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Adaptive neuro-fuzzy inference system and neural network in predicting the size of monodisperse silica and process optimization via simulated annealing algorithm

Mehrdad Mahdavi jafari, Gholam Reza Khayati*

Department of Materials Science and Engineering, Shahid Bahonar University of Kerman, Kerman, Iran.

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* *Corresponding author email: khayati@uk.ac.ir;*

ABSTRACT

In this study, Back-propagation neural network (BPNN) and adaptive neuro-fuzzy inference system (ANFIS) methods were applied to estimate the particle size of silica prepared by sol-gel technique. Simulated annealing algorithm (SAA) employed to determine the optimum practical parameters of the silica production. Accordingly, the process parameters, i.e. tetraethyl orthosilicate (TEOS), H_2O and NH_3 were introduced to BPNN and ANFIS methods. Average mean absolute percentage error (MAPE) and correlation relation (R) indexes were chosen as criteria to estimate the simulation error. Comparison of proposed optimum condition and the experimental data reveal that the ANFIS/SAA strategies are powerful techniques to find the optimal practical conditions with the minimum particles size of silica prepared by sol-gel technique and the accuracy of ANFIS model was higher than the results of ANN. Moreover, sensitivity analysis was employed to determine the effect of each practical parameter on the size of silica nano particles. The results showed that the water content and TEOS have the maximum and minimum effect on the particle size of silica, respectively. Since, water acts as diluent and synthesis of monodisperse silica in diluent solution will decrease the growth probability of nucleate, leading to a the lower silica

Keywords: *Silica particle Size; Fuzzy inference system; Simulated annealing; Artificial neural network; Process parameters, Sol-Gel methods.*

1. Introduction

Simulation is an effective technique for optimization of engineering process instead of laboratories trials with lower cost and times. The model enables us to a better understand on the behavior of a system as well as to design the new materials with unique properties. Soft computing methods help us to simulate the nonlinear phenomena and as a consequence predicting their future behavior. Artificial Neural Network (ANN) and Fuzzy Logic (FL) are the most common of soft computing methods due to their acceptable accuracy within the simulation [1-3]. The black box nature of ANN is the most drawback of its application on the simulation of such system. Therefore, a hybrid predictive model, i.e., adaptive neuro-fuzzy inference system (ANFIS) technique proposed to incorporate the desirable attributes and eliminates of disadvantages the both method, separately [3].

Silica opals are suitable candidates for Photonic Band Gap (PBG) crystal materials due to their unique properties in light manipulation. Thus, preparation of monodispersed silica is a hot topic issue for researchers [4]. Production of monodispersed silica from organic materials has been studied for decades using sol–gel technique [5], firstly proposed by Stober and Fink [6]. Consideration of various aspect of this category is a hot issue in our previous works [7-9]. According to literatures, reaction temperature, TEOS, water and NH_4 concentrations are some of the most effective parameters in sol–gel technique. The relations between these parameters are so complex and consequently applications of advanced computing methods for determination of the complexity are so beneficially. It was necessary to note that, to avoided from the complexity of determination the effect of each practical parameter on the size of products, the effect of other affected parameters on particle size (e.g., ligands, stabilizers, feeding rate, temperature, reaction time) [10-12] were ignored and all of them adjusted to a constant value in this study.

The main contribution of this work are: (1) Feasibility study of the ANN and ANFIS simulation in estimation of particle size of silica prepared by sol-gel technique on the base of experimental data reported in literature [13]; (2) Comparison of the ANFIS and ANN models with each other; (3) Investigating the effect of TEOS, water, and ammonia concentrations on the particle size with ANFIS surface plot and scrutinizing the most affecting parameters by applying of sensitive analysis (4) Integrating the best model with simulation annealing algorithm (SAA) to optimize the sol-gel reaction with the minimum particle size of silica and (5) Verifying the predicted optimum condition.

2. Description of modeling and optimizing approach

2.1 Artificial neural network (ANN)

ANN represents an effective tool for the recognition a mapping relationship between input and outputs in a nonlinear and complex systems [14, 15]. The multi-layer Feed-Forward Neural Network with a back propagation-learning strategy is the most common structure in practical usages [16, 17]. This algorithm contain of input layer, output layer and one or higher layers with a lot of nodes, which enable the network to simulate the complexity between the practical parameters (Fig. 1).

The ANN training algorithm includes of three major steps: (1) Network training; (2) Network evaluating; (3) The back propagation strategy to create and update the connection weights. This step is repeated until the accuracy of the network maximizes and could successfully predict the output. The training data can be minimized by optimization the Neural Network structure and by selecting the appropriate input parameters. However, by increasing the complexity between input and output, the number of training data must be increased [18].

The basic principal of an artificial neuron in ANN architecture illustrated in Fig. 2, that takes output of neurons in the previous layer as input "x_0", multiples them with connection weights "w_0", adds a bias "b", fed through a transfer function to generate a result which is the neuron output "a" [19]. The important activation transfer functions

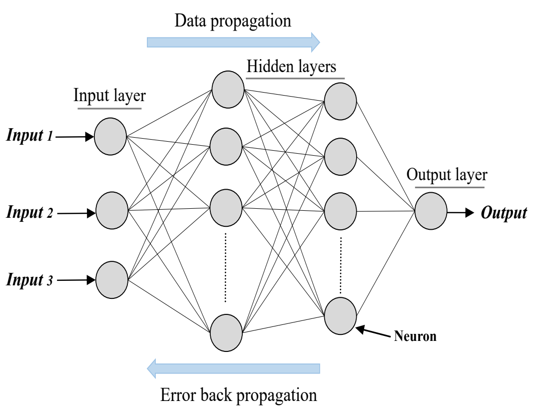

Fig. 1- Schematic representation of multilayer perceptron neural network.

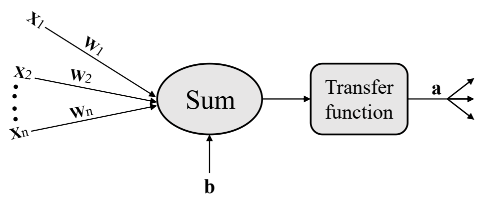

Fig. 2- A Basic Artificial Neuron.

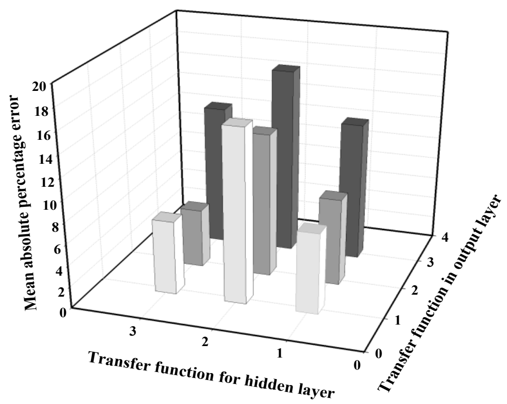

Fig. 3- Typically variation of mean absolute percentage error as a function of activation functions combinations [21].

that used for input-output fitting problems are log-sigmoidal function, hyperbolic tangent sigmoid and linear function [20]. Typically, Fig. 3 shows the mean absolute percentage error as a function of various activation function combinations [21] in which (I) activation function for hidden layer are tansig (1), purelin (2) and logsig (3); (II) Activation function for output layer are Tansig (1), purelin (2) and logsig (3); (III) Training algorithm for back propagation is scaled conjugate gradient algorithm (SCG).

A combination of these functions enables the ANN to model the nonlinear problems. Equations 1 and 2 are the hyperbolic tangent sigmoid and linear function, respectively.

$$f(x) = tan\left(\frac{1}{1+\exp(x)}\right) \quad \text{(eq. 1)}$$

$$f(x) = Bx \quad \text{(eq. 2)}$$

To the best of our knowledge, application of tan-sigmoid and linear transfer functions to regulate the weights and biases of the hidden layers as well as the output layer are so beneficially [22].

2.2. Fuzzy Logic (FL)

Artificial neural networks are the most common of soft computing. The ability of ANNs to create the knowledge on the base of limited number of trials is so beneficially for simulation of practical process [22, 23]. However, the ANN is a powerful technique when the output of process is more valuable than the relation between the practical parameters.

Fuzzy logic (FL) system utilizes human expertise in the form of fuzzy IF-THEN rules and is based on the Sugeno's architecture instead of numerical values. The "IF" part deals with generating the membership functions (MF). The "THEN" section deals with to identify the resultant variables based on the input–output relationship by one of the least squared methods, which are represented as linear combinations, i.e. $F_1 = p_1x + q_1y + r_1$ of their inputs [24]. Sugeno fuzzy model is the most common of fuzzy IF–THEN rule as follows:

Rule 1 : **IF** x is A_1 and y is B_1 **THEN**

$$f_1 = p_1x + q_1y + r_1 \quad \text{(eq. 3)}$$

Rule 1 : **IF** x is A_2 and y is B_2 **THEN**

$$f_2 = p_2x + q_2y + r_2 \quad \text{(eq. 4)}$$

In which x and y are input variables, $\mathbf{A}_i$ and $\mathbf{B}_i$ are the fuzzy sets determined for x and y, p_i and q_i are the consequent parameter of i_{th} rule and f_i is the leaner consequent function.

2.3. Adaptive neuro-fuzzy inference system (ANFIS)

Application of both of ANN and FL to construct a relation between inputs and outputs as a function is named as ANFIS is a fuzzy inference system method [25]. The basic manner that ANFIS uses for learning is the back-propagation gradient descent, which determines the accuracy recursively from the output layer backward to the input nodes. This learning algorithm is similar the back-propagation learning algorithm which used in the feed-forward neural networks [26]. The schematic representation of ANFIS illustrated in Fig. 4, include of five different layers in which each layer possesses several nodes and produce the inputs for the succeeding layer.

In the fuzzy layer, i.e., the first layer, each node creates the membership rank for every fuzzy set (like as bad, middle and good) and determines

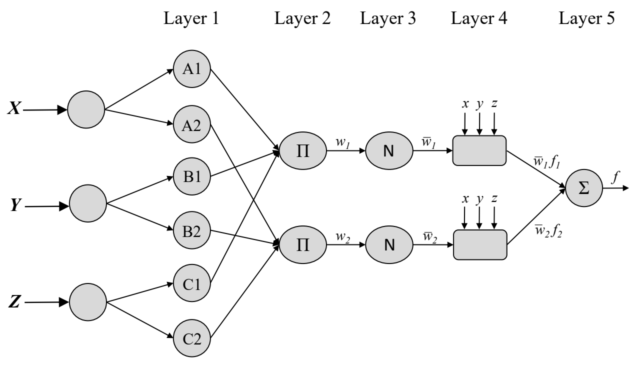

Fig. 4- Typical ANFIS architecture.

the type of MFs for ANFIS network. The layers 2–4 relates to the fuzzy rules [27]. The fifth layer expresses the overall outputs as the summation of all fourth layer signals value. Important factors that have vital effect on the accuracy of ANFIS model include the type of fuzzy based rule, the number of MFs, and the types of their MFs. In this paper, Sugeno-type fuzzy inference systems were used for predicting size of monodisperse silica particles. Thereafter, different MF types (gbellmf, gaussmf, gauss2mf, pimf, dsigmf, psigmf) are employed to estimate the best model that minimizes the mean absolute percentage error (MAPE).

2.4. Simulated annealing algorithm (SAA)

Recently, different strategies have been used to optimize the complex engineering problems. There is not unique strategy to optimize every problem. In present study, optimizers for the silica sol-gel process optimal design is proposed based on SAA. SAA is a random search methodology, which works on the base of simulation annealing process for a heat treatment of solid. The purpose of this methodology is minimizing the objective function (annealing energy) by examining of all points in data domain respect to their energy value, i.e., function value [28]. Probability function (eq. 4) enables SAA to transfer from local minimum. As eq. 5, T indicates temperature and k shows the Boltzmann constant. Let the objective function value in a specific annealing step is represented by P(E) as following:

$$P(E) = \exp\left(-\frac{E}{k\,T}\right) \quad \text{(eq. 5)}$$

The procedure of SAA is given below.

1. At the beginning of simulation, we start with an initial feasible point x_0 and consider the iteration numbers $K = 0$ and $k = 1$;
2. A new feasible point x_k is set with a random number. Then $f(x_k)$ and $\Delta f = f(x_k) - f(x_0)$ is checked;
3. If Δf value is lower than zero then x_k is determined as the new best point x_0 and go to step 4. Else, we set β value with random number that is between 0 and 1 and find the value for . If value of β is lower than p, then x_k is determined as the new best point x_0;
4. Let N shows the maximum number of trial points that are used in one iteration. If value of k is lower than N, then $k = k + 1$ and go to step 2. Else, go to step 5;
5. The process is finished when we have no acceptance after N trials. Otherwise, go to step 6;
6. If value of K is lower than the iteration limit, then decrease temperature by $\theta = \tau\theta$ (τ shows a variable less than 1). In addition, we change $K = K + 1$, $k = 1$, and go to step 2. Else, the process is finished.

3. Experimental data collection

An important part of planning an experimental work is identifying the importance variables which affecting the practical condition. Bogush et al. [13] studied the control of size and mass fraction in preparation of monodisperse silica particles. They reported that the size of monodisprese silica particles is mainly a function of 3 factors, i.e., tetraethyl orthosilicate (TEOS) concentration in the range of 0.3-0.17 M, ammonia (NH_3) content in the range of 0.5-3 M and deionized water (H_2O) in the range of 0.5-23 M were used as inputs and silica particle size as output. The data used in this work contains of 50 data from [13] that shown in Table 1. For training and testing each network, these data were randomly partitioning to the training and testing sections. Accordingly, 41 schedules were applied for training whereas 9 ones were used for testing. To increase the efficiency of networks, the input data was normalized using eq. 6:

$$Xn = 0.8 \times \frac{X - X_{min}}{X_{max} - X_{min}} + 0.1 \quad \text{(eq. 6)}$$

In which X_{max} and X_{min} are the maximum and minimum values of the independent variable X. Table 2 abbreviates the more appropriate models architecture in of ANN/ANFIS program modeling.

4. Results and discussion

4.1 Modeling development

In this section, ANN/ANFIS models are designed to estimate the particle size of monodisprese silica. To find the optimum feed-forward neural network structure with one hidden layer, a program was developed in Matlab (version 2014.b) software by changing the number of nodes in hidden layer (1-30) and training functions, i.e. scaled conjugate gradient (CGB), Levenberg Marquardt (LM) and Powell-Beale conjugate gradient (SCG). The ability of each model evaluated by mean absolute parentage error (MAPE) by the following expression:

$$MAPE = \frac{1}{L}\left[\sum_{i=1}^{L} \frac{|T_i - P_i|}{T_i}\right] \times 100 \quad \text{(eq. 7)}$$

To find the relation between input and output parameters in hidden layer, Hyperbolic tangent

Table 1- Data set of monodisperse silica particles [13]

No.	TEOS (M)	NH_3 (M)	H_2O (M)	Average diameter(nm)
1	0.17	3	10.26	587.77
2	0.17	0.5	7.58	288.89
3	0.17	1	7.00	482.19
4	0.17	1	10.00	456.86
5	0.17	1	17.00	319.48
6	0.17	2	9.13	701.38
7	0.17	0.5	3.00	156.33
8	0.17	0.5	5.00	246.24
9	0.17	0.5	10.00	273.61
10	0.17	0.5	12.00	284.12
11	0.17	0.5	12.53	318.4
12	0.17	0.5	14.54	268.83
13	0.17	1.0	2.00	137.46
14	0.17	1.0	3.00	279.00
15	0.17	1.0	5.00	446.9
16	0.17	1.0	8.00	460.71
17	0.17	1.0	12.00	416.05
18	0.17	1.0	14.00	373.09
19	0.17	1.0	15.00	382.03
20	0.17	1.0	19.00	272.17
21	0.17	1.0	23.00	223.16
22	0.17	2.0	5.10	614.01
23	0.17	2.0	5.12	703.95
24	0.17	2.0	7.12	635.21
25	0.17	2.0	7.12	671.61
26	0.17	2.0	14.11	493.14
27	0.17	0.5	0.50	12.00
28	0.17	0.5	1.00	27.00
29	0.17	0.5	2.00	82.00
30	0.17	2.0	16.12	383.71
31	0.17	3.0	8.25	592.26
32	0.17	3.0	8.25	637.23
33	0.17	3.0	13.24	542.48
34	0.17	3.0	15.25	441.62
35	0.17	3.0	15.25	480.17
36	0.17	3.0	17.26	407.15
37	0.30	0.5	3.00	99.23
38	0.30	0.5	5.00	281.86
39	0.30	0.5	7.00	468.90
40	0.30	0.5	12.00	361.76
41	0.30	1.0	14.00	403.74
42	0.30	2.0	7.14	855.73
43	0.30	2.0	9.18	761.45
44	0.30	2.0	9.13	858.15
45	0.30	2.0	14.15	542.21
46	0.30	2.0	15.16	553.31
47	0.30	2.0	16.14	522.65
48	0.30	0.5	10.00	440.66
49	0.30	2.0	11.17	733.09
50	0.30	3.0	13.21	630.02

sigmoid transfer functions was employed [29]. Since the output values (12–858) had extended ranges, a linear activation function was selected for output layer enable the structure to produce values outside of -1 to +1.

During the training phase of ANFIS, various effective parameters, i.e., the number and type of membership functions for each input variables and output parameter as well as optimization method were tested. The optimum condition using the lowest MAPE as criterion was proposed based on Matlab programing. Cross validation was employed as the stopping criterion in training section to avoid from overtraining. Fig. 5 shows the program flowchart for ANN and ANFIS.

Experimental data (target) and predicted value by ANIFS/ANN models are compared in Fig. 5. Accordingly, the accuracy of both the models is quite acceptable for training set while the testing value of ANFIS model shows higher accuracy (about 97%) respect to the experimental data. Fig. 6 clearly shows that the used ANFIS model could be capable for prediction of particle size with a minimum error within the domain covered by the training pattern. Similar results have been observed in Table 3. Fig. 7 explains the performance of the developed ANFIS model with the number of iterations for prediction of particle size. As shows, the relations between inputs and output are complex which emphasis on the necessity of iterations to minimize the root means square error during the training process.

The influence of practical parameter on the size of products is illustrated in Fig. 8. In addition, the surface plot is helpful to visualize required sol-gel parameters to achieve certain particle size. The reactant concentrations in sol–gel system had greatly affected the particles size. Due to the lower potential to increase the solid nuclei size during the precipitation in diluent solution, a smaller particle size can be produced in the lower concentration of [H_2O], [TEOS] and [NH_3]. Moreover, by decreasing the concentration of [H_2O] as well as [TEOS], the rate of hydrolysis and condensation reactions as essential step for the silica synthesis decreased and consequently the formation of smaller particle size

Table 2- Results of ANN/ANFIS program modeling

Specifications of ANN architecture				*Specifications of ANFIS architecture*			
ANN Configuration	Transfer function in hidden layer	Transfer function in output layer	Training algorithm	No. of MF for each input	Type of MF for inputs	Type of MF for output	Training method
3-[21]-1	tan-sigmoid	purelin	LM	2,2,3	dsigmf	linear	BP method

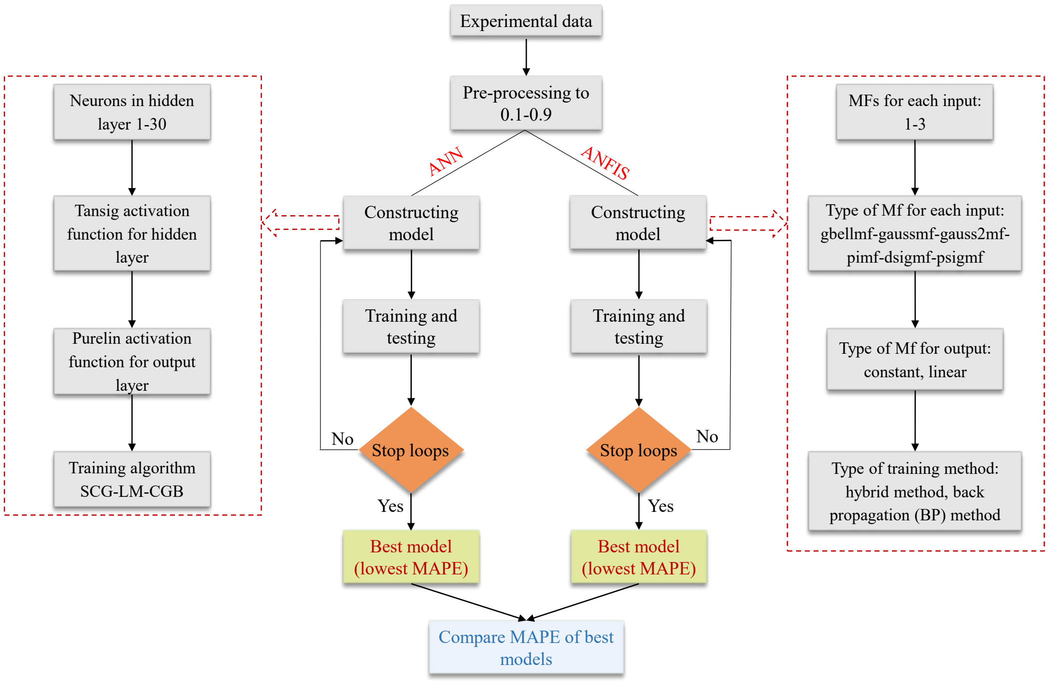

Fig. 5- Development of a model to predict the silica particle size from sol gel process parameter.

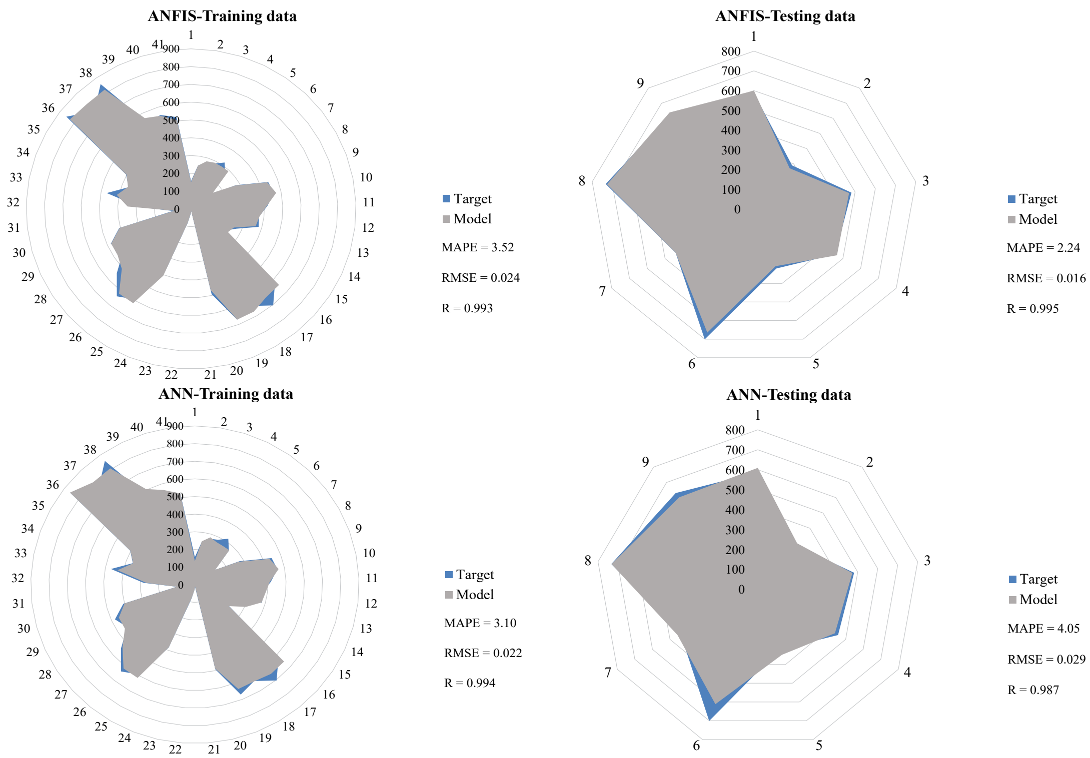

Fig. 6- Comparing the experimental data (target) and predicted target in testing and training phase using ANFIS/ANN models (The numbers on the circle and hexagonal are samples and those on the vertical axis are the particle size of the studied sol-gel conditions).

Table 3- Comparing of proposed particle size by ANN, ANFIS and experiments

Test no.	TEOS (M)	NH_3 (M)	H_2O (M)	Experimental particle size (nm)	Predicted particle size by (nm)	
					ANN	ANFIS
1	0.17	3.0	10.26	587.77	609.26	599.89
2	0.17	0.5	7.58	288.89	301.70	270.34
3	0.17	1.0	7.00	482.19	473.60	470.23
4	0.17	1.0	10.00	456.86	438.93	466.96
5	0.17	1.0	17.00	319.48	348.34	307.12
6	0.17	2.0	9.13	701.38	611.50	664.93
7	0.30	0.5	10.00	440.66	457.10	440.42
8	0.30	2.0	11.17	733.09	732.16	726.66
9	0.30	3.0	13.21	630.02	602.51	638.91

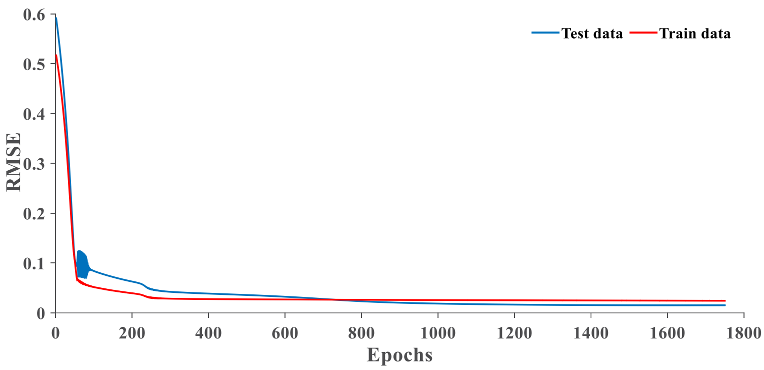

Fig. 7- The error lines of predicted particle size by ANFIS for training and testing phase.

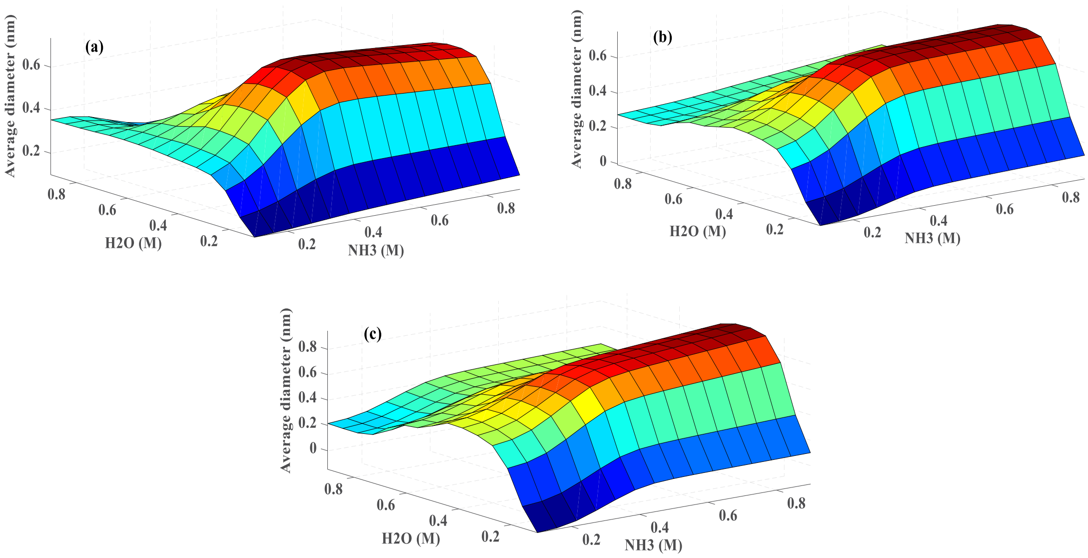

Fig. 8- Surface plot of ANFIS predicted particle size as a function of the reactant concentrations for [TEOS] = (a) 0.1 M; (b) 0.5 M; and (c) 0.9 M (i.e., the effect of two factors simultaneously on output have been drawn at fixed other factors).

encouraged [30].

As shown (Fig. 8c), the particle size values are greatly affected by the [H_2O] in a lower [NH_3]. Moreover, the [NH_3] concentration is the administrated factor on particle size when the [H_2O] content of solution decreased. Accordingly, the presence of [H_2O] and [NH_3] significantly affected the size of products. Typically, for preparation of a distinct particle size distribution, the [H_2O] value must be set at lower concentration while the [NH_3] concentration changed in the operating range. The other possible strategy is to control [NH_3] value at a lower concentration and set the [H_2O] concentration in a defined domain. This reveals that the possibility of silica particle preparation in the sub-micron size by changing [NH_3] and [H_2O] values. It seems that the effect of TEOS is lower than others. To validate this result, the sensitivity analysis was applied to determine the relative significance of practical parameters on the size of products. Moreover, the sensitivity analysis enables us to decrease the number of input parameters that haven't significant effect on the model performance as well as removing of unnecessary data collection and cost reduction.

To apply the sensitive analysis, a step-by-step technique was done on the trained ANN by changing each of the input parameter, one at a time, in a constant rate. Different constant rates (5, 10) were chosen in this paper. For every input parameter, the percentage was modified in the output as a result of the change in the input parameter. The sensitivity of each input parameters was computed by the following equation [22]:

$$S_i(\%) = \frac{1}{N}\sum_{j=1}^{N}\left(\frac{\%change\ in\ output}{\%change\ in\ input}\right)_j \times 100 \qquad \text{(eq. 8)}$$

Where S_i (%) shows the sensitivity level of an input parameter and N (= 9) is the number of datasets used for sensitivity test. According to sensitivity analysis (Fig. 9), the water content and TEOS have the maximum and minimum effect on the particle size of silica, respectively. Since, water acts as diluent and synthesis of monodisperse silica in diluent solution will decrease the growth probability of nucleate and consequently decreased the particle size of precursor, leading to a lower silica particle size.

4.2 Optimization of process using SAA

In the previous section, it has been showed that the lowest error is belonging to the 2-2-3

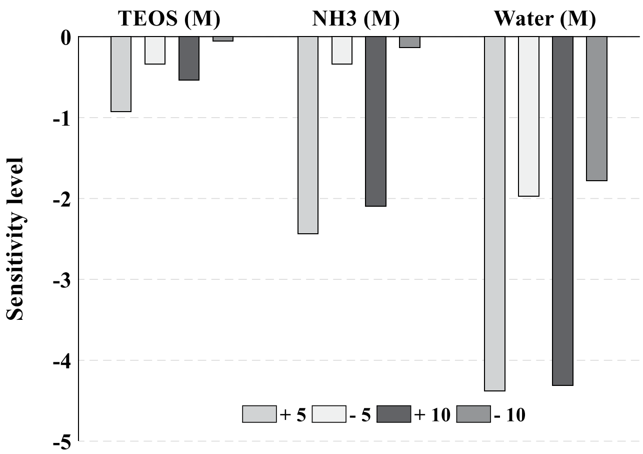

Fig. 9- Effect of input variables in silica particle size.

Table 4- Comparison of the experimental and predicted values for minimum particle size

TEOS (M)	NH_3 (M)	H_2O (M)	Particle size (nm)	
			Predicted value	Experiment value
0.17	0.5	0.5	12.56	12

ANFIS structure with dsigmf MFs. Therefore, the optimized model can be set as objective function to minimize the size of monodisprese silica particles. The flowchart of finding an optimal process of ANFIS integrated SAA is illustrated in Fig. 10. For optimization problem the objective function is defined as

Minimize $\quad$ Particle Size $= f([TEOS], [NH_3], [H_2O])$

Subject to $\quad 0.17 \le [\text{TEOS}] \le 0.3$

$0.5 \le [NH_3] \le 3$

$0.5 \le [H_2O] \le 23$

In SSA approach, Boltzmann annealing employed as annealing function that receives random steps, with size proportional to square root of temperature and exponential temperature update chosen as temperature updates function. The implementing of optimization program provided by the MATLAB commercial software tool. Simulated annealing algorithm is performed with the following settings:

Initial point x_0 = [0, 0, 0];
Initial temperature parameter (T_{init}) = 450;
Number of cycles per temperature = 100;
Boltzmann constant K=10;

The comparison of ANFIS/SAA predicted and experimental values of particle size in Table 4 reveals that this proposed algorithm is a powerful and interesting model in synthesize SiO_2 by sol-gel process. The ANFIS/SAA approach proposed the TEOS concentration of 0.17 M, NH_3 concentration of 0.5 M and H_2O concentration of 0.5 M to prepare the minimum particle size of about 12.56 nm (Table 4). As shown the minimum particle size of silica prepared at the lower concentration of reactants. Similar results have been reported according to the sensitivity analysis (Fig. 9).

5. Conclusions

This paper proposed ANN/ANFIS models for estimation of monodispersed silica particle size. Firstly, the training of network is performed using reported practical data set in literatures. According to the lowest value of MAPE as criteria, the best network is determined. Then the best model is integrated using simulation annealing algorithm (SAA) to achieve the optimal process parameters for minimization of particle size. In summary:

1. The accuracy of ANFIS model was higher than the results of ANN;
2. The H_2O content in initial solution is the

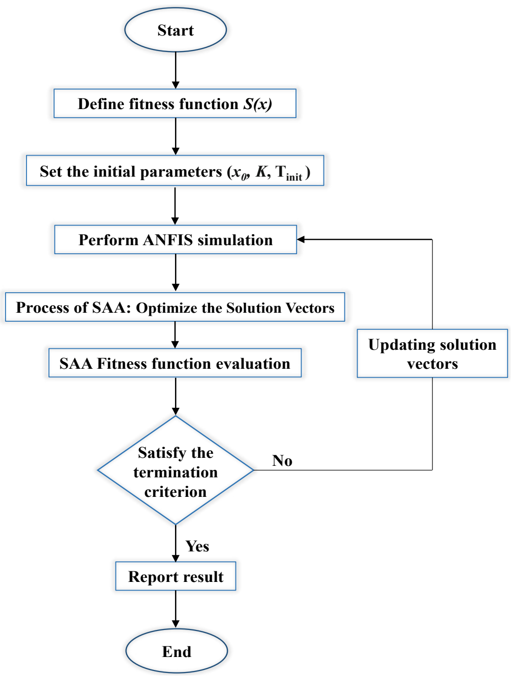

Fig. 10- The flow chart of ANFIS integrated SAA.

administrated parameter on determination of the products particle size, while the concentrations of TEOS and NH_3 have lower effect on outputs;

3. The combination of proposed ANFIS/SAA optimization method is able to predict the appropriate combinations of silica sol-gel process parameters for particle size minimization with high accuracy.

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