ANN Modelling for Predicting the Water Absorption of Composites with Waste Plastic Pyrolysis Char Fillers

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***Abstract*—Waste material was fragmented into gas, liquid and solid fractions by pyrolysis. Recently the solid fraction (char) has been used as filler in epoxy composites. Type and properties of filler affect water absorption of epoxy composites. A recent water absorption database (of 1512 data) has been obtained experimentally. Accordingly, type of paralysed plastic, waste pre–washing, pyrolysis temperature, additive dosage and water exposure time were input parameters in the estimation model developed with multilayer perceptron artificial neural network (MLP ANN) to predict the absorbed water quantity as output. Four datasets were derived with data pre-processing. Among all the configurations worked up, 0.991 training and 0.986 testing R^2 were attained as the highest R^2 values under conditions including 2e4 iterations, lr 0.04, mc 0.9, first hidden layer of 22 nodes, and second hidden layer of 15 nodes. The R^2 value attained in the optimum configuration and the average R^2 attained via 5-fold cross-validation are close to each other for both training and test. The established model will help users to predict the quantity of water that absorbed upon exposure. This will give idea about the availability of that composite for using it for particular purposes.**

Keywords—ANN, waste plastics, composite, epoxy resin, water.

I. INTRODUCTION

Water absorption tendency is a property that can be directly attributed to chemical nature and physical structure of the material. It has been long known that epoxy and epoxy composites easily absorb water when exposed to humid environments [1]. The thermoplast polymer matrix is known to absorb and diffuse less moisture because it does not contain open molecular structures and unsaturated sites, which react with water, a highly polar solvent, as does a thermoset matrix [2].

The moisture absorption by composites has several adverse effects on their properties and thus, affects their operational characteristics and long–term performance [3]-[5]. The water in epoxy is present in two states; firstly, distributed water molecules between the polymer chain and condensed water in fractures and cavities [6]. Moisture absorption results in plasticization of the matrix, and subsequently to swelling which can cause matrix cracking and fiber/matrix debonding. It reduces the stable lifetime of the material. Therefore, it is important to study the water absorption behaviour of composites in order to estimate the consequent effects on the performance of composite parts. Such studies enable users to develop strategies for controlling and minimizing water absorption [3].

Pyrolysis is one of the famous recovery processes that can be applied to many types of organic polymeric wastes to fractionate them into useful products. As a result of pyrolytic decomposition the paralysed material was fragmented into gas, liquid and solid fractions. Liquid and gas products generally contain components that can easily be used as fuel and/or feedstock after further processing. Within zero–waste approach, the solid fraction (char) has been evaluated in other areas such as adsorption, activated carbon production etc. In recent studies char has been used as filler in epoxy composites. Depending upon the paralysed material and pyrolysis conditions, char properties and char–added composite properties, which affect their utilization areas, change significantly. Water/humidity absorption properties of epoxy composites are also affected from type of filler material.

Recently artificial neural network algorithm has been preferred to predict different parameters of epoxy composites because of some important disadvantages of experimental studies such as high cost, time consuming etc. Most of these studies interested in the modelling the mechanical behavior of composite materials using ANN [7]-[9]. Although the modelling of composites' water absorption behaviour is an important problem a few published studies on modelling water absorption in composites have been conducted. Pujari et al. aimed to compare the ANN and regression models for predicting the water absorption properties of jute and banana fiber composites using interpolated values obtained from the experiment by fuzzy [3]. In a similar study, Pujari et al. compared ANN and the correlation–based method for various foundation configurations with the interpolated values of experimental works using jute and banana fiber

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composites [4].

Nazari and Azimzadegan developed (ANN) and gene expression programming (GEP) models for predicting splitting tensile strength and water absorption of concretes containing ZnO2 nanoparticles at different ages of curing [10]. Chen et al. presented ANN modelling to predict the water absorption rate of poly (methyl methacrylate) PMMA called organic glass and its composites [11].

In this study, ANN modelling was aimed to predict the absorbed water quantity for composite material by using a dataset obtained previously as a result of experimental studies. In order to find the best model configuration, the optimum parameter values of ANN were investigated.

II. MATERIAL AND METHODS

A. Dataset

A recent water absorption database has been obtained as a result of experimental studies of a completed project in which Bisphenol–A type epoxy composites prepared with plastics pyrolysis char additives [12]. Change of absorbed water quantity (mg/g) with type of paralysed waste plastic type (HDPE, LDPE, PET, PS, PP, MIX), existence of waste washing pre-process, plastic pyrolysis temperature (up to 700°C) and char additive dosage (up to 50%) in the composite and water exposure time (up to 10 days, after which composites reach saturation) was included in the dataset. ANN model was established to predict the absorbed water quantity for composite material by using this dataset.

B. Artificial Neural Network (ANN)

In this study modelling of water absorption quantity of composites was implemented depending on paralysed waste plastic type, existence of waste pre–washing, plastic pyrolysis temperature, water exposure time and char additive dosage in the composite by using Artificial Neural Network (ANN) algorithm. ANN developed by being inspired from human brain is a popular and powerfully Artificial Intelligence algorithm. Therefore, different ANN algorithms have been drawn attention in the literature such as Multi-Layer Perceptron (MLP), Levenberg Marquert (LM), Radial Based Neural Network (RBNN) etc. The common points of these algorithms are process units called as neurons and weights between neurons. These algorithms are different from each other in terms of structure, learning algorithm, activation function etc.. The structure of ANN is depending on the complexity of problem and experimentally specified. In this study, MLP was preferred to model experimental data because of its applicability and performance in prediction problems. In this study, prediction modelling with MLP was carried out by using MATLAB release R2010a.

MLP is a fully connected feedforward neural network with one or more hidden layer(s) between input and output layers and also trained with error back–propagation learning algorithm. The basic features of MLP are differentiable nonlinear activation function of each neuron in hidden and output layers and weights between neurons in different layers. Training is based on the adjusting weights in the learning process. Before the training, the input vector and expected output vector called as desired vector are presented to the network and the initial values of weights are assigned. The learning process is performed in two phase: forward and backward. In the forward phase, the input vector is propagated layer by layer through the network with the fixed weights of the network and the output signal is produced. In the backward phase, the output of the network is compared with the target vector to compute an error for each input observation. Then resulting sum error signal is propagated through the network, again layer by layer in the backward direction to adjust weights of the network [13]-[14].

There are several important parameters which affect the performance of learning process such as the number of hidden layer, the number of neurons in hidden layer(s), learning rate, momentum coefficient and the number of iteration [13]-[14]. In the learning process the optimum values of these parameters are investigated to reach the best model by changing them with different datasets in different stages.

In this study, data pre-processing was applied to see the effects of some uncontrolled extraordinary data on the modelling stages. These data include the theoretical initial (t=0) water absorption values (which were zero in the dataset) and missing experimental values (assigned as zero in the dataset) in the original dataset. Thus, by applying data pre-processing, four different datasets were generated from original dataset as described in Table 1.

TABLE I. THE RECOMMENDED FONTS.

Description	Name of Dataset	Number of Data
Original dataset includes theoretical initial water absorption values as zero and missing values as zero	DS1	1512
Zero values assigned for missing data were eliminated as the first step of data preprocessing	DS2	1498
Irrelevant and/or unexpected water absorption values were removed as the second step of data preprocessing	DS3	1482
All zero values representing theoretical initial water absorption values and also representing missing data were eliminated as the third step of data preprocessing	DS4	1373

Modelling process stages were as follows:

In the first stage of modelling (ST1), each dataset was divided into subsets for training and testing as 80% and 20%, respectively. Preliminary modelling studies were performed with DS1 to decide the number of hidden layers. Firstly the modelling was done by using MLP with one hidden layer by altering the number of neurons in the range [5 25] for 0.2 learning rate (lr) and 0.8 momentum coefficient (mc). The obtained best model was called as ST1–DS1.

In the second stage (ST2), the experiments was performed using the first dataset, DS1, in MLP with two

hidden layers by changing number of neurons in hidden layers to decide the better structure. The number of neurons in the first hidden layer and in the second hidden layer were changed in the ranges [4 25] and [5 15], respectively. As a result, it was decided that MLP with two hidden layers was more successful than the structure with one hidden layer and the subsequent studies were performed with MLP with two hidden layers.

In the third stage (ST3), the experiments were performed for each dataset by changing the number of hidden neurons in the ranges of [4 25] and [5 15] to find the best model structure (ST3–DS1, ST3–DS2, ST3–DS3, ST3–DS4).

In the fourth stage (ST4), with the best number of neurons of each dataset lr and mc were changed in the ranges [0.1 0.5] and [0.2 0.9] to find optimum values of them. For each dataset lr and mc resulting in the highest prediction performance were determined and these configurations were called as ST4–DS1, ST4–DS2, ST4–DS3, ST4–DS4.

In the fifth stage (ST5), fine tuning of lr and mc was performed for the modelling of DS4 by using the same layer structure of ST4–DS4 which resulted in the highest prediction performance. In the previous stage, the values of lr and mc for ST4–DS4 had been experimentally found as 0.1 and 0.9, respectively. However 0.1 is the lower bound of lr and 0.9 is the upper bound of mc search space. Therefore, by keeping the other optimized values constant, the experiments were carried out with DS4 by changing both lr in the range [0.01 0.2] and mc in the range [0.85 0.95] with the increment 0.01 for the same structure. The reached best model was called as ST5–DS4.

In the sixth stage (ST6), 5 fold cross validation was performed for ST5–DS4 to prove the reliability of the study. In previous steps 80% of the dataset was used for training while the remaining 20% was used for testing. In each fold of cross validation another 20% portion of the dataset was used for testing while the remaining 80% of it was used in training. The calculated errors and R2 were listed at the end of each run and statistically compared to indicate the reliability of model performance. The advantages of this method are that every data point gets to be in a test set exactly once, and gets to be in a training set 4 times [15]

In the previous stages, the number of iteration had been selected as 2e4. In the seventh stage (ST7), the effect of iteration number was investigated by using the same optimum configuration in the fifth stage, ST5–DS4.

C. Performance Criteria

Comparisons were made between the model estimation results and the experimental results by calculating the Mean Absolute Error (MAE), Mean Square Error (MSE), Root Mean Square Error (RMSE), and the coefficient of specificity (R^2). The equations used in these calculations are given in Eq.1-4.

$$MAE = \frac{1}{N}\sum_{i=1}^{N}\left|O_{Exp}(i) - O_{Model}(i)\right| \quad (1)$$

$$MSE = \frac{1}{N}\sum_{i=1}^{N}(O_{Exp}(i) - O_{Model}(i))^2 \quad (2)$$

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(O_{Exp}(i) - O_{Model}(i))^2} \quad (3)$$

$$R^2 = \frac{\left[\sum_{i=1}^{N}(O_{Exp}(i) - \overline{O_{Exp}})(O_{Model}(i) - \overline{O_{Model}})\right]^2}{\sum_{i=1}^{N}(O_{Exp}(i) - \overline{O_{Exp}})^2 \sum_{i=1}^{N}(O_{Model}(i) - \overline{O_{Model}})^2} \quad (4)$$

in which N is the number of data in dataset; $O_{exp}(i)$ and $O_{Model}(i)$ are i_{th} experimental and model output values of absorbed water quantity, respectively. The average values of experimental output in the dataset and model output are indicated as $\overline{O_{Exp}}$ and $\overline{O_{Model}}$.

The condition which yielded the lowest error and the highest R^2 was selected as the best condition. These error calculations were made both for the training, and the test data, and training and test performances were assessed separately.

III. RESULTS

A 'one factor at a time' model run was implemented. In each step the optimized value of the previous step and all the conditions were kept constant except the variable of that step. Accordingly, lr (0.2), mc (0.8) and the number of iteration (2e4) were assigned initially. First, the MLP prediction performances for one and two hidden layers were compared with original data set (DS1). The performace of prediction of models for each structure (ST1–DS1 and ST3–DS1) were compared in Table 2. The error values (MAE, MSE and RMSE) for MLP with two hidden layers (ST3–DS1) were lower than the model with one hidden layer (ST1–DS1) while R^2 value was higher than ST1–DS1. As better results were achieved by using the structure with two hidden layers, subsequent studies were performed with two hidden layer MLP for all datasets.

TABLE II. COMPARISON OF ONE AND TWO HIDDEN LAYER STRUCTURE MODEL PERFORMANCES.

	Number of Neurons		**Parameters**		**Training Results**				**Test Results**			
	HL1	**HL2**	**lr**	**mc**	**MAE**	**MSE**	**RMSE**	**R2**	**MAE**	**MSE**	**RMSE**	**R2**
ST1–DS1	28	0	0.2	0.8	0.508	0.438	0.662	0.967	0.649	0.801	0.895	0.957
ST3–DS1	20	14	0.2	0.8	0.295	0.160	0.400	0.989	0.429	0.338	0.581	0.982

After the decision of the number of layers the number of neurons in each hidden layer were optimized for each dataset to find the best approximation to real water absorption quantity values. At this third stage lr (0.2), mc (0.8) and the number of iteration (2e4) were kept constant and only number of neurons were changed. Then the effects of lr and mc were studied by changing their values in the fourth stage The prediction performances of the best models of these two consecutive stages, ST3 (ST3–DS1, ST3–DS2, ST3–DS3, ST3–DS4) and ST4 (ST4–DS1, ST4–DS2, ST4–DS3, ST4–DS4) are presented in Table 3. The results indicated that DS4 was the dataset obtained the lowest errors and highest R2. In addition, the model was run for ST4–DS4 by performing fine tuning of lr and mc in the fith stage and the results (ST5–DS4) were added as the last row of Table 3. The scatter plot of predicted test values of ST5–DS4 model against real experimental values is given in Figure 1. Both the slope of the trendline and R2 are close to 1 as expected.

TABLE III. OPTIMIZATION OF NUMBER OF NEURONS AND PARAMETERS FOR EACH DATASET.

	Training				Test				Number of Neurons		Parameters	
Model	**MAE**	**MSE**	**RMSE**	**R2**	**MAE**	**MSE**	**RMSE**	**R2**	**HL1**	**HL2**	**lr**	**mc**
ST3–DS1	0.295	0.160	0.400	0.989	0.429	0.338	0.581	0.982	20	14	0.2	0.8
ST4–DS1	0.302	0.166	0.408	0.988	0.422	0.338	0.581	0.982	20	14	0.1	0.9
ST3–DS2	0.281	0.133	0.365	0.991	0.442	0.392	0.626	0.978	22	15	0.2	0.8
ST4–DS2	0.283	0.136	0.369	0.991	0.434	0.378	0.615	0.979	22	15	0.1	0.9
ST3–DS3	0.315	0.168	0.410	0.989	0.457	0.363	0.603	0.975	17	13	0.2	0.8
ST4–DS3	0.309	0.161	0.401	0.990	0.413	0.318	0.564	0.978	17	13	0.3	0.9
ST3–DS4	0.270	0.131	0.362	0.991	0.383	0.281	0.531	0.985	22	15	0.2	0.8
ST4–DS4	0.263	0.124	0.352	0.991	0.381	0.277	0.526	0.985	22	15	0.1	0.9
ST5–DS4	0.263	0.124	0.353	0.991	0.378	0.272	0.521	0.986	22	15	0.04	0.9

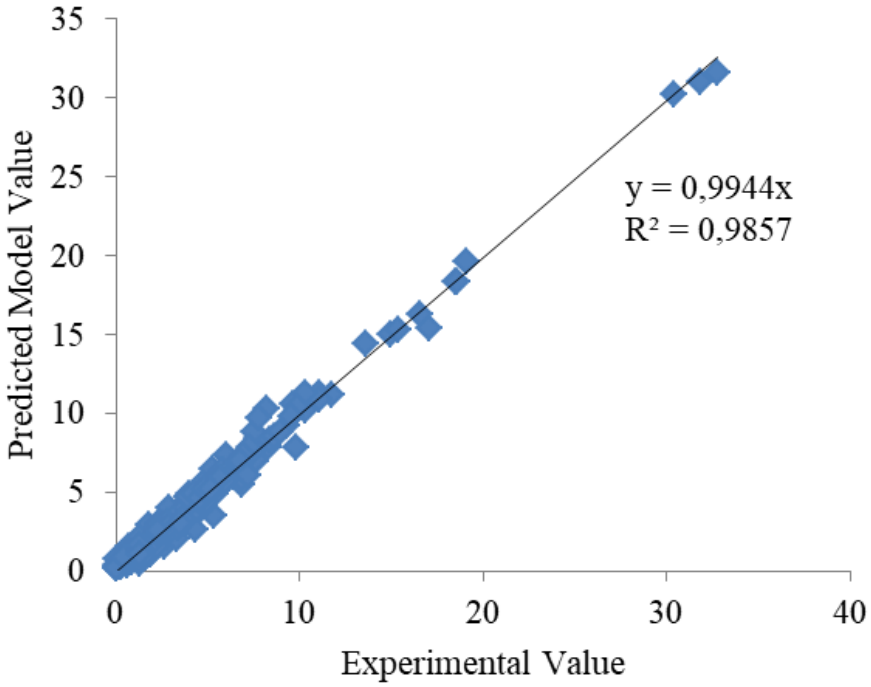


Fig. 1. Scatter plot of best model configuration ST5–DS4.

lr is expressed as the size of the step taken while approaching the minimum over the error function. The fact that the *lr* has a low value means that learning process takes place in small steps, i.e., brings together a slow learning process [16]. On the other hand, the network may not reach to the absolute minimum when higher learning coefficients are used to increase learning speed. This situation is called oscillation. Thus, *lr* should be taken so as not to cause oscillation. For this reason, in the studies conducted, the learning ratio is generally assigned a value between 0 and 1, preferably between 0-0.2 [17-18]. In the light of this information and with no significant change in error ratios, 0.2 which gives the lowest error and the highest R^2 in all error functions for the test data was selected as the optimum *lr*. mc was optimized changing the mc between 0.1-0.9 in accordance with the approach in the literature that the selection range should be between 0-1[13]. For all models, the highest prediction performance was obtained when all zero values in the dataset representing theoretical initial water absorption values and also representing missing data were eliminated i.e, with DS4. This indicates that theoretical values should not be included in the dataset for such a prediction model. Comparison of prediction performances of the best

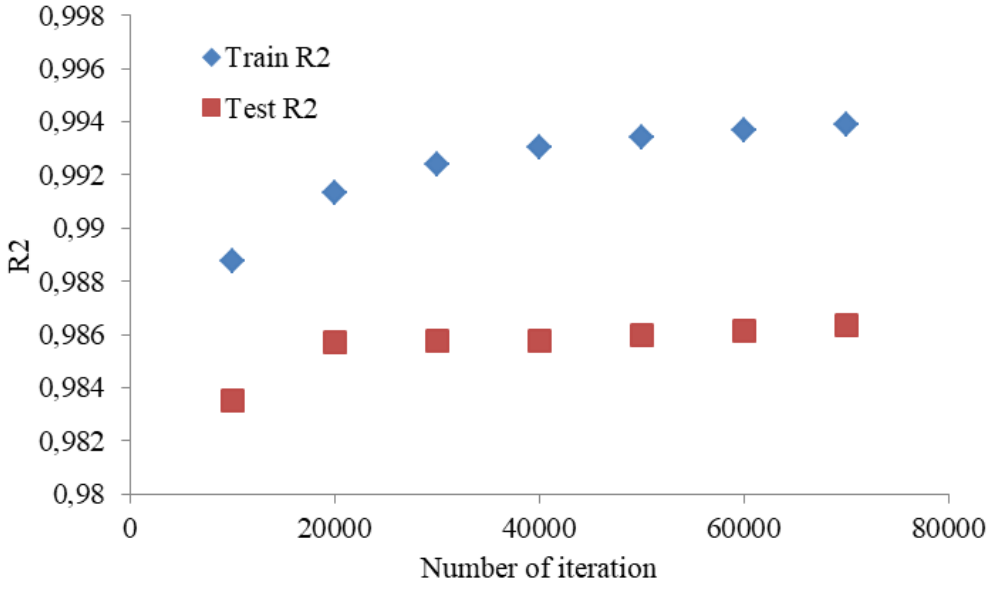
configuration of models with DS2 and DS4 datasets indicated that, during data preprocessing eliminating not only zero values assigned for missing data in the dataset, but also elimination of theoretical zero values is essential for higher prediction performance. Correct data preprocessing is important in modeling. The lowest prediction performance was achieved with DS3, indicating that removing any data by considering it as irrelevant and/or unexpected value is not meaningful as data preprocessing.

In this study, the best model configuration was reached in the above five stages. In the sixth stage, 5-fold cross–validation was applied to DS4 dataset by using ST5–DS4 model configuration to indicate the reliability and success of it. The obtained error and R2 values of each fold, and their mean and standart deviation values for both training and testing were given in Table 4. When the values attained in the previously specified optimum configuration and the average values attained via cross-validation are compared, training R^2 values were close to each other, while there was not a quite big difference between the test R^2 values. The fact that R2 values are close to each other for both training and test values indicates the success of the optimum configuration attained. Standard deviations are low and mean R2 values were close to maximum values indicating that this model can predict water absorption values with high performance.

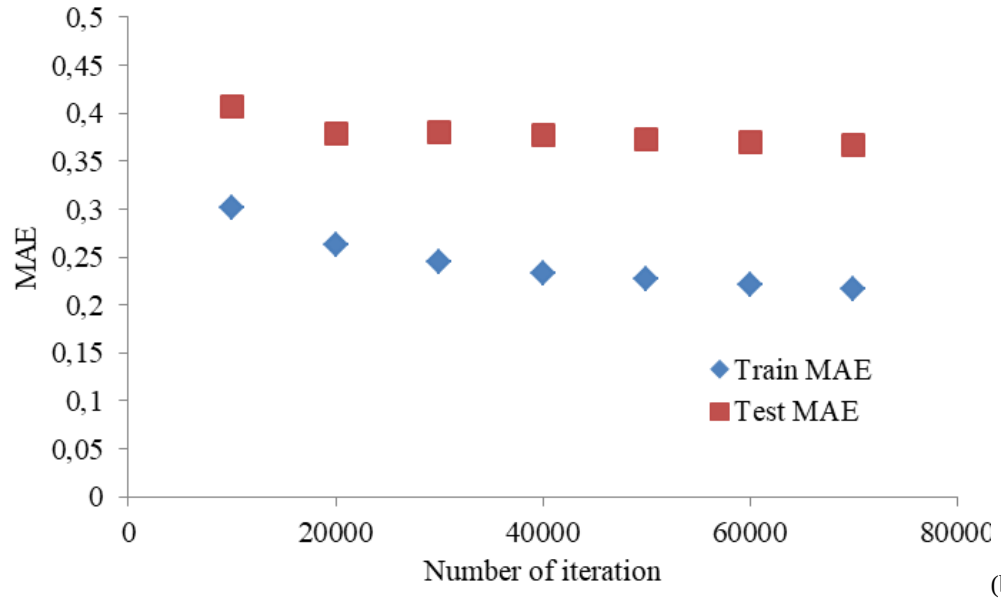
TABLE IV. PERFORMANCE RESULTS AND THEIR STATISTICS FOR CROSS VALIDATION.

Cross	**Training**				**Test**			
Val.	**MAE**	**MSE**	**RMSE**	**R2**	**MAE**	**MSE**	**RMSE**	**R2**
1st Fold	0.2629	0.1245	0.3528	0.9913	0.3785	0.2716	0.5211	0.9857
2nd Fold	0.2736	0.1370	0.3701	0.9908	0.4369	0.5707	0.7554	0.9685
3rd Fold	0.2501	0.1160	0.3406	0.9928	0.4252	0.3643	0.6036	0.9711
4th Fold	0.2500	0.1114	0.3337	0.9925	0.4260	0.7066	0.8406	0.9586
5th Fold	0.2558	0.1152	0.3394	0.9929	0.3794	0.3876	0.6226	0.9665
Mean	0.2585	0.1208	0.3473	0.9921	0.4092	0.4601	0.6687	0.9701
Std Dev	0.0089	0.0092	0.0130	0.0009	0.0251	0.1568	0.1142	0.0089

Finaly, the effect of iteration number was investigated on the best model configuration (ST5–DS4) in the last stage and the results were shown in Figure 2. It is seen from the figure that the model performance increases when the number of iteration increased from 1e4 to 2e4 but above 2e4 iteration, there is no significant change in the model performance as the number of iteration increase.



(a)



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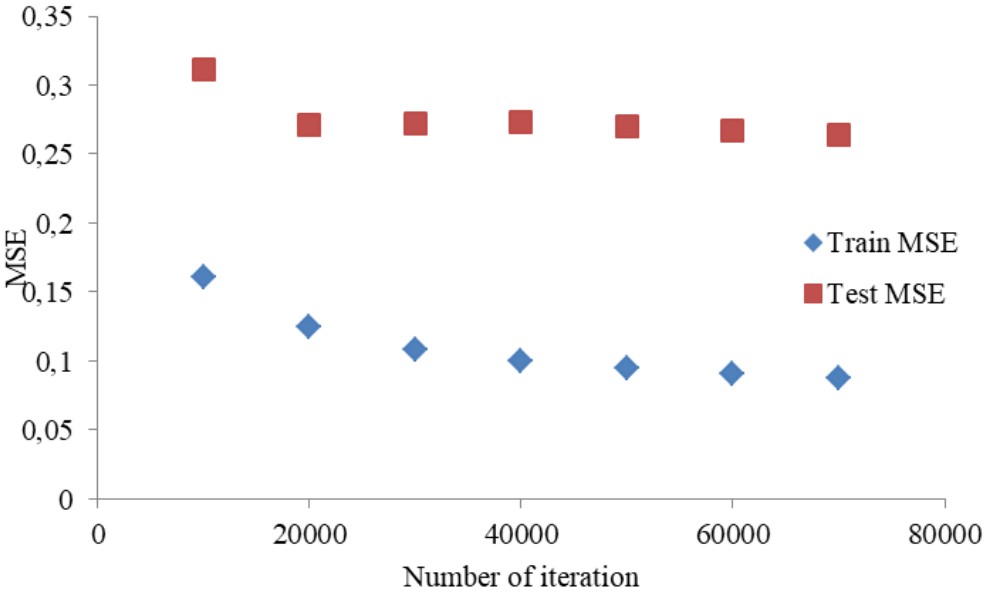

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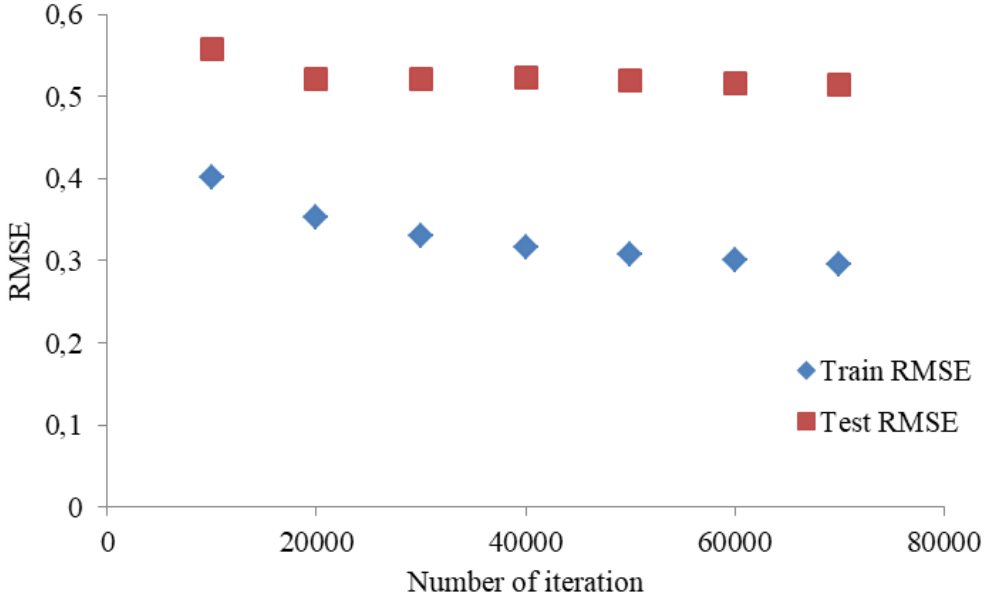

(d)

Fig. 2. Effect of number of iterations on (a) R2 values, (b) MAE values, (c) MSE values, (d) RMSE values for both model training and test

I. CONCLUSION

In this study, water absorption quantity of epoxy composites with pyrolysis char additives was modeled by using MLP. The established prediction model was improved by optimizing the model configuration in consecutive stages. The input parameters of MLP are pyrolysed waste plastic type, existence of waste pre–washing, plastic pyrolysis temperature, water exposure time and char additive dosage in the composite while the output parameter of model is water absorption quantity of composites. The most important aim of modelling is the best prediction of water absorption quantity. The effects of data preprocessing, MLP structure and the number of iterations were also investigated.

This study indicated that data preprocessing is important in increasing model prediction performance. Before ANN modeling it is necessary to review all the data. If there is missing data in the experimental dataset, it should not be replaced with zero or with another value. This reduces the model performance. Theoretically for exposure time is zero (i.e. at the beginning of water exposure) absorbed water quantity is zero as well. This can be used in plotting the change of absorbed water quantity against time. However, this theoretical approach should not be used in the dataset in prediction model, otherwise the prediction performance decreases.

Among all the configurations worked up, the lowest error and the highest R^2 values were obtained under hyperbolic tangent transfer function configuraton including 2e4 iterations, *lr* 0.04, *mc* 0.9, 22 nodes in the first hidden layer, and 15 nodes in the second hidden layer. R^2 values attained in this configuration were found respectively as 0.991 in the training, and as 0.986 in the testing. The R^2 attained in the optimum configuration and the average R^2 attained via cross-validation are very close to each other for both training and test values proves the success of the optimum configuration attained.

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