Attaining material sustainability by incorporating nanoparticles additives to improve the mechanical properties of polypropylene composites: data driven modelling

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Abstract. Polypropylene is commonly employed in several industrial applications such as packaging, cable insulation and automotive. Research interest has focused on how to improve its mechanical properties to reduce the effect of low impact toughness of polypropylene. One of the sustainable ways to achieve this is by incorporating graphene nanoplatelets to form a composite. This study investigates the application of a hybrid support vector machine (SVM) and artificial neural networks (ANN) model to predict the effect of incorporating graphene on the mechanical properties of polypropylene composites. The effect of parameters such as maleic anhydride grafted polypropylene (MAPP), Talc, and exfoliated graphene nanoplatelets on the tensile strength and modulus of the polypropylene composites was modelled by using ANN. Testing various topologies was accomplished. An optimized ANN structure of 3-7-2 indicating 3 input-layer, 7 hidden layer, and 2 output-layer was tested. Both the SVM and the ANN predict well the mechanical properties of polypropylene composites. However, the ANN with $R^2$ of 0.999 offers the best predictions.

Keywords: Artificial neural networks; Support vector machine; Propylene; Graphene nanoplatelets

1. Introduction

Polypropylene is an important polymer product widely used for various purposes [1]. Due to the unique chemical properties which include semi-rigidity, translucent, good chemical resistance, toughness, fatigue resistance, and heat resistance, polypropylene finds wide applications in packaging, construction, and automobile industries [2]. In order to improve end-use performance as well as protecting the polypropylene, various types of additives are added during production [3]. To further improve the properties of polypropylene for enhanced usage, there is a growing interest in the production of polypropylene-composite materials [2, 3]. Low-cost natural fibres have been explored as reinforced materials to improve the thermal and mechanical properties of polypropylene composite. Various studies on the use of natural fibres revealed that the chemical, mechanical, and physical properties of the natural fibre-based polypropylene composites are strongly dependent on the cellulosic contents of the natural fibres [4].

Besides natural fibres, the effects of other materials on the properties of polypropylene composite have been investigated. Papageorgiou et al. [5] reported the effect of adding a mixture of glass fibres and graphene on the properties of polypropylene composite. The study revealed that the mechanical properties of the polypropylene composite as a function of the Young modulus was higher compared to the pristine polypropylene. Moreover, a higher thermal conductivity was obtained for the hybrid multifunctional graphene/glass-fibre polypropylene composites compared to that of the pristine polypropylene. In a similar study, Peng et al. [6] investigated the effect of lignin content on mechanical and thermal properties of polypropylene reinforced microparticles of spray-dried cellulose nanofibrils.
The tensile strength and modulus of the reinforced polypropylene composite were found to be significantly improved compared to the pristine polypropylene. Obaid et al. [7] investigated the stress relaxation behaviour of glass-fibre reinforced polypropylene composites using both experimental and analytical model prediction methods. The predicted stress relaxation behaviour of the glass-fibre-reinforced polypropylene using the analytical model methods strongly agrees with the experimental values.

Machine learning algorithms such as the SVM and ANN can be employed to model the non-linear relationship between the various parameters and the polypropylene composite. Uysal and Tanyildizi [8] employed ANN to estimate the compressive strength of self-compacting concrete containing polypropylene fibre and mineral additives exposed to high temperature. The study revealed that the artificial neural network (ANN) model accurately predicts the capability of the loss in compressive strength of the self-compacting concrete mixtures after exposure to elevated temperature. Clemente et al. [9] employed a backpropagation ANN model to predict the flexural and compressive strength of concrete reinforced with polypropylene fibre. Kazi et al. [10] investigated the prediction of optimal filler content for cotton fibre-polypropylene composite-based mechanical properties using an artificial neural network. The study showed that the ANN model was efficient to predict the optimum filler content. In this study, SVM and ANN were employed to model the effect of adding graphene nanoplatelets on the mechanical properties of polypropylene nanocomposites.

2. Materials and Methods

2.1 Experimental and Data Acquisition
The detailed experimentation which includes the materials used, the preparation of the polypropylene composites, the characterization, and the testing of the composite for its mechanical properties has been reported by Ghasemi et al. [11]. The datasets employed for the modelling consists of 15 experimental runs generated using experimental designs. The datasets are combinations of the amount of talc, maleic anhydride grafted polypropylene (MAPP), and graphene nanoplatelets. The effects of these combinations of the parameters on the mechanical strength of the polypropylene composite were investigated.

2.2 Model Development
The modelling was performed using SVM and ANN [12]. Support vector machine is a supervised machine learning model that can be employed for classification and regression analysis [13]. Support vector machine can perform non-linear classification and regression analysis by mapping the input of the process into high dimensional features [14]. The ANN model was built to imitate the human neuron system [15]. It consists of the input layers, the hidden layer, and the output layers linked with interconnections of artificial neurons. The artificial neurons received signals from the input layer, process them, and subsequently transmit them to the output layer for prediction of the output. The input data which consists of the amounts of talc in the composite mixture, the amount of MAPP in the composite mixture, and the amount of graphene nanoplatelets were loaded into the network through the input layer. For efficient prediction, the network was trained in order to determine the differences between the process output and the target otherwise known as the network predictive error. For an efficient prediction, the network error can be minimized as much as possible by using the best hidden neuron. The best hidden neuron is determined by testing a range of neurons to obtain the one that will minimize the network error. The datasets are divided into three portions for training, testing and validation at the ratio of 0.7, 0.15 and 0.15, respectively. MATLAB version 2019 b (MathWorks Inc.) was employed for the SVM and the ANN. The network error was measured as a function of the mean square error (MSE) which is the average square difference between the predicted values and the experimental value. The coefficient of determination ($R^2$) which measures the proportion of variance in the independent variables that is predictable from the independent variable was employed to measured how closely related the predicted values are to the experimental values. The effect of the amounts of
talc in the composite mixture, the amount of MAPP in the composite mixture, and the amount of graphene nanoplatelets on the mechanical properties of the polypropylene composite was investigated using Garson algorithm.

3. Results and Discussions

Table 1 shows a summary of the results obtained from the detailed analysis to determine the best-hidden neurons by adjusting the artificial neurons at the hidden layer from 1 to 20. It can be seen that the MSE, R, and R² changes with the model structure. Each of the model structure depicts the number of input neurons, hidden neurons, and output neurons. The training, validation, and testing of the artificial network revealed that the model with the 3-7-2 has the best performance with MSE values of 116.84, 171.54, and 112.34 for the training, validation, and testing, respectively. Hence, the 3-7-2 ANN model depicted in Figure 1 was employed for modelling the prediction of the effect of the various additives on the mechanical properties of the polypropylene composite.

| Model structure | Training | Validation | Testing |
|-----------------|----------|------------|---------|
|                 | MSE      | R          | R²      | MSE      | R          | R²      | MSE      | R          | R²      |
| 3-1-2           | 7376     | 0.974      | 0.949   | 3354.64  | 0.968      | 0.937   | 13265    | 0.952      | 0.906   |
| 3-2-2           | 68470.1  | 0.874      | 0.764   | 5900.92  | 0.998      | 0.996   | 30562    | 0.936      | 0.876   |
| 3-3-2           | 1126.89  | 0.996      | 0.992   | 211.61   | 0.999      | 0.998   | 20330    | 0.995      | 0.990   |
| 3-4-2           | 7047.36  | 0.985      | 0.970   | 3746.83  | 0.993      | 0.986   | 26.32    | 0.999      | 0.998   |
| 3-5-2           | 10793.5  | 0.965      | 0.931   | 1373.96  | 0.997      | 0.994   | 7236     | 0.988      | 0.976   |
| 3-6-2           | 11313    | 0.964      | 0.929   | 12566    | 0.989      | 0.978   | 26267    | 0.999      | 0.998   |
| 3-7-2           | 116.84   | 0.999      | 0.998   | 171.54   | 0.989      | 0.978   | 112.34   | 0.988      | 0.976   |
| 3-8-2           | 2696.55  | 0.995      | 0.990   | 10492.3  | 0.988      | 0.976   | 54306.9  | 0.927      | 0.859   |
| 3-9-2           | 5031.71  | 0.985      | 0.970   | 52742.2  | 0.858      | 0.736   | 16538    | 0.994      | 0.988   |
| 3-10-2          | 26000    | 0.991      | 0.982   | 26399    | 0.939      | 0.882   | 17242    | 0.961      | 0.924   |

Figure 1: Configuration of the ANN Model with the best performance

The predictability of the SVM and the ANN algorithm in modeling the effect of adding graphene nanoplatelets to the propylene composite. As it can be seen in Figure 2 (a) and (b), both the SVM and the ANN were used for modelling the prediction of the tensile strength and the tensile modulus. The
ANN has a better prediction of the tensile strength and the tensile modulus compared to the SVM. The predicted tensile strength and modulus by the ANN are in proximity to the experimental values. The better predictability of the ANN model in modeling the effect of graphene nanoplatelets addition on the mechanical properties of the polypropylene composite can be attributed to several factors. The neural network models possess an inbuilt algorithm that has the capability to learn non-linear functions. The ANN also has the ability to detect faults associated with missing information. The predictability of the ANN model used in this study is consistent with that reported in the literature. Benimam et al. [13] performed a comparative analysis of modeling the activity coefficient at infinite dilution of water in ionic liquids using ANN and SVM. The results revealed that the artificial neural networks model was robust in modelling the prediction of activity coefficient at infinite dilution of water in ionic liquids with \( R^2 \) of 0.99997. In a similar study, Abbas et al. [14] employed support vector machine and artificial neural networks to model the prediction of lost circulation. The result revealed that both SVM and ANN showed great potential in modelling the prediction of lost circulation during drilling of crude oil.

The sensitivity analysis showing the effect of the various additives on the mechanical properties of the polypropylene composite is depicted in Figure 2 (c). All three additives namely talc, MAPP, and graphene nanoplates significantly influence the predicted mechanical properties of the polypropylene composite. The normalized importance of the talc, MAPP, and graphene nanoplates additives are estimated as 75.8%, 38.7%, and 100%, respectively. This implies that the graphene nanoplatelets have the most significant effects on the predicted mechanical properties. Ahmad et al. [16] reported the mechanisms of reinforcement of polypropylene by graphene nanoplatelets. The study revealed that the addition of graphene nanoplatelet significantly improves the Young modulus of the polypropylene composite. Song et al. [17] reported that graphene plays a significant role in improved thermal conductivity of graphene-polypropylene composite with excellent heat dissipation when used as a thermal management material in LED integration.

The effect of the talc, MAPP, and graphene addition on the tensile strength and modulus of the polypropylene composite are depicted in Figures 3 and 4, respectively. In Figure 3, it can be seen that the talc, MAPP, and graphene nanoplatelets have a significant effect on the tensile strength. The increase in the talc, MAPP, and the graphene nanoplatelets contents resulted in a corresponding increase in the tensile strength of the polypropylene composite. As shown in Figure 3 (a), an increase in MAPP and talc contents resulted in a corresponding increase in the tensile strength of the polypropylene composite. Similarly, in Figure 3 (b), an increase in graphene nanoplatelets and MAPPs resulted in an increase in the polypropylene composite. A similar trend can also be observed for an increase in the graphene nanoplatelet and talc in the polypropylene composite as shown in Figure 3 (c). The addition of the graphene nanoplatelets had the optimal significant influence on the tensile strength. Also, Figure 4 depicts the three-dimensional plots showing the effects of the talc, MAPP, and graphene addition on the tensile modulus. As illustrated in Figure 4 (a) and Figure 4 (c), the increase in the talc and the graphene nanoplates resulted in a corresponding increase in the tensile modulus. However, the optimum MAPP effects were attained at 2%.
Figure 2: Dispersion plots for the prediction of (a) the tensile strength (b) the tensile modulus and (c) the sensitivity analysis of the polypropylene composite
Figure 3: The effect of interaction between (a) MAPP and Talc (b) Graphene nanoplatelets and Talc (c) Talc and Graphene nanoplatelets on the tensile strength of the polypropylene composite
Figure 4: The effect of interaction between (a) MAPP and Talc (b) Graphene nanoplatelets and Talc (c) Talc and Graphene nanoplatelets on the tensile modulus of the polypropylene composite.
4. Conclusion

The study investigates the predictive modelling of polypropylene composite mechanical properties prepared by the addition of MAPP, talc and graphene nanoplatelets. An artificial neural model (ANN) of 3-7-2 was employed for the ANN model. Both the support vector machine (SVM) and the ANN models predicted well the mechanical properties of the polypropylene composite. However, the ANN model offers a better prediction of the tensile mechanical properties of the polypropylene composite. The sensitivity analysis revealed that the talc, MAPP, and the graphene nanoplatelet significantly influence the predicted tensile strength and modulus. The addition of graphene nanoplatelets to the polypropylene significantly influences the predicted tensile mechanical properties of the polypropylene composite. The addition of the various nanoparticles in the preparation of the polypropylene composite creates a new opportunity for a highly efficient sustainable nanocomposite materials. In the case whereby the process is to be scaled-up, this study will serve as a guide to employ the appropriate algorithm to determine the right proportion of the various additives.

5. References

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Acknowledgement
May Ali Al-saffar wish to acknowledge the financial support of the Department of Chemical Engineering, University of Technology, Iraq?