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Solubility Prediction of Gases in Polymers Using Fuzzy Neural Network Based on Particle Swarm Optimization Algorithm and Clustering Method

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ABSTRACT: A four-layer fuzzy neural network (FNN) model combining particle swarm optimization (PSO) algorithm and clustering method is proposed to predict the solubility of gases in polymers, hereafter called the CPSO-FNN, which combined fuzzy theory's better adaptive ability, neural network's capability of nonlinear and PSO algorithm's global search ability. In this article, the CPSO-FNN model has been employed to investigate solubility of CO_2 in polystyrene, N_2 in polystyrene, and CO_2 in polypropylene, respectively. Results obtained in this work indicate that the proposed CPSO-FNN is an effective method for the prediction of gases solubility in polymers. Meanwhile, compared with traditional FNN, this method shows a better performance on predicting gases solubility in polymers. The values of average relative deviation, squared correlation coefficient (R^2) and standard deviation are 0.135, 0.9936, and 0.0302, respectively. The statistical data demonstrate that the CPSO-FNN has an outstanding prediction accuracy and an excellent correlation between prediction values and experimental data. © 2013 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 129: 3297–3303, 2013

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INTRODUCTION

Solubility is one of the most important physicochemical properties, which determines the compatibility of blending system. Scrutiny of the literature[1–3] reveals the great interest that too many researchers have focused their attention on the solubility of gases in polymer melts. The data of solubility of gases in polymers obtained by experiment or prediction are essential to the optimal design process.[4] Experimental methods mainly include phase separation method,[5] pressure decay method,[6] gravimetric method,[1,2,7] volumetric method,[8,9] chromatographic method,[5] etc. However, some experiments are difficult to implement because of many restricted conditions for equipment and techniques. Considering it is costly and time consuming to measure in laboratory, there is a strong demand to develop friendly, effective, accurate, and reliable methods that can predict the solubility of gas–polymer compound. Traditional method for prediction of gas solubility in polymer mainly consists of perturbed-hard chain theory,[10] lattice-fluid theories,[5,8] cubic equation of states,[11] etc. Unfortunately, most traditional methods have some shortcomings such as large observed deviations compared with the results that obtained from experiment.[4,10]

Recent years, the interdiscipline of information science and intelligent technology has a broad application perspective.[4,12,13] With the popularization of artificial neural networks (ANN), the determination of ANN structure, parameters and bias becomes the most crucial factors because the training process of ANN could be considered as a classical optimization problem.[12] Recently, researchers discovered that many intelligent algorithms such as genetic algorithm,[13] simulated annealing algorithm,[14] fuzzy logic theory,[15] gravitational search algorithm,[16] wavelet analysis,[17] ant colony optimization algorithm,[18] particle swarm optimization algorithm (PSO),[12,19–21] chaos theory,[22] and so on, can all be used for this determination. Therefore, ANN combined with intelligent optimization algorithms namely hybrid neural network has become one of the most active subject.

So far as solubility of gases in polymers is concerned, it is affected by temperature, pressure, and sometimes it can also be affected by the interactions with the groups of the macromolecular chains.[23] As a result of the nonlinear relationship of these

factors, traditional methods of prediction of gases solubility in polymers are insufficient to meet the requirements; therefore, there is a strong demand to develop an effective and accurate prediction method for gases solubility in polymers.[1,8] Bakhbakhi[10] presented a comparison between ANN and equation of state for the prediction of solubility of 2-naphthol in ternary systems and demonstrated that the ANN method was a powerful approach with better accuracy. Recently, some literatures have mentioned the using of fuzzy theory in developing statistical estimator.[24,25] Khajeh and Modarress[26] developed adaptive neuro-fuzzy inference system (ANFIS) and Radial Basis Function Neural Network for solubility prediction of gases in polystyrene (PS) and indicated that the ANFIS had better accuracy by comparing with the classical methods. At the same time, PSO has become a popular optimizer and has been widely applied in solving practical problem.[19] Ahmadi[12] proposed a feed-forward ANN model optimized by unified PSO algorithm to predict asphaltene precipitation and indicated that the proposed model was superior.

Although these works have achieved better prediction accuracy, there are still some defects in some case. For example, the traditional ANN methods take a long time for training and it is easy to get into the local optima.[27] As far as the ANFIS is concerned particularly, the ANFIS gains an advantage in modeling nonlinear functions and is faster in convergence when compared to the other fuzzy models,[26] but when expanding the search space or increasing the fuzzy rule, or when the prerequisites of model parameters are indeterminate, the learning ability of ANFIS is still unsatisfactory.

Therefore, in this article, a faster, better learning ability and more satisfied modeling has been proposed for the solubility prediction of gases in polymers in a wide range of temperature and pressure, which involves technologies of neural network, fuzzy theory, PSO algorithm, and k-means clustering method, hereafter called the CPSO-FNN method. In the CPSO-FNN method, fuzzy neural network (FNN) trained by PSO algorithm has an excellent convergence rate; the k-means clustering method uses to determine the number of clusters with the purpose of improving the learning efficiency. The CPSO-FNN method effectively improves the shortcomings of traditional method of time consuming and easy to get into local optima. The prediction accuracy and reliability of the proposed model were checked based on the experimental data obtained from literatures. The comparison between different neural networks was carried out in detail to reveal the advantage of the proposed model.

THEORY AND ARCHITECTURE

The CPSO-FNN model is a hybrid system combining clustering method, PSO algorithm, fuzzy logic theory, and neural networks, which is considered as a special kind of neural networks. In CPSO-FNN model, fuzzy rules are generated in FNN, and then in order to avoid the curse of dimensionality as the number of input dimension gets larger,[28] a clustering method for reducing the number of fuzzy rules has been proposed. The PSO algorithm has been employed to train the FNN so as to optimize the structure and improve the learning efficiency.

Fuzzy Neural Networks

FNN is a hybrid system combining the theories of fuzzy theory and ANN, which can make use of easy interpretability of fuzzy theory as well as superior learning ability and selfadaptive capability of ANN. It has a broad application in areas of intelligent control, signal processing, prediction and forecasting, nonlinear system identification, intelligent optimization, pattern recognition, etc.[15,29] To get an intuitional understanding of FNN, Figure 1 shows a highly simplified schematic diagram of the structure of a four-layer FNN.[29]

Layer 1: Input layer. In this layer, each node represents an input variable. The node only transmits input values to the node in layer 2.

$$\mathrm{net}_j^1 = \mathrm{x}_i^1, j = i, \mathrm{y}_j^1 = \mathrm{net}_j^1 \quad (1)$$

where net_j^k is the network input of jth node in kth layer; x_i^k is the ith input in kth layer; y_i^k is the output of jth node in k_{th} layer.

Layer 2: Fuzzification layer or membership function layer. Nodes in layer 2 are arranged into some groups, each group representing the if-part of fuzzy rule. Each node computes the value of membership function.

$$\mathrm{net}_j^2 = -\frac{(\mathrm{x}_i^2 - m_{im})^2}{(\sigma_{im})^2}, j = 6(i-1) + m, \mathrm{y}_j^2 = \exp(\mathrm{net}_j^2) \quad (2)$$

where m_{im} and σ_{im} are the mean deviation and standard deviation of the ith input of the mth fuzzy subset.

Layer 3: Fuzzy inference layer or rule layer. The number of nodes in this layer is equal to the number of fuzzy sets. The automatic k-means clustering method, which tends to define the number of fuzzy rules based on the closeness of data points has been employed here. It is a method of cluster analysis that

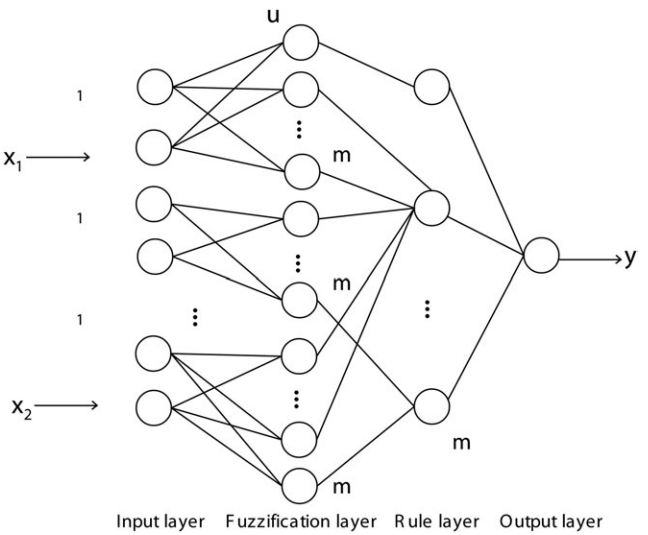

Figure 1. Structure of a four-layer FNN.

aims to partition n observations into k clusters in which each observation belongs to the cluster with the nearest mean.[28,30]

In FNN, given a set of fuzzy rules (x_1, x_2, …, x_n), where each rule replaces a node, automatic k-means clustering aims to partition the n rules into k fuzzy sets ($k \leq n$) $S = \{S_1, S_2, \ldots, S_k\}$, so as to minimize the within-cluster sum of squares:[31]

$$\arg\min_{S} \sum_{i=1}^{k} \sum_{x_j \in s_i} || x_j - u_i ||^2 \quad (3)$$

where μ_i is the mean of points in S_i:

$$\mathrm{net}_j^3 = \mathrm{x}_i^3 \mathrm{x}_m^3, j = 6(i-1) + (m-6), \mathrm{y}_j^3 = \frac{\mathrm{net}_i^3}{\sum_{i=1}^{M} \mathrm{net}_i^3} \quad (4)$$

Layer 4: Defuzzification layer or output layer. Nodes in this layer perform defuzzification. Output is the prediction value of FNN:

$$\mathrm{net}_o^3 = \sum_{i=1}^{M} \theta_i \mathrm{x}_i^4, \mathrm{y}_o^4 = \mathrm{net}_o^4 \quad (5)$$

where θ_i is the weight and y_o^4 is the output of FNN.

PSO Algorithm

PSO is an evolutionary computation algorithm inspired by social behavior and collective behavior of bird flocking or fish schooling, and swarm theory to yield the best of the characteristics among the population.[12,20,29] The search of PSO algorithm is based on the orientation by tracing a local variable (Plbest) that is each particle's best position in its search history, and tracing a global variable (Pgbest) that is all particles' best position in their history; it can rapidly arrive around the global optimum.[19,21]

The PSO algorithm works by initializing a flock of birds randomly in the search space. Every bird is called as a particle represented as a potential solution, flies through the search space with a certain velocity following the current optimum particles and finds the global best position after some iteration. Each particle can adjust its velocity and position vector based on its momentum and the influence of its best position (Plbest) and the best position of its neighbors (Pgbest).[12,16,20] The position and velocity are updated using the equations as follows:

$$v_{i,d}^{k+1} = v_{i,d}^{k} + c_1 \,\mathrm{rand}(p_{i,d}^{k} - \mathrm{x}_{i,d}^{k}) + c_2 \,\mathrm{rand}(p_{g,d}^{k} - \mathrm{x}_{i,d}^{k}) \quad (6)$$

$$x_{i,d}^{k+1} = x_{i,d}^{k} + v_{i,d}^{k+1} \quad (7)$$

where $i = 1,\ldots,m$; $x_{i,d}^{k}$ and $v_{i,d}^{k}$ are the position and velocity of ith particle at d-dimensional and the kth iteration; c_1 and c_2 are the acceleration constants with positive values, called training factor too; $p_{i,d}^{k}$ is the best position of ith particle in d-dimensional, whereas $p_{g,d}^{k}$ is the global best position.

Architecture and Evaluation

Input and output parameters of neural network should be defined for solving the practical problem.[13,32,33] As far as ANN is concerned, the required parameters include layers and the number of nodes in every layer. As a result the input and output parameters can be extracted from experimental data.[34,35] In this article, the CPSO-FNN model with four layers was designed for the prediction of gases solubility in polymers in a wide range of temperature and pressure. Therefore, two process variables (T, P) have been chosen to develop the prediction model, where T is the temperature and P is the pressure.

All experimental data for this model were obtained from literatures.[3,6,7,9,36–39] After comprehensive evaluation of the data we gathered, after removing the redundant and invalid data points, a database containing 283 data points was finally established for the CPSO-FNN model. Table 1 shows the sources of statistical data utilized in the present article.

The available data were divided into three categories by different systems of gases and polymers, which are CO_2 in PS, N_2 in PS,

Table I. Experimental Data in This Article

Polymer	Gas	Temperature (K)	Pressure (Mpa)	Solubility (g/g)	Data points	Reference
PS	CO_2	338.22-362.50	3.710-24.650	0.03714-0.19110	15	3
		372.20-383.22	2.068-42.810	0.01231-0.16056	34	3,6,7,37
		402.51-413.20	5.140-20.036	0.01462-0.20190	16	3,6
		423.15-473.15	2.159-20.151	0.00688-0.06870	37	6,7,37
PS	N_2	170.00-313.20	3.110-69.470	0.00217-0.02144	23	3,36,38
		332.20-333.23	3.050-50.110	0.00109-0.02077	21	3,36
		352.20-373.20	2.989-62.500	0.00106-0.01813	32	3,6,36
		413.20-453.20	6.452-18.011	0.00334-0.00983	13	6
PP	CO_2	373.15-373.20	2.930-12.550	0.02050-0.10860	21	9,39
		393.20-423.15	4.120-14.960	0.02260-0.10050	25	9,39
		432.20-453.20	4.960-24.910	0.03230-0.26170	30	36,37,39
		464.20-483.70	4.970-24.910	0.03020-0.24290	16	36,37,39
Total		170.00-483.70	2.068-69.470		283	

and CO_2 in PP, and then the data of each category was divided into three subsets including training set, validation set, and testing set. The training set is used to generate and train the CPSO-FNN model, the validation set is applied to verify the reliability of the model, and the testing set is used to test the prediction capability of the proposed model. The random method was proposed for the process of division of the data into subsets. In order to improve the effect of training, the training set had 70% of the data points; the validation and test sets had 15% data set each. In other words, in the 283 data points' database, 199 data points were employed to train and the rest were employed to validate and test.

The predictability of the model was evaluated by calculating average relative deviation (ARD), standard deviation (SD), and squared correlation coefficient (R^2).[4,15] The ARD and SD are defined as

$$\mathrm{ARD} = \frac{1}{N}\sum_{i=1}^{N}\frac{|\mathrm{Pre}(i) - \mathrm{Exp}(i)|}{\mathrm{Exp}(i)}100\% \quad (8)$$

$$\mathrm{SD} = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(x_i - x_0)^2} \quad (9)$$

where N is the number of data points; $\mathrm{Pre}(i)$ is the predicted value of model and $\mathrm{Exp}(i)$ is the experimental data; the x_0 is the average of the N data points.

RESULTS AND DISCUSSION

In this article, a four-layer neural network model based on fuzzy theory, PSO algorithm, and clustering method was proposed to predict the solubility of gases in polymers. For this purpose, we have built a 2-12-36-1 CPSO-FNN model, which consisted of one input layer with two nodes that represented temperature and pressure, one fuzzification layer with 12 nodes, one fuzzy inference layer with 36 nodes, and one defuzzification layer with one node represented the solubility.

The original experimental data were extracted from literature and shown in Table I, consisting the data of two gases (carbon dioxide/CO_2 and nitrogen/N_2) in PS and one gas (CO_2) in polypropylene (PP). That could also mean the database was divided into three categories by different polymers and gases, which were CO_2 in PS, N_2 in PS, and CO_2 in PP. The CPSO-FNN model had been employed to investigate their solution behavior of the three categories in this article. In Figures 2–4, the prediction of solubility of CO_2 in PS, N_2 in PS, and CO_2 in PP are plotted against the experimental data for the training, validation, and testing sets. In these figures, lines show the ideal modeling that the prediction values equal to the experimental data, whereas the asterisk, x shape, and square are the correlations between experimental data and prediction values in training, validation, and testing sets, respectively.

As shown in these figures, the outputs of the CPSO-FNN model show a fair agreement with the experimental data, no matter the training, the validation, or the testing sets. That could also mean the CPSO-FNN model has an excellent prediction capability and has a good correlation with the experimental data. Particularly, for CO_2 in PP, as observed in Figure 4, shows a better correlation between the prediction and the experiment, the close proximity of the best linear fit to the perfect fit. These results also show the superiority of the prediction of solubility by CPSO-FNN.

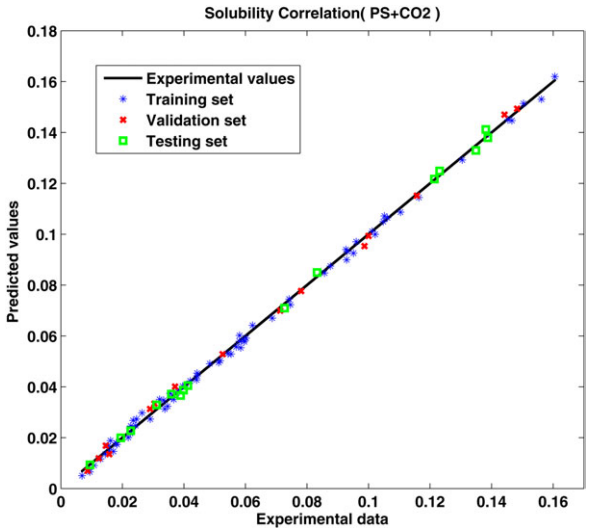

Figure 2. Predicted solubility of CO_2 in PS by CPSO-FNN vs. experimental data. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The simulation performance of the CPSO-FNN model was also evaluated by comparing with the traditional FNN model that established beforehand based on calculating ARD, SD, and R^2. It is certain that a fair comparison is achieved when these models are based on the same testing data. Therefore, another testing database with three subsets consisted of CO_2 in PS, N_2 in PS, and CO_2 in PP has been established by random method, each

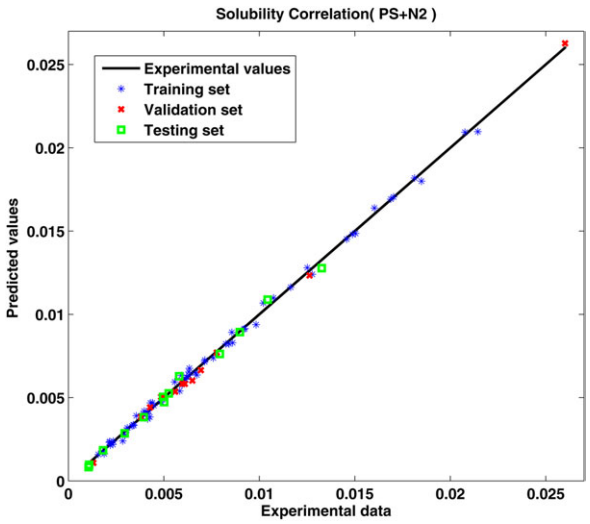

Figure 3. Predicted solubility of N_2 in PS by CPSO-FNN vs. experimental data. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

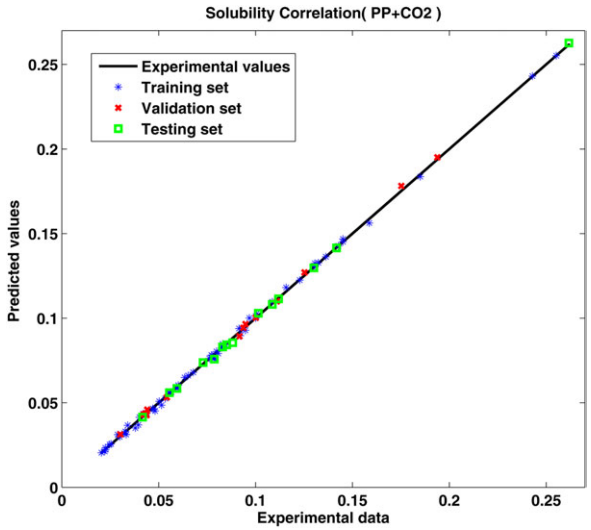

Figure 4. Predicted solubility of CO_2 in PP by CPSO-FNN vs. experimental data. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

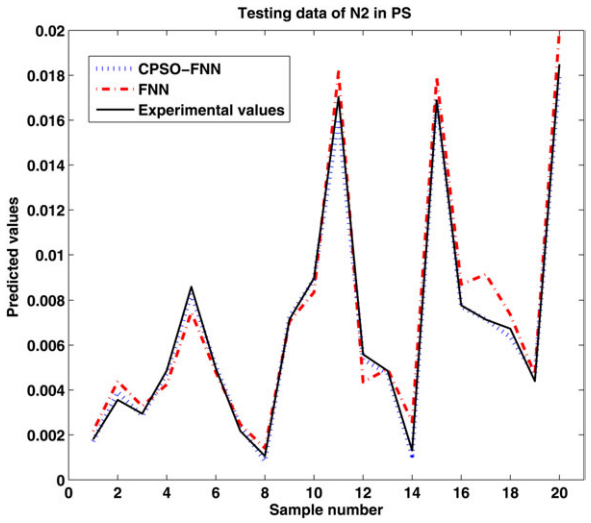

Figure 6. Comparison between experiment and prediction of N_2 in PS. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

subset contained 20 data points. The testing database was employed in this comparison. Figures 5–7 show the plots between the experimental and prediction solubility of CO_2 in PS, N_2 in PS, and CO_2 in PP by FNN and CPSO-FNN models. The statistical results of the two prediction models are compared in Table II.

Figures 5–7 and Table II reveal that, in the same testing set, the CPSO-FNN model has better performance with higher accuracy than the traditional FNN model, no matter for CO_2 in PS, N_2 in PS, or CO_2 in PP.

In order to show the predictivity of the present algorithm model, the results in this article were compared with the data obtained from associated literatures. So far as time of training is concerned, the traditional feed-forward multilayer perceptron ANN proposed by Lashkarbolooki et al.[27] needed about 500 iterations, whereas the model proposed in this article needed about 160 under the same training error. The ARD for ANFIS and Radial Basis Function Neural Network proposed by Khajeh and Modarress were 0.2543 and 0.6498 for CO_2 in PS,[26] whereas the ARD was 0.133 in this article; The R^2 for unified PSO method proposed by Ahmadi[12] was 0.99493, whereas the best value of R^2 was 0.9975 in this article. Obviously, the comparison also shows that the CPSO-FNN has better performance with higher accuracy and has better correlation and robustness.

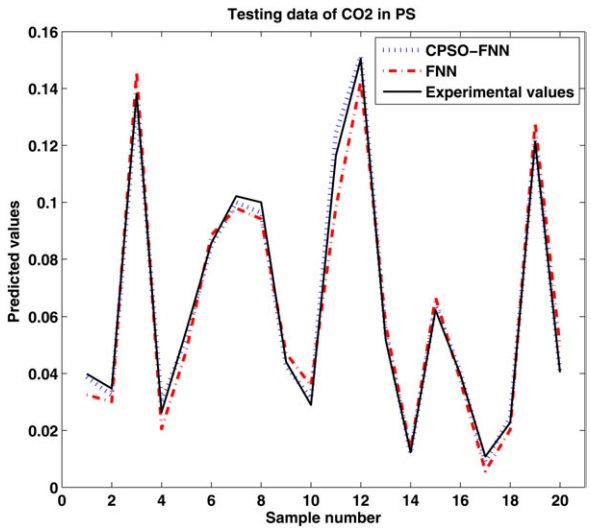

Figure 5. Comparison between experiment and prediction of CO_2 in PS. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

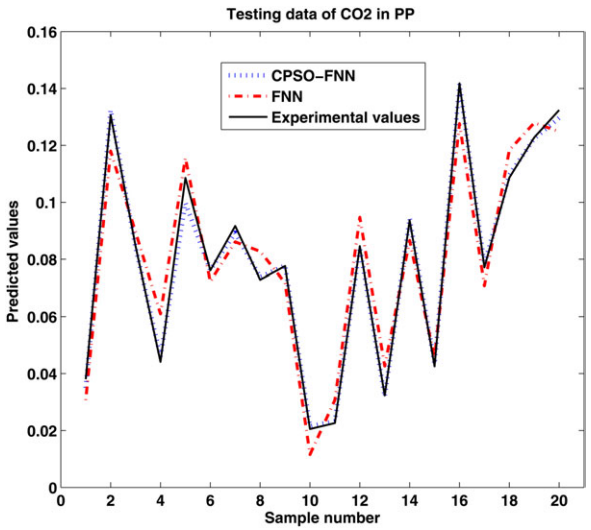

Figure 7. Comparison between experiment and prediction of CO_2 in PP. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table II. Statistical Comparison Results Between CPSO-FNN and FNN

	FNN			CPSO-FNN		
Compounds	ARD	R^2	SD	ARD	R^2	SD
PS, CO_2	0.311	0.9726	0.0718	0.133	0.9935	0.0301
PS, N_2	0.314	0.9718	0.0815	0. 175	0.9898	0.0324
PP, CO_2	0.235	0.9896	0.0603	0.097	0.9975	0.0281
Average	0.287	0.9780	0.0712	0.135	0.9936	0.0302

The major reasons for the superiority of the CPSO-FNN model should owe to the proposed training algorithm. The performance advantages of the training algorithm includes the FNN's superior learning rate, higher inference, and decision ability, the PSO algorithm's keen capability in global search and optimum performance for optimizing the structure of FNN, and the automatic *k*-means clustering method's keen cluster ability for determining the number of clusters.

CONCLUSIONS

This article presents a prediction model, which aims to replace the costly and time-consuming measurement in laboratory. The CPSO-FNN model combining the technologies of clustering method, PSO algorithm, and fuzzy logic theory has developed to predict the solubility of two gases in polymers. The performance of the CPSO-FNN model was evaluated based on calculating the ARD, R^2, and SD. The results indicate that the CPSO-FNN model is a reliable and accurate model for the solubility of gases in polymers, and is a practicable method for the analysis and design of polymer processing technology. This article shows that the proposed modeling is supposed to have high application value. In the future research works, we will follow-up on this subject all the time and focus on how to apply this model to solve more realistic problems.

NOMENCLATURE

ANN	Artificial neural network
PSO	Particle swarm optimization
FNN	Fuzzy neural network
PS	Polystyrene
PP	Polypropylene
ARD	Average relative deviation
R^2	Squared correlation coefficient
SD	Standard deviation

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