Journal of Scientific & Industrial Research
Vol. 79, May 2020, pp. 369-371

Application of the LM-trained Model for Predicting the Retardance of Citrate Coated Ferrofluid

Jing-Fung Lin
Department of Industrial Design, Far East University, Taiwan, R.O.C.
Address: No.49, Zhonghua Rd., Xinshi Dist., Tainan City 74 448, Taiwan R.O.C.

Received 14 March 2018; revised 19 November 2018; accepted 20 January 2020

This paper has focused on developing an optimized artificial neural network (ANN) modeling to highly predict the retardance in ferrofluid of citrate coated magnetite nanoparticles. Utilizing the previously measured retardances as a training dataset, ANN models in architectures of single/double-hidden layer were trained by the Levenberg-Marquardt (LM) algorithm. From the testing of neural network with double-hidden layer (4-4-30-1); the correlation coefficient and average absolute relative error of predicted/simulated (ANN/multiple regression) retardance values were determined as 0.878 and 1.19%, respectively. Hence, a highly predictive LM-trained ANN model is obtained.

Keywords: Artificial Neural Network, Multiple Regression, Nanoparticle, Retardance

Introduction

Nowadays, iron oxide magnetic nanoparticles (MNPs) are used in various fields such as magnetic fluids, hyperthermia, magnetic resonance imaging, drug/gene delivery, and environment protection.[1–3] Surface modification of iron oxide MNPs with small organic molecules, diverse polymers and inorganic materials is a challenge.[4] The preparation of biocompatible citrate coated magnetite nanoparticles was investigated in details in the literature. Citrate coated ferrofluids (FFs) with high magnetic retardance were obtained by the adoption of Taguchi experimental design method and a developed Stokes polarimeter.[5]

The measured retardances of citrate coated FF samples were utilized to train the artificial neural network (ANN) by the gradient descent with momentum and adaptive learning rate (GDX) algorithm.[6] However, the detection of retardance around the excellent program decided by Taguchi design method were not precise enough. Moreover, the longer time for the ANN simulation was required. In this study, the Levenberg-Marquardt (LM) algorithm was particularly used for training the ANN model with single or double-hidden layer. The predicted retardance around the excellent program was well estimated by a well-trained neural network with double-hidden layer. Additionally, a comparison was made between ANN methods with a multiple regression (MR) analysis.[5]

Author for Correspondence
E-mail: jacklin@mail.feu.edu.tw

Experimental results of Taguchi design method

Considering process parameters in the co-precipitation method to produce the magnetite MNPs and finding optimal parameters for citrate (citric acid, CA) coated Fe_3O_4 FFs provided with high retardance value, the Taguchi design method was adopted. Experiments trials were conducted using a four parameter, three level (L9). Process parameters including pH of coated suspension (4.5, 5, 5.5), molar ratio of CA to Fe_3O_4 (0.03, 0.06, 0.12), CA volume (10ml, 20ml, 40ml), and coating temperature (70°C, 80°C, 90°C) were chosen. Using the intuition range analysis, the optimal parameter combination (excellent program) were assessed as coated suspension of pH 4.5, molar ratio of 0.12, CA volume was 40 ml, and coating temperature was 70°C. The orthogonal results were shown in Table 1. The excellent program was [4.5, 0.12, 40, 70] in simple representation. If it is noted that the retardance of A7 was low and negative, then it was not adopted in training neural network afterward.

ANN modeling for predicting the retardance of CA coated FF

In this study we used a feed forward multilayer perceptron (MLP) network to predict the retardance of CA coated FF. The MLP network generally consisted of successive layers, from input layer to output layer, through the hidden layer(s). The number of neurons in the input layer was 4 (4 process parameters) and the number of neurons in the output layer was only 1 (retardance) in this work. ANN models in

Table 1 — Orthogonal test results of retardance

No.	A	B	C	D	Retardance (deg)
A1	1	1	1	1	7.8228
A2	1	2	2	2	11.9872
A3	1	3	3	3	29.3618
A4	2	1	2	3	4.0525
A5	2	2	3	1	23.6294
A6	2	3	1	2	18.1662
A7	3	1	3	2	-0.4628
A8	3	2	1	3	4.5877
A9	3	3	2	1	20.2021

architectures of single/double-hidden layer were constructed in sequence. In the MLP network with single-hidden layer, tan-sigmoid transfer function was used between the input and hidden layer, and log-sigmoid transfer function was used between the hidden and output layer. In a preliminary stage, the series combination of these two transfer functions was chosen by a better training result.

The Levenberg-Marquardt (LM) algorithm combines two minimization methods including the gradient descent method and the Gauss-Newton method.[7] Gauss-Newton method is simple and has quadratic convergence rate, while gradient descent method is less dependent on the initial weights. The LM algorithm is very fast and accurate. Therefore, neural network training was done using the LM algorithm. Neural network toolbox of MATLAB software was used.

When training neural network with single-hidden layer, both training cycle (epoch) and performance goal (mean square error, MSE) were set in advance. The maximum number of training epoch (net.TrainParam.epochs) was set as 1.e5, and a performance goal (net.TrainParam.goal) was set as 1.e-7. The neuron number of single-hidden layer ranging from 4 to 14 in increments of 2 was considered. The neural network was trained by the LM algorithm for the retardance value. The root mean square error (RMSE) results of the trained network with single-hidden layer for different neuron numbers were shown in Fig. 1. While in the 4-4-1 ANN model (4 neurons in the single-hidden layer), the RMSE (predicted/measured retardances) was the lowest as 2.2916. The simulation only took 2 s to execute. The correlation coefficient R was 0.965. The average absolute relative error (AARE) was 9.35%. However, in the testing stage for the excellent program of [4.5, 0.12, 40, 70] and two programs of [5, 0.12, 40, 70] and [5.5, 0.12, 40, 70], the R value (predicted/simulated retardances, 32.70°, 33.11°, and

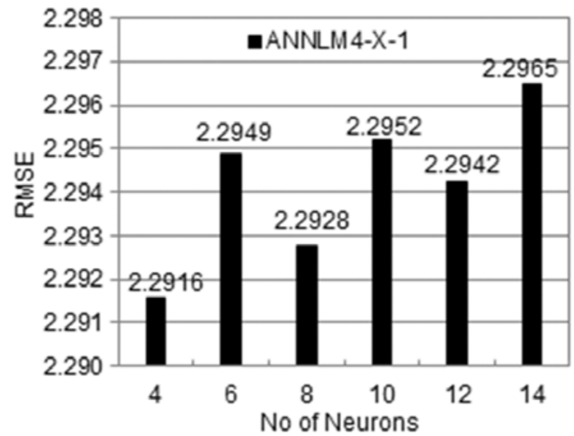

Fig. 1 — RMSE results of different neuron numbers in the single-hidden layer by the LM algorithm

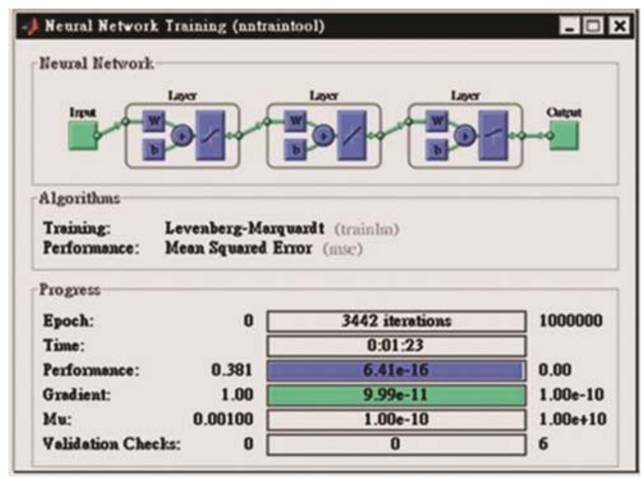

Fig. 2 — ANN trained by the LM algorithm (double-hidden layer with neurons of 4 and 30)

33.52°, which were obtained by MR[5]) was determined as -0.875 (negative) and the AARE was 1.35%.

Subsequently, in order to find a LM-trained ANN model with high generalization ability[8], the ANN model constructed by a double-hidden layer was further considered. The maximum number of training epoch was set as 1.e6, and a performance goal was set as 0. The transfer function between the second-hidden layer and the output layer was chosen as purelin. Then the neural network trained with double-hidden layer (neuron number in the first hidden layer was chosen as 4, neuron number in the second hidden layer was varied from 10 to 50 in increments of 10, 5 neurons was also included) was performed.

As evident in Fig. 2, the optimum number of neurons in the second hidden layer could be determined as 30. For the 4-4-30-1 ANN model, the training stopped at 3442 epochs with a MSE of 6.41e-16 (much close to

Table 2 — Statistical index of the ANN model with double-hidden layer (4-4-X-1)

Neuron No.	5	10	20	30	40	50
RMSE	6.100	16.042	0.529	0.429	0.527	7.016
R	-0.942	-0.864	-0.866	0.878	-0.986	-0.770
AARE (%)	16.47	28.73	1.25	1.19	1.26	17.95

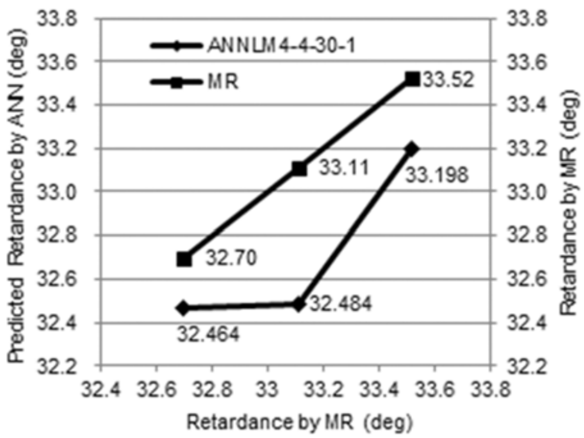

Fig. 3 — Comparisons of predicted/simulated retardances at three programs by LM-trained ANN (4-4-30-1) and MR

zero) and took 1 min and 23 s to execute. From the performance plot in Fig. 2, the 4-4-30-1 ANN model had excellent generalization ability; the MSE decreased continuously and began to rise until 3442 epochs. The AARE (predicted/measured retardances) was 9.30%, which was less than 9.35% obtained in the 4-4-1 ANN model. A double-hidden-layer neural network with high predictive ability was found.

Testing of the 4-4-30-1 ANN model around the excellent program and the comparisons with simulated retardances by MR was performed. As shown in Table 2, the RMSE (AARE) was determined as the lowest as 0.429 (1.19%), which was lower than that of 0.526 (1.35%) in the 4-4-1 ANN model. The *R* value (predicted/simulated retardances) was 0.878 (positive and >0.85 for practical applications).

Compared to the testing results (Fig. 3) obtained using GDX in the 4-6-1 ANN model[6] (AARE of 1.44% and *R* of -0.998), the obtained LM-trained ANN model (4-4-30-1) was of better accuracy (lower AARE of 1.19%) and high generalization ability (positive *R* of 0.878). Concerned with training process and accuracy[7], the LM algorithm is superior to GDX algorithm (For the 4-6-1 neural network, the training stopped at 244921 epochs with a MSE of 1.e-7 and took time over 44 min). Simulation results illustrated that LM algorithm could speed up learning process and reduce training time greatly. Therefore, it could be applied to retardance prediction. As for the reliability and validity of the proposed ANN models, they are concerned with the measurement precision of the developed Stokes polarimeter and need to be further verified by experiments indeed, respectively.

Conclusions

A LM-trained ANN model with double-hidden layer (4 neurons in the first hidden layer and 30 neurons in the second hidden layer) was well developed to predict retardance precisely. The corresponding ANN prediction was found to be in extremely good agreement with that by a MR analysis. From the testing results; *R* and AARE (predicted/simulated retardances) were determined as 0.878 and 1.19%, respectively. The developed LM-trained network for retardance was highly predictive. Above all, a certain large amount of samples with the well measured properties are required for the precise prediction in the ANN study. There is no precise methodology to determine the number of neurons in the hidden layer(s) exactly.

Acknowledgements

Thanks for the financial support of this study from the Ministry of Science and Technology in Taiwan under grant MOST 105-2221-E-269-001.

References

1 Hedayatnasab Z, Abnisa F & Daud W M A W, Review on magnetic nanoparticles for magnetic nanofluid hyperthermia application, *Mater Design*, **123** (2017) 174–196.

2 Shen L Z, Li B & Qiao Y S, Fe_3O_4 nanoparticles in targeted drug/gene delivery systems, *Mater*, **11** (2018) 324–352.

3 Saini P, Sharma R, Pant R P & Kotnala R K, Ultrafast adsorption of organic dyes by activated-carbon@Fe_3O_4 nanoscale composites: an effective solution for water purification, *Indian J Pure Appl Phys*, **56** (2018) 187–195.

4 Sonali K K, Functionalization of Fe_3O_4 nanoparticles, *Indian J Pure Appl Phys*, **56** (2018) 728–731.

5 Lin J F, Wu J S & Sheu J J, Modeling of retardance in ferrofluid with Taguchi-based multiple regression analysis, *Proc SPIE*, 2015, 930229–1–6.

6 Lin J F & Sheu J J, Modeling and prediction of retardance in citric acid coated ferrofluid using artificial neutral network, *J Magn Magn Mater*, **404** (2016) 201–208.

7 Kumar G S & Rajasekhar K, Performance analysis of Levenberg-Marquardt and Steepest Descent algorithms based ANN to predict compressive strength of SIFCON using manufactured sand, *J Eng Sci Technol*, **20** (2017) 1396–1405.

8 Maran J P & Priya B, Comparison of response surface methodology and artificial neural network approach towards efficient ultrasound-assisted biodiesel production from muskmelon oil, *Ultrason Sonochem*, **23** (2015) 192–200.