artificial neural network model. The data were divided into training and testing using a 70:30 ratio.

MODEL DEVELOPMENT

The model was built to predict the nanocomposite's tensile strength based on five variables: melt flow rate, initial tensile strength, graphene's surface area, graphene content, and compatibilizer content. The model consisted of a feedforward neural network varying its number of hidden layers, the number of neurons in each hidden layer, and the activation function of the hidden layers to reach the topology with the best results. Figure 1 represents the tested ANN used in the present work.

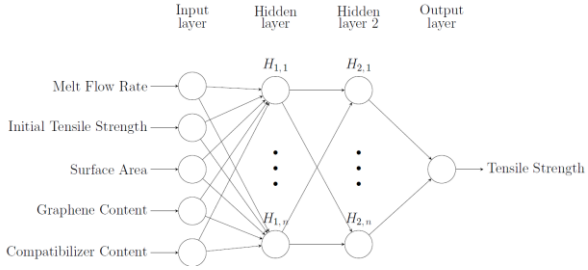


Figure 1: Generalized representation of the ANN used in the present work.

The networks were tested using the Tensorflow package in Python 3. The optimizer used was the Adam class (stochastic gradient descent method) with the default settings, except for the learning rate, which was set to 0.01. The objective function used to train the network was the mean squared error function (MSE – Equation 1). The maximum number of iterations was 1000 with a callback if the error function value did not decrease for 50 consecutive iterations.

Aside from the objective error function, the determination coefficient (R^2 – Equation 2) was also used for performance evaluation. The tested activation functions were sigmoid (Equation 3), hyperbolic tangent (tanh – Equation 4), and rectified linear unit (ReLU – Equation 5).

$$\text{MSE} = \frac{\sum_{i=1}^{n}(x_i - \hat{x_i})^2}{n} \tag{1}$$

$$\text{R}^2 = \frac{\sum_{i=1}^{n}(\hat{x_i} - \bar{x})^2}{\sum_{i=1}^{n}(\hat{x_i} - \bar{x})^2 + \sum_{i=1}^{n}(x_i - \hat{x_i})^2} \tag{2}$$

where x_i is the experimental value, $\hat{x_i}$ is the model value, $\bar{x}$ is the average value, and n is the sample size in the input layer.

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

$$\tanh(x) = \frac{e^{x} - e^{-x}}{e^{x} + e^{-x}} \tag{4}$$

$$\text{ReLU}(x) = \begin{cases} x, \text{ if } x > 0 \\ 0, \text{ if } x < 0 \end{cases} \tag{5}$$

where x is the value from the previous layer

RESULTS AND DISCUSSION

DATA ANALYSIS

Figure 2 shows the pair plots of the independent (melt flow rate, initial tensile strength, surface area, graphene content, and compatibilizer agent) and the dependent (tensile strength) variables. Most of the independent variables show only one significant peak, which can result in a non-complete understanding of the influence of each variable in the output.

The melt flow rate and graphene content show only a single peak. The first shows a significant concentration of values between 0 and 20 g.10 min^{-1} and the latter with values between 0 and 5 wt.%. The melt flow rate directly impacts the processing of the composite. PP with higher values tends to be easier to process and add components as reinforcements. The lack of data of higher values may induce that there is no relation between the tensile strength and the melt flow rate, as can be seen in the last row of the first column in Figure 2 and also in the correlation matrix in Figure 3, with a correlation value of 0.049. Similar behavior is seen for the graphene content, with a correlation value of -0.044. Figure 4 shows that these variables have too slight variation, presenting a lot of outliers. Therefore, their actual representation may be compromised by the lack of data.

Regarding the surface area and compatibilizer content, there are smaller peaks besides the highest one. This might help further understand the influence of such variables on the model's output. However, both behave the same as the ones mentioned above. They show almost no direct correlation with the tensile strength (last row of Figure 2 and Figure 3), despite showing an apparent more distributed data. The correlation values are still very low for both these variables, -0.024 and -0.15 for surface area and compatibilizer content, respectively. Unlike the melt flow rate and graphene content, these two variables show no outliers (Figure 4), indicating that the data may be better distributed. Nevertheless, the median value still is very low, pulling the data toward a concentrated region.

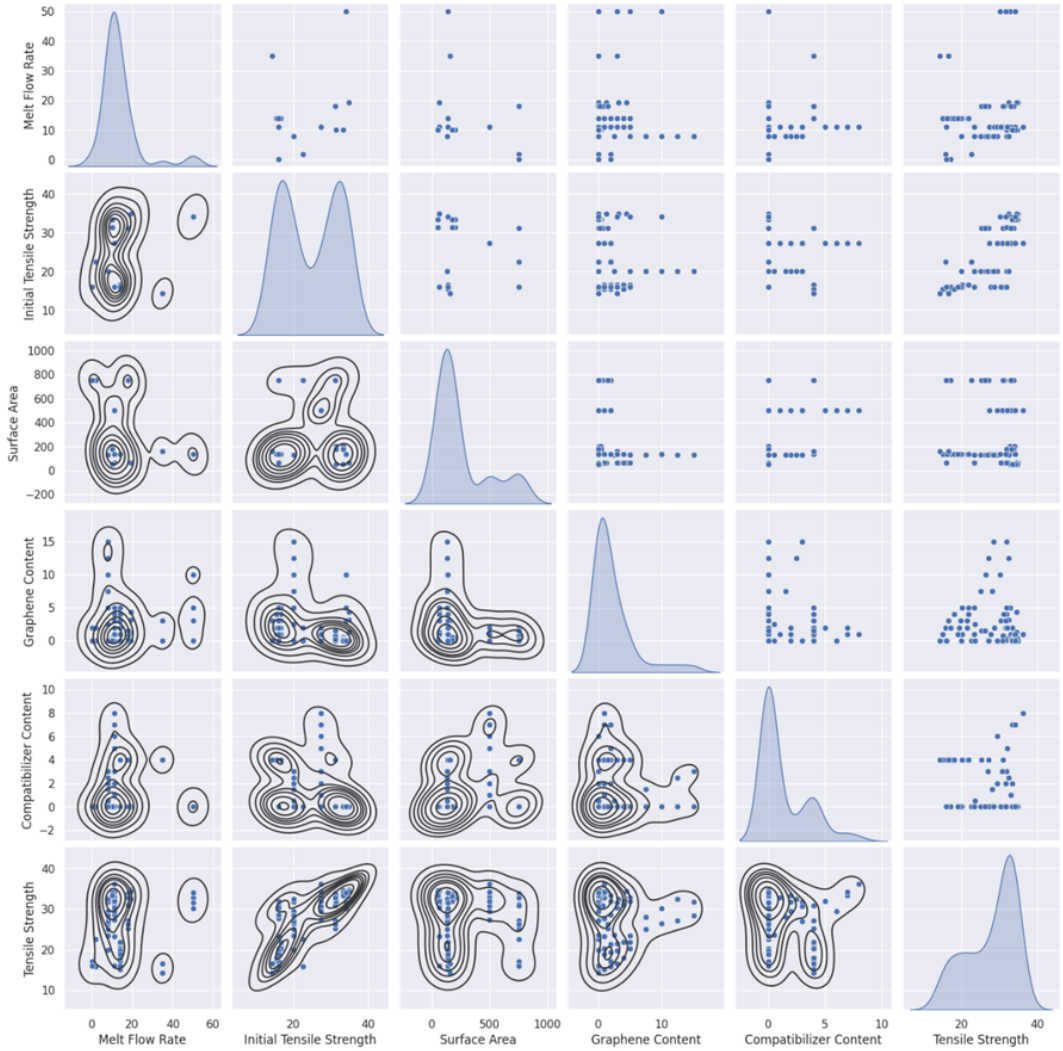


Figure 2: Pair plot of the independent variables.

Figure 3: Correlation matrix of independent and dependent variables.

It is noticeable that almost all independent variables seem to have little to no influence on the output, except the initial tensile strength. This variable is the only one that shows two significant peaks in its distribution, representing a more representative dataset. The frame in the last row of the second column in Figure 2 shows a somewhat direct correlation between the initial and final tensile strength, which is corroborated by the correlation value in Figure 3 (0.77). Similar to the surface and compatibilizer content, it displays no outliers (Figure 4). The median value is the one closest to the center, indicating that the data is better distributed.

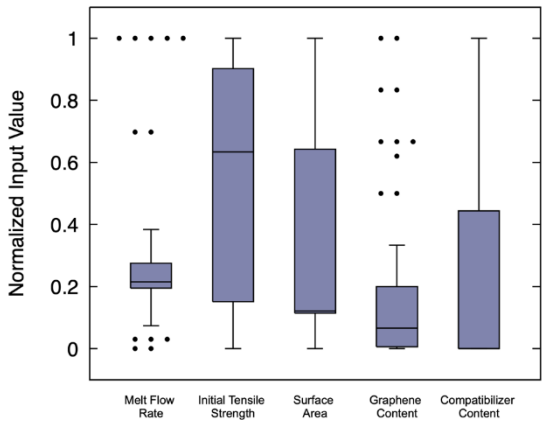


Figure 4: Boxplot of normalized input data.

ANN MODELING

Some tested architectures (number of hidden layers, number of neurons in each hidden layer, and transfer functions) with their respective performance results (R^2 and MSE values) are shown in Table 1. The overall results were satisfactory, considering that all the data came from different studies in the literature. By acquiring data using this method, some relevant information regarding the composition or production process may be omitted. Nevertheless, some architectures reached an accuracy of over 80 %. The best case (tenth row) was obtained with a relatively simple topology, with only a single hidden of 5 neurons and using the sigmoid function in its activation.

Table 1: Topology of the tested architecture and their performance stats.

Hidden layer 1		Hidden layer 2		MSE	R2
Number of neurons	Transfer function	Number of neurons	Transfer function		
8	sigmoid	-	-	10.52	0.775
5	ReLU	-	-	20.44	0.591
5	sigmoid	5	tanh	8.56	0.819
4	sigmoid	3	tanh	13.49	0.709
5	sigmoid	5	sigmoid	10.22	0.792
3	sigmoid	-	-	10.31	0.785
5	tanh	-	-	14.13	0.692
5	sigmoid	8	sigmoid	12.29	0.730
6	sigmoid	-	-	9.48	0.811
5	sigmoid	-	-	8.47	0.829

The results in Table 1 also indicate that more complex networks do not always have the best results, with only one case (third row) having a result with R^2 above 0.8. Moreover, the ReLU function is the one that had the worst results, and the presence of the sigmoid function seems to increase the networks' overall performance.

Figure 5 depicts the training performance of the best network. There is a rapid reduction in the objective function up to the 400th iteration. From this iteration onward, the error decrease occurs only in small values, representing a fine adjustment. The learning rate value (0.01) is critical in this phase as it can make the objective function reach its minimum value with fewer or higher iterations. The network could have gone a lower objective function value using a learning rate lower than 0.01; however, this would probably occur in iterations above the maximum amount (1000), given that the training phase ended with more than 950 iterations. On the other hand, a higher learning rate value could lead to a faster function minimization but also could "miss" the global minima due to the more rapid rate.

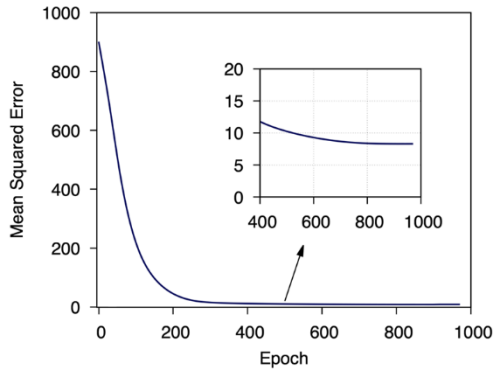
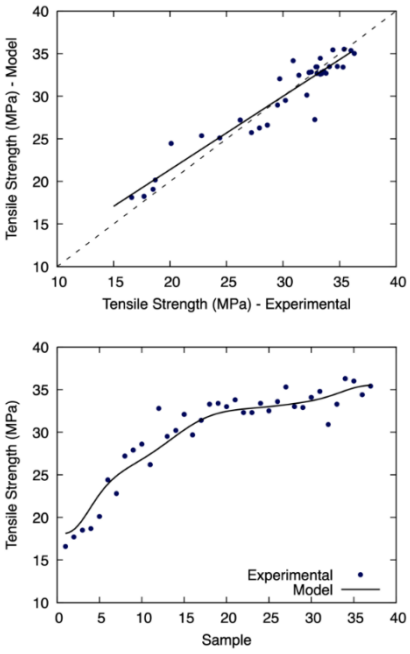


Figure 5: Training performance of the best-fitted network.

A comparison between the experimental and simulation results is displayed in Figure 6. The linear regression is depicted in Figure 6(a), showing a relatively good agreement between both data. Some noticeable discrepancies can be better seen in Figure 6(b). The experimental data has a scattered behavior, with no regular shape formed. This behavior could be due to the conditions used in each experiment. Some other components might have been added, and the articles did not report it.



(a)

(b)

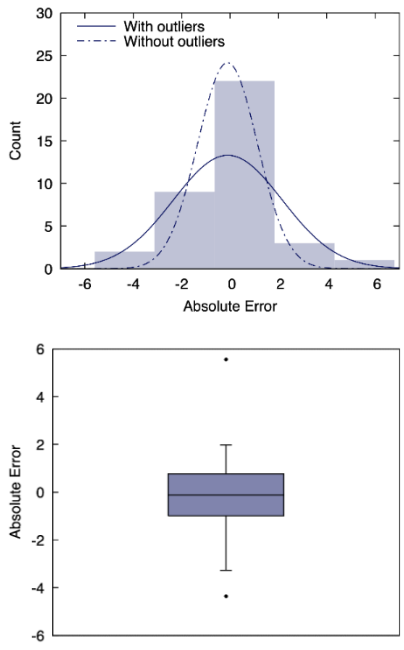
Figure 6: Experimental and model results: (a) linear regression; (b) samples.

Moreover, this preliminary study did not consider the experimental conditions, such as temperature. Such conditions have a significant impact on the final conditions of the material. Therefore, the results could be improved by adding these as input variables.

Regardless of the uncertainty concerning the experimental data, the simulations were able to follow a similar pattern as the literature data. Other than the experimental conditions, the network might be improved using more experimental results. The more data the network has, the more it can "understand" the influence of each input on the output. A better-trained network can give more reliable results to predict the mechanical performance of the PP and graphene composite without the need for so many experiments.

Figure 7(a) shows the absolute error distribution between experimental and simulation values. The average absolute error was -0.09, with a standard deviation of 2.24. Considering these values, most errors are between -2.5 and 2.5, with few cases of higher error values. This behavior was addressed in Figure 6, in which most model values were close to the experimental ones. However, Figure 7(b) shows that some error results can be seen as outliers (errors above 4 and below -4).

Therefore, the distribution gets narrower as shown in Figure 7(a) if these values are removed from the analysis. The average value is still very close to the obtained with the outliers, -0.10. On the other hand, the standard deviation drops from 2.24 to 1.23. Accounting this to the network results, the determination coefficient increases to 0.93 and the MSE value decreases to 2.58. These differences could be caused by some of the variables that are not being evaluated by the model, such as the manufacturing conditions.



(a)

(b)

Figure 7: Absolute error distribution of the experimental and model tensile strength values: (a) histogram; (b) boxplot.

CONCLUSIONS

Graphene and derivatives are materials that can be interesting alternatives to reinforce polymer materials. The prediction of the polymer composites' mechanical properties is highly complex due to the different characteristics of the initial materials and the various processing possibilities. The present work proposed to build a neural network model capable of predicting the final tensile strength based on five input variables: melt flow rate, initial tensile strength, surface area, graphene content, and compatibilizer content. Among the different tested architectures, the best one consisted of a single hidden layer with 5 neurons using the sigmoid transfer function. This network had a satisfactory prediction capacity with a R2 value of 0.83 and MSE value of 8.47 as performance parameters. Adding data to the network may increase the network's efficiency. Therefore, feeding more information regarding the manufacture processing (temperature, rotor speed) could be tested as it may enhance the ANN results.

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