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Performance characterization and optimization of activated carbons are extensively studied using artificial intelligence modeling. In this study, the effect of several parameters on the preparation of activated carbon by chemical activation is investigated. Various preliminary parameters have been considered. The study has resulted in finding four parameters, which are of higher importance compared to the others. These parameters include chemical agent type, chemical agent to precursor ratio, activation temperature, and activation time. In our previous study, 36 activated carbon (AC) samples were prepared using the aforementioned parameters at various levels. In the present investigation, these experimental results have been used for the modeling.

As a novel approach, an adaptive neuro-fuzzy inference system (ANFIS) is also applied to the experimental data presented in this study. ANFIS is established by combining artificial neural network (ANN) with fuzzy inference system. After determining the model parameters, some additional data points are used to validate the models. Finally, the outcomes are compared with the experimental results. The normalized mean square error (NMSE) has been obtained as 0.00327, which is very satisfactory for the model validation.

These attempts to simulate the preparation stage of activated carbons would provide a simple and flexible route with various AC preparations. Such an effort is essential to develop the adsorbed natural gas (ANG) technology.

Keywords activated carbon; methane storage; ANFIS; Artificial Intelligence

INTRODUCTION

Activated carbon is one of the most common adsorbents in various industrial processes such as separation, purification, pollution control, and recovery. Due to energy conservation and lower pollution, separation processes are the largest consumers of this kind of adsorbent (1, 2).

Another application of activated carbon, which nowadays is very important, is the capability to adsorb and store certain gas molecules, such as hydrogen or methane to achieve non-fossil fuels.

The ability to store natural gas as an adsorbed phase in porous materials is known as Adsorbed Natural Gas (ANG) technology, which can be perceived as a solution to overcome the drawbacks of the CNG alternative. Using adsorbent materials in storage tanks, to store natural gas (containing high percentage of methane) at relatively low pressure (3.5 MPa) and room temperature, is an opportunity to produce cars and appliances with natural gas consumption competing with other products (3, 4).

Consequently, it has been confirmed by scientists that the requirements for an ANG adsorbent include high adsorption capacity, high adsorption/desorption rates, good mass transfer properties, all of which are extremely hydrophobic and inexpensive for the end user. In addition to high packing density, to ensure that the storage capacity and the energy density on a volumetric basis is high, the adsorbent should also be predominantly microporous with pores around 0.8 nm, i.e., greater than the thickness of two methane molecules, to maximize the deliverability at ambient pressure. Furthermore, an appropriate adsorbent possesses low adsorption heat and high heat capacity to minimize changes in the vessel temperature during the adsorption and desorption cycles (1).

Basically, activated carbons are produced by physical or chemical activation methods. Various researchers produced activated carbons for methane storage using the chemical activation method (5, 6). The aim of the present study is to find optimum conditions for producing activated carbons through the chemical activation method towards a high storage methane achievement.

Characterization and optimization of activated carbon have been noticeably tried by many researchers (3–11). Coal and lignocellulosic material are often used as raw materials for ACs production. Many attempts have also been made in the

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preparation of ACs from solid waste recently (5–8). In this study, anthracite coal was used as a precursor for ACs preparation.

Characterization approaches for activated carbons are usually costly processes. Therefore, researchers are looking for some new methods, which are not expensive and at the same time have reliable results for the prediction of activated carbons characteristics. The artificial intelligence methods can be considered as a suitable gadget for such investigations.

One of the admired integrated systems is an ANFIS structure, which was established by combining artificial neural network (ANN) with fuzzy inference systems. The fuzzy systems and neural networks are harmonizing technologies. The most important reason for coupling fuzzy systems with neural networks is to make use of the learning potential of the neural network. From one point of view, since a neuro-fuzzy system is found on linguistic rules, it can easily integrate previous knowledge into the system, considerably shortening the learning procedure. Since ANFIS has the possibility of solving nonlinear problems and can attain the input–output mapping effortlessly, it can be looked up as a perfect way to solve the predicting problem.

Many efforts to use artificial intelligence have been done comprehensively for empirical modeling of different chemical engineering processes (13, 15, 21). However, there have been only few efforts made (1, 2, 22) to use artificial intelligence for characterization and optimization of activated carbons. Among them, employing the adaptive neuro-fuzzy inference system (ANFIS) for such a task is quite new.

In the present investigation, new methods are employed based on ANFIS to predict the iodine numbers of activated carbons produced by Tabas anthracite (a kind of local Iranian anthracite with 85.1% carbon content). Iodine number is one of the important factors indicating the adsorption capacity of carbonaceous adsorbents.

EXPERIMENTAL

Iranian anthracite coal with 7.9 wt% ash content is used as the precursor in this study. The specification of this precursor is shown in Table 1.

Anthracite coal has been chosen, since it is an appropriate precursor for producing activated carbons with high surface areas. Chemical activation has been carried out with KOH and NaOH agents. For both carbon series, precursor with particle size of 250 to 1000 μm has been applied.

The coal powder samples were completely dried for 24 hr at the temperature of 150°C inside an oven and then crushed and sieved. Based on our previous investigation, the optimum range of chemical to coal ratio of 2–3 at 700°C temperature have been chosen. It has also been found that physical mixing of coal with chemical agent leads to the optimal result (9–12).

Since in the preparation of this particular type of activated carbon, high temperature is required and at that temperature NaOH and KOH salts convert to liquid form, there is no need to use the impregnation method. It should also be noted that in the present experimental series, the single-step activation has been carried out.

In these series of experiments, different temperatures of 670, 730, and 790°C were applied. The detailed experimental procedure was reported in the literature (13–15). In each experiment, 15 g of coal have been mixed with certain weight ratio (2, 2.5, and 3) of the above-mentioned agents. Then, samples were placed into a steel crucible inside the furnace and were heated from room temperature to the final carbonization temperature by 5°C/min under 500 mL/min nitrogen flow. Samples were kept at carbonization temperature for different activation times of 1 or 2 hr before cooling down.

The prepared samples were then washed subsequently with hot distillated water and dilute hydrochloric acid for several times to remove any remaining chemical agents. Once the activation agents were removed, samples were dried at 140°C for 24 hr.

TABLE 1
Proximate analysis of precursor (Tabas anthracite coal)

Carbon	Mineral matter (Ash)	Volatile	Moisture
85.1%	7.9%	5.9%	1.1%

MODELING

Adaptive Neuro-Fuzzy Inference System (ANFIS)

Neural Network Model

An artificial neural network (ANN) is a computer model adopted from a simplified concept of the brain (16, 17). It is a system of interconnected neurons, which in contrast to the other conventional numerical computing techniques is more powerful. This is caused by the intensive parallel processing feature of neural networks. ANN has already been used in a large number of cases as predictor, forecasting tools, controller, etc., and has shown great performances. Artificial neural network is an influential computational tool, which follows a learning procedure from a set of examples with recognized inputs and outputs (18).

Feed-Forward Artificial Neural Networks (FFNN)

In general, feed-forward ANN include a layer of input neurons, a layer of output neuronsand one or more layers of hidden neurons (19, 20). The most common ANN training method is the back-propagation (BP-ANN), which refers to the class of supervised learning methods.

Neurons in each layer are fully interconnected to the preceding and following layer neurons and each interconnection has a united connection strength named weight. The activation function employed in the hidden and output layers' neurons is non-linear. Information flows from one layer to the other in a feed-forward approach.

Since the model outputs are different from the desired responses, an error term evaluates and readjusts the weights

in an iterative process with the aim of approaching network responses close to the desired ones (21).

Fuzzy Inference System

A Fuzzy inference system (FIS) is a nonlinear system that uses fuzzy if–then rules developed by Zadeh (22). With respect to the ability of modeling the qualitative aspects of human knowledge, fuzzy logic is particularly useful in the development of expert systems.

A fuzzy set may be featured by a membership function. This function gives the grade of the membership within the set. The most commonly used membership functions are triangular, trapezoidal, Gaussian, generalized bell, and sigmoid (23, 24). Although there is one softness associated with the membership of elements in a fuzzy set, a fuzzy set is a set without obvious boundaries or without binary membership characteristics (25).

Fuzzy logic modeling techniques can be classified into two main categories, linguistic (Mamdani-type) and Takagi–Sugeno–Kang (TSK) (26). In linguistic models, both the antecedent and the consequence are fuzzy sets, contrary to the TSK model that the antecedent is made up of fuzzy sets and the consequence consists of linear equations.

ANFIS

Based on the TSK model, adaptive network-based fuzzy inference system (ANFIS) has been established (25). Since the TSK system is more convenient and computationally more efficient than a Mamdani system, it lends itself to the application of adaptive techniques for constructing the fuzzy models. These adaptive techniques can be applied to customize the membership functions to improve the fuzzy performance. ANFIS is a fuzzy inference system executed as a five layered feed-forward neural network structure shown in Fig. 1. The first and the fourth layers consist of adaptive nodes, while the second, third, and fifth layers consist of fixed nodes. The adaptive nodes are associated with their respective parameters, get duly updated with each subsequent iteration, while the fixed nodes are devoid of any parameters (27).

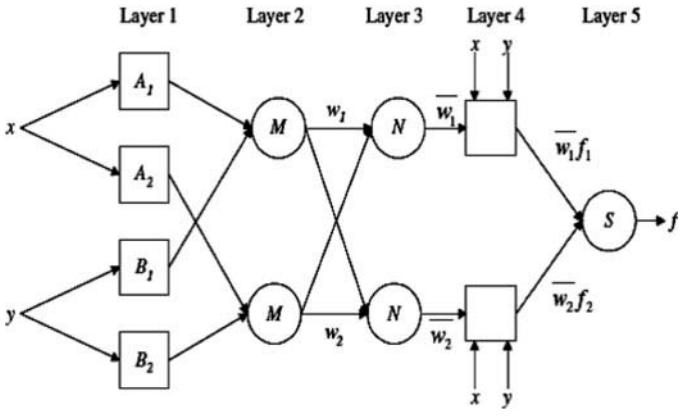

FIG. 1. Basic ANFIS structure.

The main benefit of this kind of representation is that the FIS parameters are adjusted as weights in the neural network and thus can be optimized through powerful renowned neural net learning methods. This technique is mostly suitable for the modeling of nonlinear systems. Generally, if-then rules are in the following forms:

$$\begin{aligned}&\text{Rule 1 : If (x is A1) and (y is B1) then } (f1 = p1x + q1y + r1)\\&\text{Rule 2 : If (x is A2) and (y is B2) then } (f2 = p2x + q2y + r2), \ldots\end{aligned} \tag{1}$$

where x and y are inputs, A_i's and B_i's are fuzzy sets, f_i's are outputs within the fuzzy region specified by the fuzzy rule, p_i, q_i, and r_i are design parameters that are determined during the training process.

Model Specification

Data Processing

To establish a proper model for the design, the input data have been disarranged and divided into three categories, 75% allotted for training, 12.5% for validation, and 12.5% for testing. These data include iodine adsorption capacity (iodine number) versus temperature, activation time, chemical agent type, and agent ratio.

ANFIS Structure

In order to train and validate the neural network and the ANFIS model, iodine numbers (IN) of several samples with various experimental conditions were used as input data points. Activated carbon production methods are different and are affected by various parameters. If all the data were used in one simulation program, the result would have not been satisfactory; therefore, activated carbons were classified with respect to their base (Raw materials) and production type (Physical or Chemical).

The important parameters in the production of chemically activated carbons, which have been addressed in previous work, could be listed as type of chemical agent, chemical agent to precursor ratio, activation time, activation temperature, nitrogen flow rate, drying time, particle size of precursor, mineral impurities, water content and type of precursor, and type of mixing.

Based on our experiences, the following four parameters have been chosen from the above list as the more effective ones:

1. Type of chemical agent
2. Chemical agent ratio
3. Activation temperature, and
4. Activation time.

Among them, by comparing different ANFIS structures three parameters are spotted as the most influential ones. Activation time seems to have lower effect on iodine number

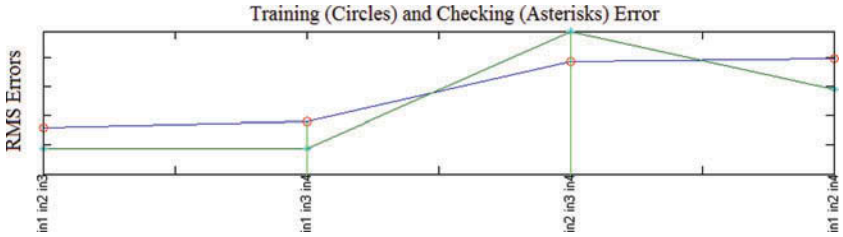

FIG. 2. Comparing various combination of parameters for selecting the more effective ones (int1 = agent type, int2 = agent ratio, int3 = activation temperature).

of ACs. The results are shown in Fig. 2. It should be noted that the interaction of these three parameters and their influences on the iodine numbers have been examined for all cases.

Two ANFIS models were employed in this study. The rules, which have been applied in both models, are given in Table 2 and the other similar training characteristics of the models are addressed in Table 3.

The first ANFIS model was designed based on the grid partitioning algorithm and a hybrid learning approach was employed to obtain a fast identification of the parameters. This method utilizes the back propagation algorithm to tune or modify the membership function parameters along with the least squares method. In this model, a simple search process was applied to tune and find the best input membership functions. The final achieved membership functions are shown in Fig. 3.

In the second ANFIS model, a Gaussian function was used as the membership function for all input variables and a back propagation algorithm was employed to train the structure. The Gaussian membership function is defined as:

$$f(x;\sigma,c) = \exp\left(-\frac{(x-c)^2}{2\sigma^2}\right) \tag{2}$$

TABLE 2
The rules applied to the ANFIS models

No.	Rule
1.	If (Agent Type is KOH) and (Agent Ratio is Medium) then (Iodine Number is out1) (1)
2.	If (Activation Temperature is Medium) and (Activation Time is High) then (Iodine Number is out2) (1)
3.	If (Agent Ratio is High) and (Activation Temperature is Medium) then (Iodine Number is out1cluster3) (1)
4.	If (Agent Ratio is Low) then (Iodine Number is out4) (1)
5.	If (Activation Temperature is Low) and (Activation Time is Low) then (Iodine Number is out1cluster5) (1)
6.	If (Agent Type is Low) and (Activation Temperature is Low) and (Activation Time Low) then (Iodine Number is out1cluster6) (1)
7.	If (Activation Temperature is Low) then (Iodine Number is out1cluster8) (1)

TABLE 3
The characteristic of ANFIS models used in the present work

Characteristic	Type
Defuzification	Weight average
And method	Prod
OR method	Probor
The number of Neurons in hidden Layers for Model (1)	
1th hidden layer	10
2th hidden layer	6
3th hidden layer	6
4th hidden layer	1
The number of Neurons in hidden Layers for Model (2)	
1th hidden layer	15
2th hidden layer	9
3th hidden layer	9
4th hidden layer	1

The membership functions for this model are presented in Fig. 4.

Model Performance Evaluation

Three types of error were employed to determine the accuracy of the model: average relative error (ARE), absolute average relative error (AARE) and normalized mean square error (NMSE). In addition, the coefficient of determination values (R^2) was used to represent the precision of our simulations. Employing NMSE error instead of MSE has been due to the high values of outputs obtained by the model. Naturally, in this case the MSE error would be deceptive. These errors are calculated through the following equations:

$$ARE = \frac{1}{N}\sum_{i=1}^{N}\left(\frac{X_{\exp.(i)} - X_{cal.(i)}}{X_{\exp.(i)}}\right) \tag{3}$$

$$AARE = \frac{1}{N}\sum_{i=1}^{N}\left(\left|\frac{X_{\exp.(i)} - X_{cal.(i)}}{X_{\exp.(i)}}\right|\right) \tag{4}$$

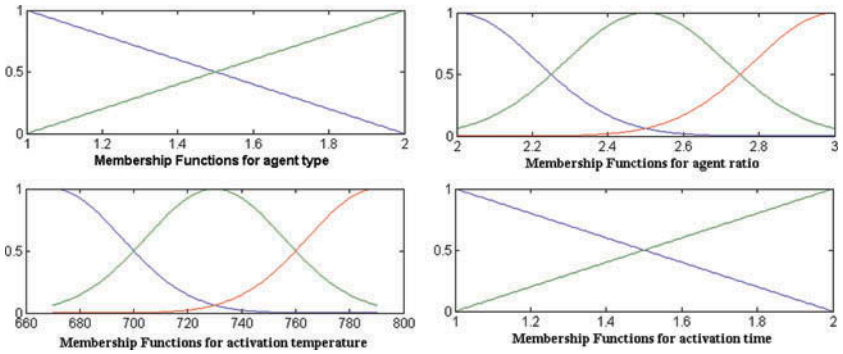

FIG. 3. The final achieved membership functions for model (1).

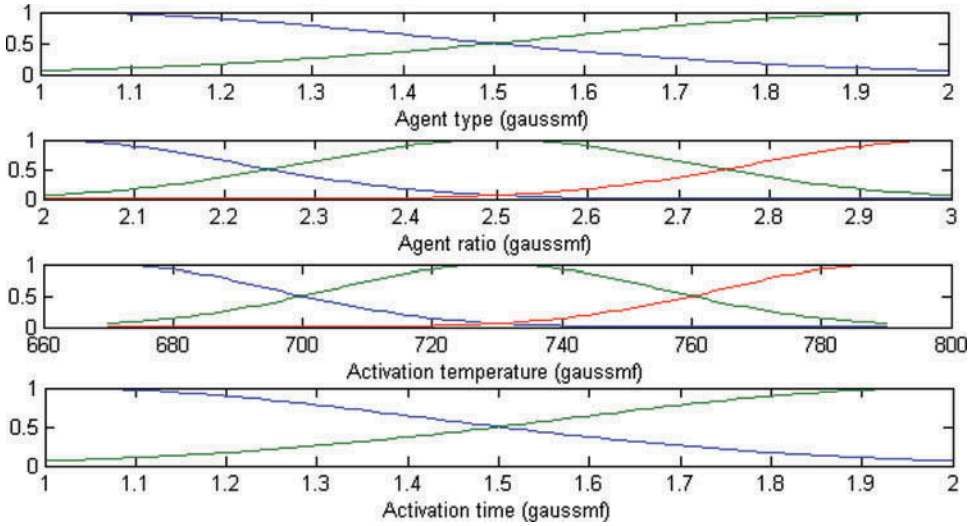

FIG. 4. The final achieved membership functions for model (2).

$$NMSE = \frac{1}{\sigma^2 N}\sum_{i=1}^{N}\left(X_{\exp.(i)} - X_{cal.(i)}\right)^2 \tag{5}$$

$$R^2 = \frac{\left(\sum XY - n\bar{X}\bar{Y}\right)^2}{\left(\sum X^2 - n\bar{X}^2\right)\left(\sum Y^2 - n\bar{Y}^2\right)} \tag{6}$$

Note that NMSE is equal to the average square of predicted error normalized by the variance.

RESULTS AND DISCUSSION

The data presented in the columns 2 to 6 of Table 4 and 5 were used to train the ANFIS model.

The simulation results are shown in Figs. 5 and 6 illustrate good agreement between the experimental data and the predicted ones.

Additionally, both experimental and predicted iodine numbers are shown in Tables 4 and 5. The small differences between these data are so obvious. In these tables the relative deviations (RDs) are presented to show the differences between predicted and actual values. RD is calculated as:

$$RD(\%) = \left|\frac{X_{\exp.(i)} - X_{cal.(i)}}{X_{\exp.(i)}}\right| \times 100 \tag{7}$$

The average statistical values of two trained ANFIS models are shown in Table 6. The comparison of determination coefficients (R^2) with similar values presented in the literature shows better prediction of our models for the activated carbon production process (3, 4, 28, 29).

As it was revealed before, there are several interactions among the activation parameters that affect AC preparation. Therefore, it is better to discuss the combination of all these parameters simultaneously in one unique qualitative factor called "degree of activation".

Figures 7–10 demonstrate the three-dimensional plots of iodine number versus input parameters, i.e., activation temperature, activation time and agent ratio.

As seen in Fig. 7, at lower NaOH agent ratios, an increase in the activation temperature leads to an increase in the iodine

TABLE 4
ANFIS simulation results for model (1)

No.	Agent type	Agent Ratio	Activation Temperature (°C)	Activation Time (h)	Experimental Iodine No.	Predicted Iodine No	RD (%)
1	KOH	2.5	730	2	2586	2586.418	0.016148
2	KOH	2.5	670	1	2308	2308.096	0.004154
3	KOH	3	670	2	2499	2499.139	0.005572
4	KOH	3	790	2	1946	1945.725	−0.01416
5	NaOH	2.5	730	1	2121	2120.774	−0.01065
6	NaOH	2	670	1	1844	1843.873	−0.00687
7	NaOH	3	790	2	555	555.0066	0.001194
8	NaOH	2.5	670	2	2054	2053.89	−0.00535
9	KOH	2	670	2	2185	2184.96	−0.00185
10	KOH	2.5	790	2	2081	2080.73	−0.01295
11	NaOH	2.5	790	1	1054	1053.934	−0.00624
12	KOH	2	790	2	2064	2063.79	−0.01015
13	NaOH	2.5	790	2	685	685.1756	0.025641
14	NaOH	2	730	2	2165	2164.885	−0.00532
15	NaOH	3	790	1	850	850.05	0.005877
16	NaOH	2	730	1	1942	1942.137	0.007063
17	KOH	2	730	2	2308	2308.008	0.000339
18	KOH	2	670	1	2080	2079.505	−0.02382
19	KOH	3	790	1	1886	1886.231	0.012261
20	KOH	2.5	670	2	2290	2289.782	−0.00953
21	KOH	3	670	1	2495	2494.154	−0.03392
22	KOH	3	730	1	2552	2552.148	0.005781
23	KOH	2	730	1	2190	2190.077	0.003515
24	NaOH	2.5	730	2	1481	1480.935	−0.0044
25	NaOH	2.5	670	1	2013	2012.922	−0.00385
26	KOH	3	730	2	2454	2453.957	−0.00174
27	KOH	2.5	790	1	2005	1887.157	−5.87746
28	NaOH	3	670	1	2258	2013.273	−10.8382
29	NaOH	3	730	1	2309	2297.21	−0.51061
30	NaOH	2	790	2	1191	1513.607	27.08704
31	KOH	2.5	730	1	2351	2419.054	2.894679
32	NaOH	3	730	2	1346	850.9606	−36.7786
33	NaOH	2	790	1	1316	1541.581	17.14142
34	KOH	2	790	1	2002	2369.921	18.37769
35	NaOH	2	670	2	1884	2021.08	7.275989
36	NaOH	3	670	2	2008	2054.521	2.316765

number up to a certain point with a sudden drop afterwards. Enhancing the iodine number is a consequence of activation improvement, caused by the temperature rise. In other words, the temperature rise intensifies the reaction between precursors and the chemical agent and results in an enhancement of the iodine number. On the other hand, since this coal is medium rank anthracite, more intensified reaction leads to pore growth. Consequently, micropores convert to mesopores leading to a significant reduction of iodine numbers. As illustrated in Fig. 7, the higher the agent ratio, the sooner the critical temperature will be reached. For example, at the agent ratio of 3, the critical temperature has been found as 740°C. Moreover, at lower activation temperatures, the higher agent ratio leads to a higher iodine number. However, at higher activation temperatures (more than 760°C), the trend is opposite and growth in the agent ratio reduces the iodine number.

Similarly, as shown in Fig. 8, for activation period of 2 hr, at lower temperatures the iodine number decreases with the activation temperature. However, while the activation temperature grows excessively, a sudden decrease in iodine number occurs

TABLE 5
ANFIS simulation results for model (2)

No.	Agent	Agent	Activation	Activation	Experimental	Predicted	RD (%)
1	KOH	2.5	730	2	2586	2585.563	−0.01691
2	KOH	2.5	670	1	2308	2308.18	0.007815
3	KOH	3	670	2	2499	2498.611	−0.01557
4	KOH	3	790	2	1946	1945.793	−0.01064
5	NaOH	2.5	730	1	2121	2120.896	−0.0049
6	NaOH	2	670	1	1844	1844.101	0.005476
7	NaOH	3	790	2	555	555.0612	0.011025
8	NaOH	2.5	670	2	2054	2054.23	0.011175
9	KOH	2	670	2	2185	2184.968	−0.00145
10	KOH	2.5	790	2	2081	2081.262	0.012585
11	NaOH	2.5	790	1	1054	1053.684	−0.02995
12	KOH	2	790	2	2064	2063.295	−0.03415
13	NaOH	2.5	790	2	685	685.2625	0.038323
14	NaOH	2	730	2	2165	2164.647	−0.01629
15	NaOH	3	790	1	850	849.8774	−0.01442
16	NaOH	2	730	1	1942	1942.005	0.000275
17	KOH	2	730	2	2308	2308.061	0.002656
18	KOH	2	670	1	2080	2080.011	0.000543
19	KOH	3	790	1	1886	1886.187	0.009919
20	KOH	2.5	670	2	2290	2290.05	0.002195
21	KOH	3	670	1	2495	2495.415	0.016635
22	KOH	3	730	1	2552	2551.788	−0.00831
23	KOH	2	730	1	2190	2190.127	0.005815
24	NaOH	2.5	730	2	1481	1481.009	0.000576
25	NaOH	2.5	670	1	2013	2012.857	−0.00713
26	KOH	3	730	2	2454	2454.155	0.006312
27	KOH	2.5	790	1	2005	1894.702	−5.50113
28	NaOH	3	670	1	2258	2116.34	−6.27369
29	NaOH	3	730	1	2309	1976.333	−14.4074
30	NaOH	2	790	2	1191	1514.867	27.19283
31	KOH	2.5	730	1	2351	2419.455	2.911732
32	NaOH	3	730	2	1346	928.825	−30.9937
33	NaOH	2	790	1	1316	1588.394	20.69864
34	KOH	2	790	1	2002	2368.566	18.30997
35	NaOH	2	670	2	1884	2026.933	7.586702
36	NaOH	3	670	2	2008	2057.593	2.469776

(i.e., in an activation temperature of 690°C and for agent ratios of 2 to 3). This fact is related to the rank of precursor.

As has been revealed in the literature, the carbonization reaction for NaOH and KOH as chemical agents is as follows:

$$6NaOH + 2C \rightleftarrows 2Na + 2Na_2CO_3 + 3H_2 \quad (1)$$

$$6KOH + 2C \rightleftarrows 2K + 2K_2CO_3 + 3H_2 \quad (2)$$

Carbonization reaction is an exothermic reaction, and consequently, the activation temperature rise causes more severity in the carbonization reaction. This brings about more microporosity and iodine number subsequently. From another point of view, since the rank of anthracite is medium (85% carbon content), as the activation temperature rises excessively, the surface of coal cannot provide the required carbon for the

TABLE 6
The statistical analysis results for two ANFIS models

Model	AARE	ARE	NMSE	R^2
Model (1)	3.593	0.584	0.00146	0.95
Model (2)	3.736	0.4409	0.0015	0.94

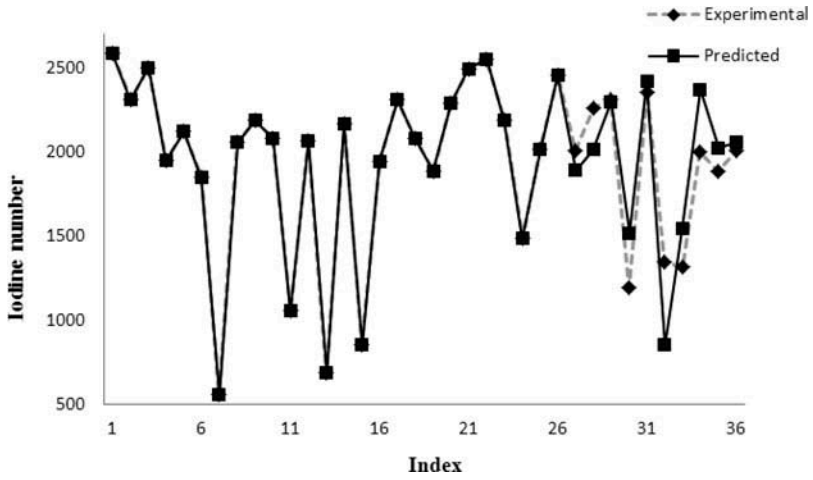

FIG. 5. Comparison between the experimental and predicted iodine numbers in model (1).

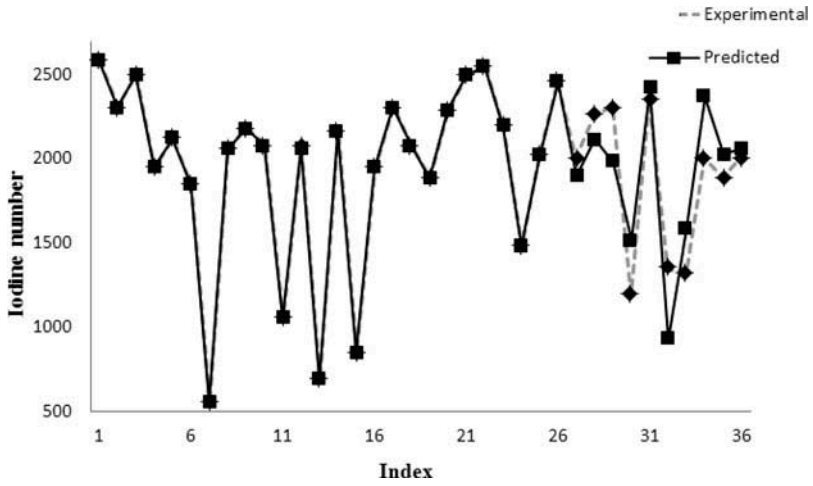

FIG. 6. Comparison between the experimental and predicted iodine numbers in model (2).

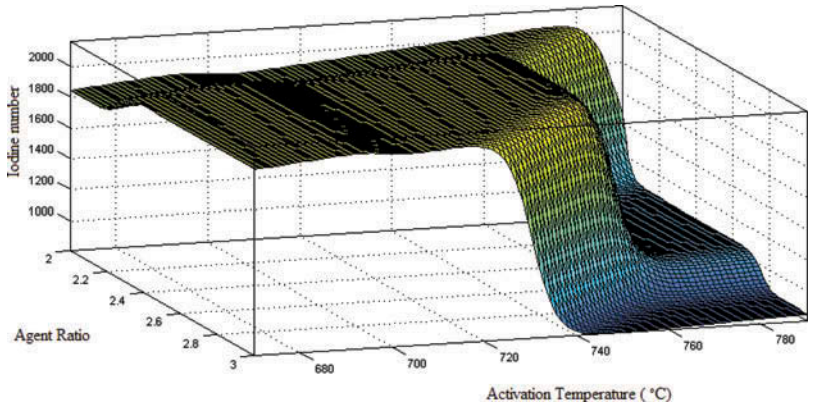

FIG. 7. Three dimensional plot of iodine number versus activation temperature and agent ratio, for NaOH agent and activation time of 1 hr.

carbonization reaction. Thus, the chemical agents commence reacting with carbon atoms inside the pores, resulting in pore widening and changing micropores to mesopores. This progress causes the iodine number to decrease at the first stage and then leads to decomposition of the activated carbon structure in the next step.

According to Fig. 8, for activation time of 2 hr, due to the enhancement of activation degree in comparison with the activation time of 1 hr, the sudden drop in iodine number achieved at lower temperature and agent ratio. As discussed before, other researchers have also found a similar procedure.

Analysis of 3D plot for KOH agent at 1 hr activation time (Fig. 9) indicates that while the activation temperature is below 760°C, the iodine number increases with agent ratio. But, at higher temperatures, the iodine number has an opposite trend with respect to the agent ratio.

On the other hand, when activation time increases to 2 hr, the optimal degree of activation is achieved earlier (Fig. 10). Finally, it should be noticed that the optimal degree of activation for NaOH agent is reached sooner in comparison with the KOH agent.

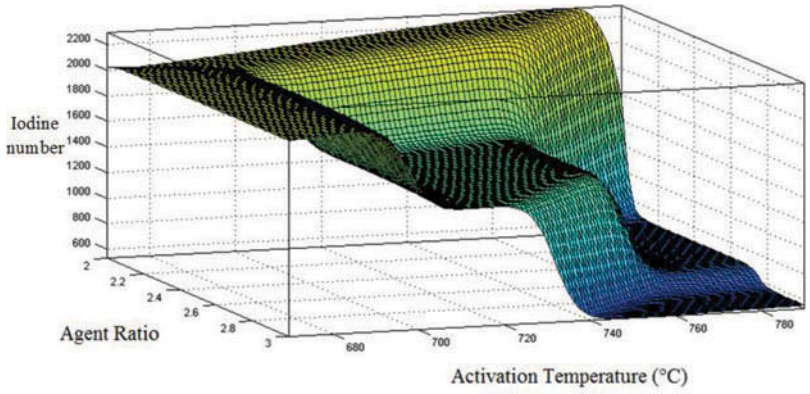

FIG. 8. Three dimensional plot of iodine number versus activation temperature and agent ratio, for NaOH agent and activation time of 2 hr.

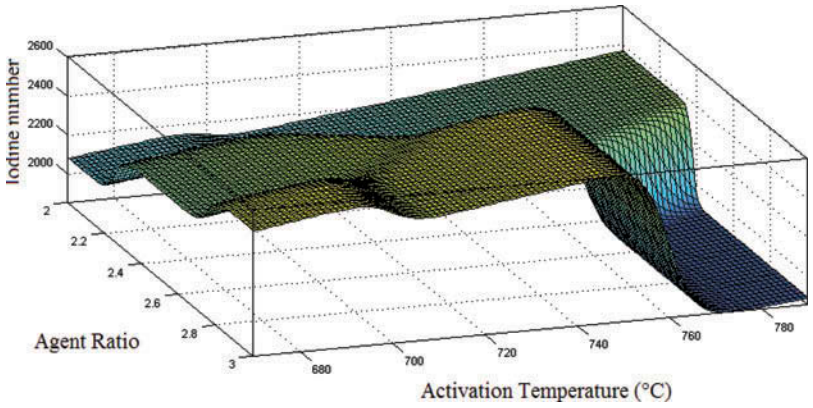

FIG. 9. Three dimensional plot of iodine number versus activation temperature and agent ratio, for KOH agent and activation time of 1 hr.

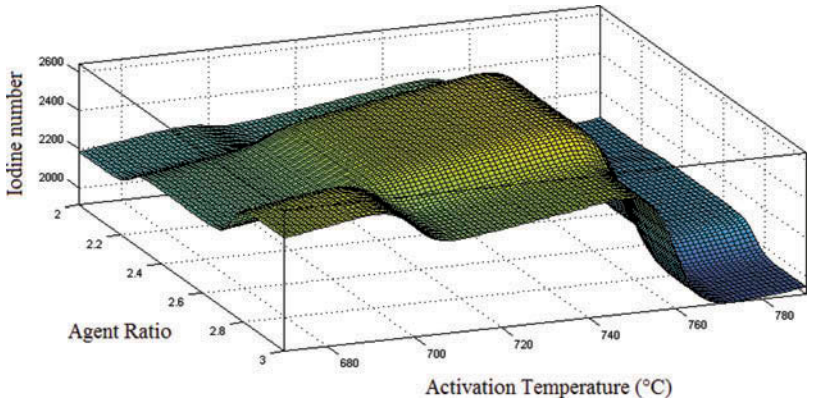

FIG. 10. Three dimensional plot of iodine number versus activation temperature and agent ratio, for KOH agent and activation time of 2 hr.

To the best of our knowledge, the performance of degree of activation is similar to the behavior of the iodine number. Thus, accurate prediction of the degree of activation is beneficial for the prediction of the iodine number, which could direct us to appropriate ACs for methane storage.

Analysis of the effect of activation degree on ACs pore structures indicates that there are two activation mechanisms for two distinct ranges of pores. Within the first range and for low degrees of activation, the micropore volume increases mainly by creation of new micropores possessing similar average widths. For higher degree of activation, the micropore volume decreases through the widening process of already existing micropores. While ultra micropores and mesopores are almost unaffected, changing the degree of activation from low to medium leads to the development of super micropores. Such a growth in the super micropore volume enhances methane storage in the AC products. However, enhancing pore volume reduces the apparent density of ACs.

Besides, the model predicts that as activation temperature increases from 760°C to 790°C, especially in the case of NaOH, the iodine number will reduce. Therefore, this activation temperature range is not appropriate for producing ACs for methane storage.

In overall, controlling the preparation condition and the degree of activation helps us to prepare ACs with desirable iodine numbers. The optimum iodine number would lead to find maximum methane storage.

CONCLUSIONS

In this study, ANFIS models are provided to evaluate qualitatively the effect of preparation parameters on activated carbons prepared from the local anthracite coal. For this purpose, the iodine numbers were predicted under various experimental conditions by changing major parameters. Initially, by referring to the previous results, four parameters have been chosen. By carrying out the process, the subsequent parameters were considered as the more effective ones: "agent type, agent ratio, activation temperature, and activation time."

Later on, with the help of ANFIS models three of these mentioned parameters were spotted as the most influential.

Finally, the model indicated that as activation temperature increases from 760°C to 790°C, especially in the case of NaOH, the iodine number reduces. Due to a decrease in specific surface area, samples prepared at this temperature range are not appropriate for methane storage. This conclusion is consistent with the results obtained previously.

In addition, the current study might be the starting point to achieve better ways for modeling the effective parameters in activated carbon production and thus reduces the number of experiments. This means lower cost and time saving.

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