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NEURAL NETWORK MODELING OF THE SULFUR DIOXIDE REMOVAL BY ACTIVATED CARBON SORBENT

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ABSTRACT: - An artificial neural network (ANN) model of three-layers was advanced to predict the efficiency of the sulfur dioxide (SO_2) removal from the flue gas stream (SO_2+air) in a fixed bed reactor using granulated activated carbon sorbent. The experimental data were collected from varying six process variables, namely, initial SO_2 concentration, reaction temperature, flue gas flow rate, sorbent particle size, bed height and reaction time. The data were used to create input-base information to train and test the NN strategy. Back propagation algorithm with two hidden layers was used for training and tests the NN. The neural network predictions of SO_2 removal efficiency agree with experimental data with the minimum mean squared error (MSE) for training and testing with values of $0.112*10^{-4}$ and $0.817*10^{-3}$, respectively.
Keywords: *Artificial Neural Networks, Sorption, Removal Efficiency.*

1- INTRODUCTION

The growth of industrialization makes it imperative to reduce the amounts of sulfur dioxide (SO_2) released into the atmosphere. Sulfur dioxide is produced from some chemical processes, mainly fossil fuels combustion. Many attempts have been made for developing a new methods to control the SO2 emissions into the atmosphere [1, 2]. The removal of the SO_2 from flue gases may be obtained by chemical or physical means, using gaseous, liquid or solid substances. The employment of activated carbons as sorbent materials for removal SO_2 is becoming increasingly to a greater extent widespread in various applications of industrial [1-3].

Recently, a sustained attempt has been made in the direction of finding the modeling of the kinetics of the sulfation reaction. The concept of modeling should be developed for an efficient operation to predict the reaction efficiency and design. ANN has been used comprehensiveness in chemical engineering as a tool of powerful modeling in different processes such as membrane filtration [4-10], ultra filtration [11-13], reverse osmosis [14], water treatment [15-18] and flue gas desulfurization [3,19-20].

In continuation of previous experimental work on SO_2 removal from flue gases [21-22]. The present paper reports the suitability models of a three layer neural using a back propagation (BP) to predict the removal efficiency of activated carbon for SO_2. The data had been collected for experiments were initially divided on four types; training, testing, interpolation and extrapolation. Finally, the output obtained from the ANN prediction was compared with the experimental data.

2. EXPERIMENTAL WORK

The experiments were performed on a laboratory scale fixed bed reactor following the previous documented work [21]. The test section is a QVF column, 7.5cm inside diameter and

50cm long. Industrial grade, activated carbon (AC) was used as sorbent with the part size (0.7) cm, (1.5) cm. The required amount of the sorbent was fixed in the center of the reactor by borosilicate glass wool. The external wired heater was heating up the reactor to the desired temperature and bed temperature monitoring by thermocouple. The Humidified air stream was combined with the required concentration of SO_2 gas and fed to the reactor. The outlet SO_2 gas concentration was determined using the titration method. The iodine was titrated with the sodium thiosulfate to calculate the SO_2 concentration in the outlet stream. The schematic diagram of the experimental setup is shown in Fig. (1). the amount of titrated iodine is equal to the residual SO_2 in the trap of analysis section. The SO_2 removal efficiency was calculated from the ratio of SO_2 concentration that was removed by AC to the initial concentration of SO_2 gas fed to the bed as shown in the equation below:

$$\eta\,(\%) = \frac{C_0 - C_{so_2}}{C_0} * 100 \qquad \ldots.(*)$$

η: SO_2 removal efficiency
C_0: Initial concentration of Sulfur Dioxide (ppm)
C_{SO2}: Effluent concentration of Sulfur Dioxide (ppm)

3. NEURAL NETWORKS MODELING

The artificial neural network is important role in modeling and predicting the linear and nonlinear problems in several engineering field. Many researchers sought to develop models from the experimentally data using statistical techniques.

3.1. Background of Neural Network – Multiplayer – Perceptron Neural Network

The neural network exemplary architecture of multilayer perceptron is illustrated in Fig. (2). The data are divided into training, testing, interpolation and extrapolation database, the status variety getting good description. The SO_2 removal efficiency is predicted by ANN Levenberg-Marquardt BP models using a MATLAB program version 7.10. The Levenberg-Marquardt algorithm determines the best weight connection by permitting error to release from layers (output toward hidden to input). The errors are calculated by comparing the output results with desired results. The training process persists for weight adjusting until the output of a network equals with an output of desired. Reducing the difference between network output and desired output by altering the weights and biases. The training process is ended at difference drop lower a specified target. Statistical parameters such as mean square error (MSE) and correlation coefficient (R^2) values are used for evaluating the performance of the network prepared as follows[23]:

$$MSE = 1/N \sum_{t=1}^{N} \left(t_j - o_j\right)^2 \qquad \ldots.(1)$$

$$R^2 = 1 - \left[\frac{\sum_j (t_j - o_j)^2}{\sum_j (o_j)^2}\right] \qquad \ldots.(2)$$

Where t is a target value, o is an output value, and N is a pattern.

Therefore, the performance of the network measures by using MSE as the error function. Consequently, network possesses maximum R^2 and minimum MSE is chosen as better NN model [24].

3.2. Backpropagation Network Algorithm

The prediction, of the multilayer feed forward ANN training involves conditioning input–output examples to the network and minimizing the objective function (i.e., error function) by use optimization method a first-order or a second-order. Training can be shaped as one of minimizing a function of the weight, the sum of the nonlinear least squares between the observed and the predicted outputs defined by the following equation:

$$E = \frac{1}{2} \sum_{p=1}^{n} (Y_o - Y_p)^2 \qquad \ldots.(3)$$

Where: n is the observations, Y_o is the target output, and Y_p the predicted output. In the BP training, the steepest decent method is used to reduce the error and to calculate the gradient of the error function by applying the chain rule on the hidden layers of NN. The hidden layer involves M neurons, and the network is presented by Eq.(4) and Eq.(5) [25]:

$$net_{PJ} = \sum_{I=1}^{N} W_{ji} X_{pi} + W_{jo} \quad \ldots.(4)$$

$$g(net_{PJ}) = \frac{1}{1+e^{-net_{PJ}}} \quad \ldots.(5)$$

Where: net_{PJ} is the weight inputs into the jth hidden unit, n is the total number of input nodes, W_{ji} is the weight from input unit i to the hidden unit j, x_{pi} is a value of the ith input for pattern P, W_{jo} is the threshold (or bias) for neuron j, and $g(net_{PJ})$ is the jth neuron's activation function assuming that g is a logistic function. The input units do not entire the operation on the data, but simply pass it onto the hidden nodes. The output unit receives a net input of:

$$net_{PK} = \sum_{J=1}^{M} W_{kj}.g\left(net_{PJ}\right) + W_{ko} \quad \ldots.(6)$$

$$y_{pk=}g(net_{Pk}) \quad \ldots.(7)$$

Where: M is the number of hidden units, W_{kj} represents the weight connecting the hidden node j to the output k, W_{ko} is the threshold value for neuron k, and $y_{pk}$is the kth predicted output. The set of weights W_{ji} is finding by The ultimate goal of the network training, connecting input units I to the hidden unit j and W_{kj}, connecting the hidden unit j to output k, that minimize the objective function in Eq.(1) [26]. Since Eq.(1) is not lucid function of the weight in the hidden layer, the first partial derivatives of E in Eq.(1) is evaluated with respect to the weights using the chain rule, and the weights are moved in the steepest descent direction. That is designed mathematically as follows:

$$\Delta W_{kj} = -\eta \frac{\partial E}{\partial W_{kj}} \quad \ldots.(8)$$

Where: η is the learning rate that measures step size. The approximate, in back propagation training contains in selection η in conformity with the relation 0\η\1. For adjusting the weights and biases of the network the learning rule is a measure [25].

3.3. Model Structure and Parameters for Neural Network

The model of artificial neural network in this paper has six neurons in input layer and one neuron in the output layer as illustrated in Fig.(3). The values for input layers are the initial SO_2 concentration, reaction temperature, flue gas flow rate, sorbent particle size, bed height and reaction time. The removal efficiency of SO_2 represents the value of the output layer. The input parameter range has been shown in the table (1).

In the hidden layer the transfer function tangent sigmoid (*tansig*) and in the output linear transfer function (*purelin*) is used. Through iterations, the model was trained. Tested the trained model with the input values, and the results of predicted were close to the experimental results.

NN model performance mostly to rely on the architecture and parameter settings of the network. Finding this optimal network architecture is hard tasks in studies of ANN that is founded on specification of values of ideal layers and neurons for hidden layers through a trial-and-error approach [27].

Matlab ANN mathematical software was used in ANN applications. Which treats the trial-and-error process; a program has been developed in Matlab to overcome optimization difficult [28]. Both first and second hidden layers the program tries various numbers of layers and neurons in the hidden layer.

4. RESULTS AND DISCUSSION

4.1. Interpolation

The SO_2 removal efficiency computed from the train and test of ANN plotted against the experimental results, as shown in Fig.(4). The MSE for training and test set was $1.1212*10^{-5}$ and $8.1753*10^{-4}$, respectively. On the other hand, the R^2 for training and testing

set was 0.99991and 0.99924, respectively. The ability of the NN to predict the data not used in training are demonstrated in Fig.(4b). Fig.(4) shows the results of the NN activity as a strong modeling tool. Evaluating the ability of NN to interpolate, a sorbent activity test was conducted within the ranges of the six process variables displayed in table (1). Different interpolation parameters were used to interpolate the SO_2 removal efficiency as shown in table (1). The flue gas flow rate was set to be 30 (l/min), which is inside the range of flue gas flow rate used for training (20-70 l/min) can be shown in a table (2). Fig.(5) showing distinguished agreement and to prove the model's capability to interpolate the multidimensional input space of the sulfation process with satisfactory accuracy. The values of MSE are $5.6148*10^{-6}$ and 0.0026 for Fig.(5a) and Fig.(5b) respectively.

4.2. Extrapolation

Although the NN have excellent interpolation properties, it can offer reliable predictions outside the training or test domain because of its empirical nature. A test of significant of the predictive capability of the NN is the range to which it can predict efficiency of SO_2 removal outside its training domain. Fig.(6) and Fig.(7) shows the results for two cases whose input conditions were outside the ranges of the training domain can be seen in table (3 and 4). In the case 1, Fig.(6 a,b) shows the efficiency of SO_2 removal of a sorbent activity test carried out with the same all parameters used in the training phase except the flue gas flow rate used at 80 (l/min). In the case 2, shown in Fig.(7 a,b) all parameters were inside the training domain while the initial SO_2 concentration was set at 2000 ppm. It is obvious that the predictions given by the NN are very close to the actual values, yielding the values of MSE for case 1 Fig.(6a) and Fig.(6b) are $7.6661*10^{-5}$ and $1.0608*10^{-4}$, respectively. For case 2 the MSE for Fig.(7a) and Fig.(7b) are $8.0204*10^{-5}$ and $8.1322*10^{-5}$, respectively.

The excellent extrapolation properties of the NN may be ascribed to the fact that at the higher flue gas flow rate of 80 (l/min) and the higher SO_2 inlet concentration (2000) ppm tested the kinetic profiles still closely resemble those inside the training domain. Apparently, the training set used in this work contained a large enough database to allow extrapolation. This suggests that when sufficiently large training sets of data are available, purely empirical NN are able to give accurate predictions on extrapolation.

Fig.(8) shows the SO_2 removal efficiency calculate from NN against the corresponding experimental data at constant reaction temperature of 80 ^{0}C, constant bed height of 18 cm, constant sorbent particle size of 0.7 mm and constant initial concentration of SO_2 of 500 for different flue gas flow rates for an hour interval. It can be seen the maximum overall removal efficiency can be obtained at 40 ℓ/min flue gas flow rate. Beyond this value the overall removal efficiency decreases.

It's clear that the model is suitable for predication of the SO_2 removal efficiency and the resulted, values were very closed to experimental values. The best value of MSE is $2.0026*10^{-4}$.

The Kinetic behavior varied with flow rate and desulfurization property was controlled by diffusion at flow rates below 40 ℓ/min, and controlled by adsorption or catalytic reaction at flow rates above 40 ℓ/min.

5. CONCLUSIONS

The feasibility of using a neural network to predict the SO_2 removal efficiency from flue gas stream (SO_2 + air) in a fixed bed reactor using activated carbon sorbent. The predictions of the neural network shown an excellent agreement with the experimental data based on the category of MSE for both training and testing. The neural network is shown to give comparable predictive capability when used for interpolation and extrapolation, for a variety of reactions.

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Table (1) The range of the input parameters in ANN models.

Inputs	Range
Initial SO_2 concentration (ppm)	500-1500
Reaction temperature (^oC)	30-80
Flue gas flow rate (l/min)	20-70
Sorbent particle size (mm)	0.7-1.5
Bed height (cm)	10-24
Reaction time (min)	0-60

Table (2) The parameters value used in the interpolate of the SO2 removal efficiency for ANN model.

Inputs	Interpolation a	Interpolation b
Initial SO_2 concentration (ppm)	500	500
Reaction temperature (^oC)	30	80
Flue gas flow rate (l/min)	30	30
Sorbent particle size (mm)	1.5	0.7
Bed height (cm)	24	18
Reaction time (min)	0-60	0-60

Table (3) The parameters value used in the extrapolation of the SO_2 removal efficiency for ANN model.

Inputs	Extrapolation a	Extrapolation b
Initial SO_2 concentration (ppm)	500	500
Reaction temperature (^oC)	80	30
Flue gas flow rate (l/min)	80	80
Sorbent particle size (mm)	0.7	1.5
Bed height (cm)	18	24
Reaction time (min)	0-60	0-60

Table (4) The parameters value used in the extrapolation of the SO_2 removal efficiency for ANN model.

Inputs	Extrapolation a	Extrapolation b
Initial SO_2 concentration (ppm)	2000	2000
Reaction temperature (^oC)	80	80
Flue gas flow rate (l/min)	20	20
Sorbent particle size (mm)	1.5	0.7
Bed height (cm)	24	24
Reaction time (min)	0-60	0-60

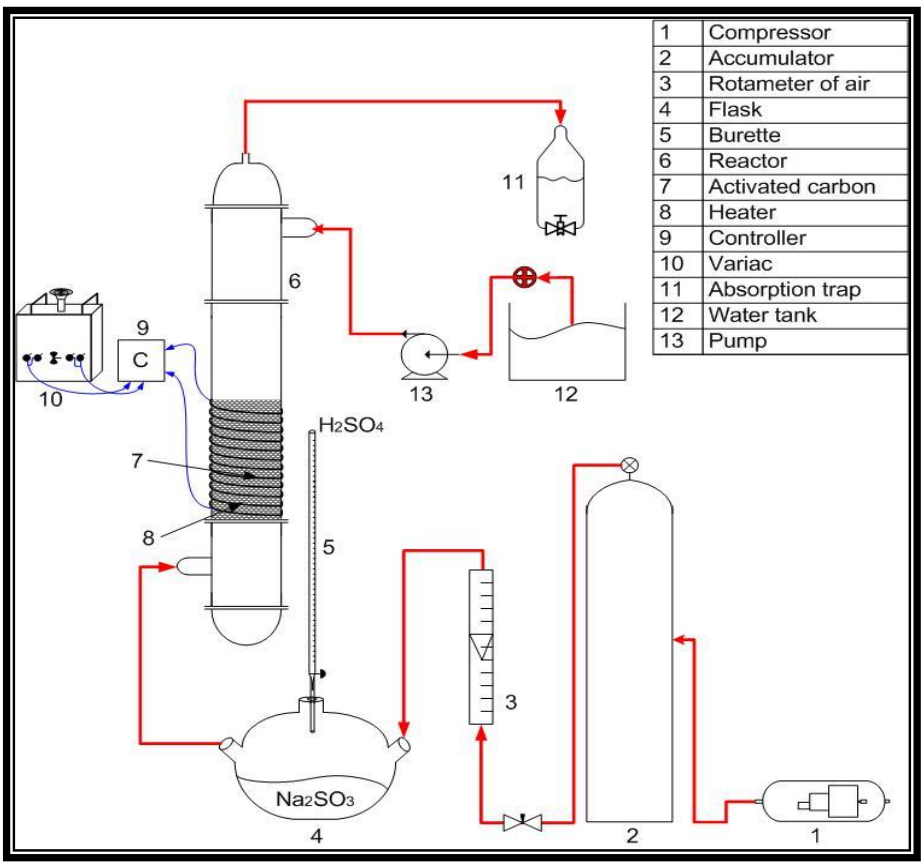

Fig. (1) Schematic diagram of the experimental setup

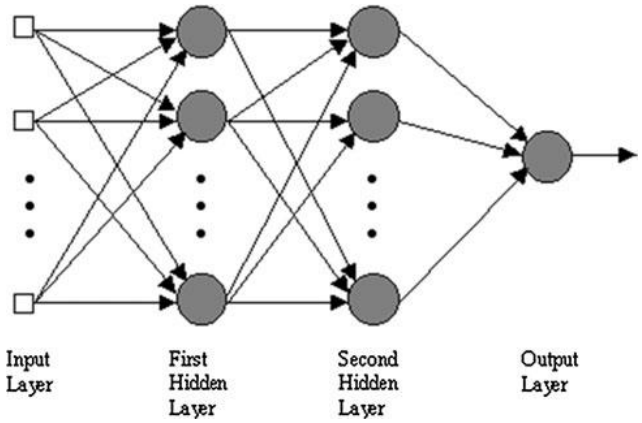

Fig.(2) The exemplary architecture of multilayer perceptron neural network [25].

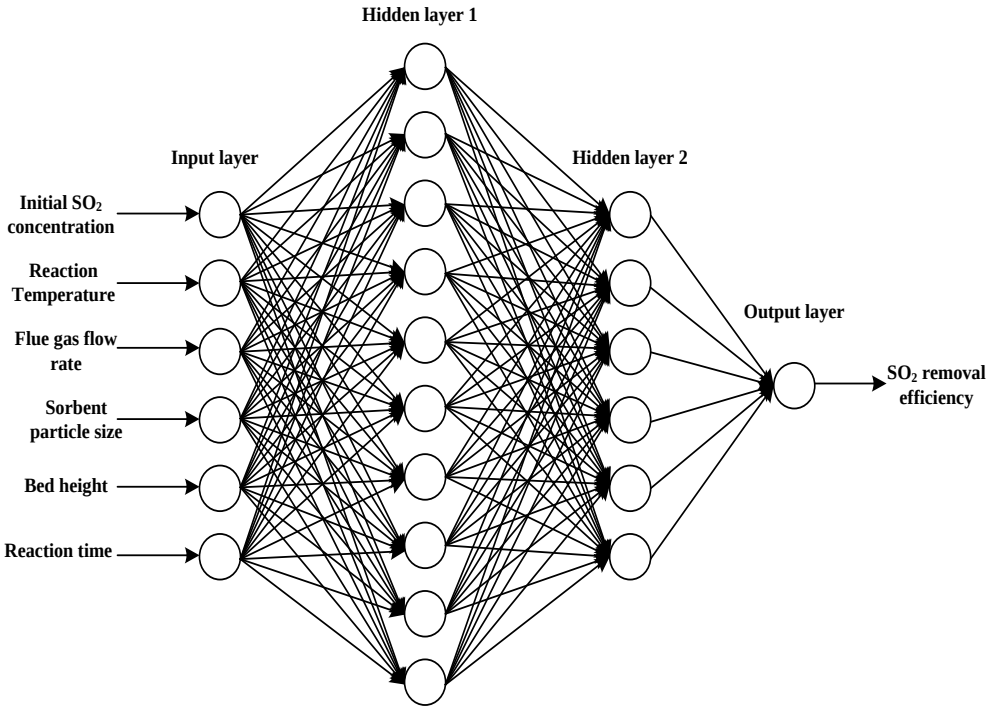

Fig.(3) The system used in the ANN model.

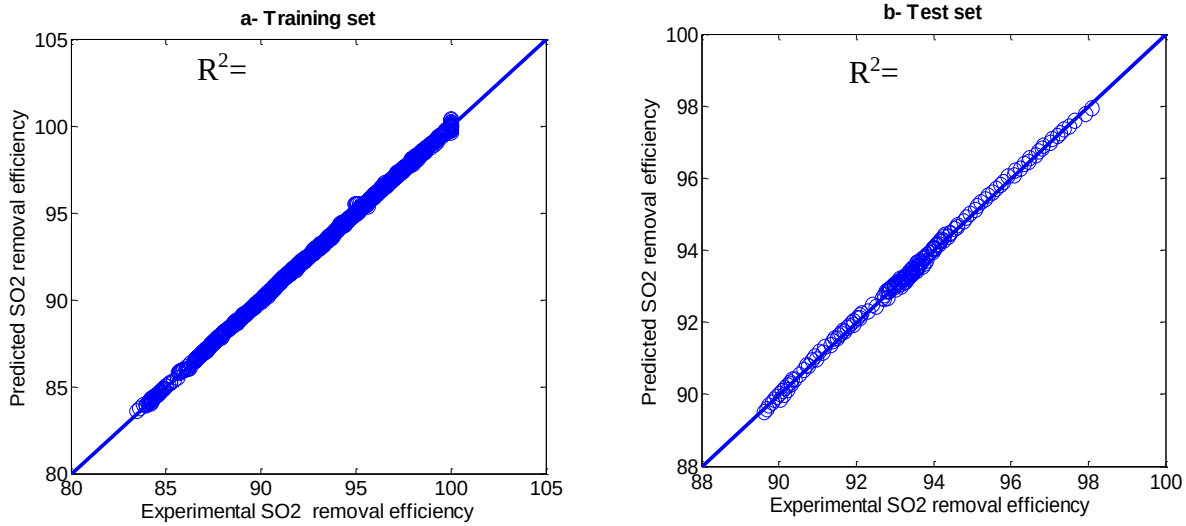

Fig.(4) Scatter plot showing experimental vs. neural network of the SO_2 removal efficiency.

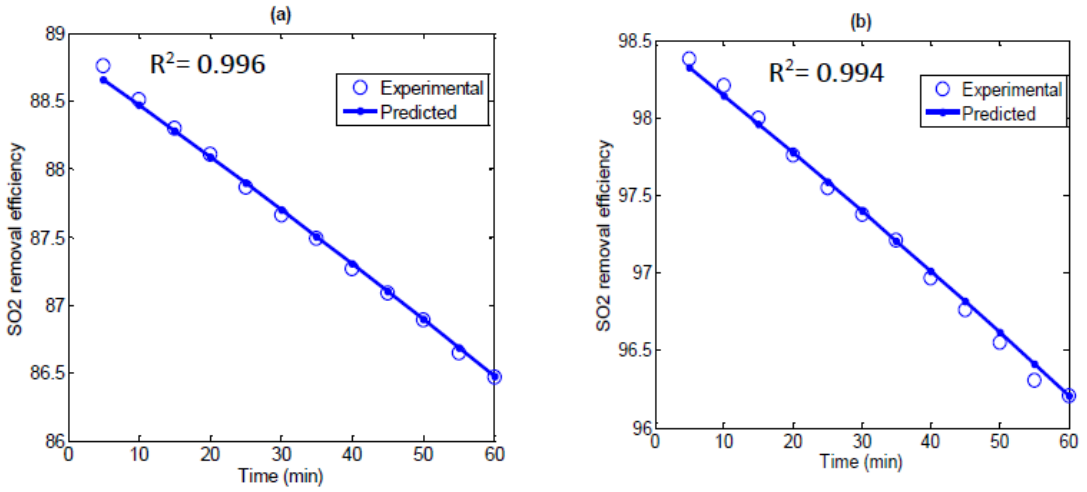

Fig.(5) (a,b) SO_2 removal predicted by the neural network vs. experimental of the SO_2 removal efficiency for interpolation data.

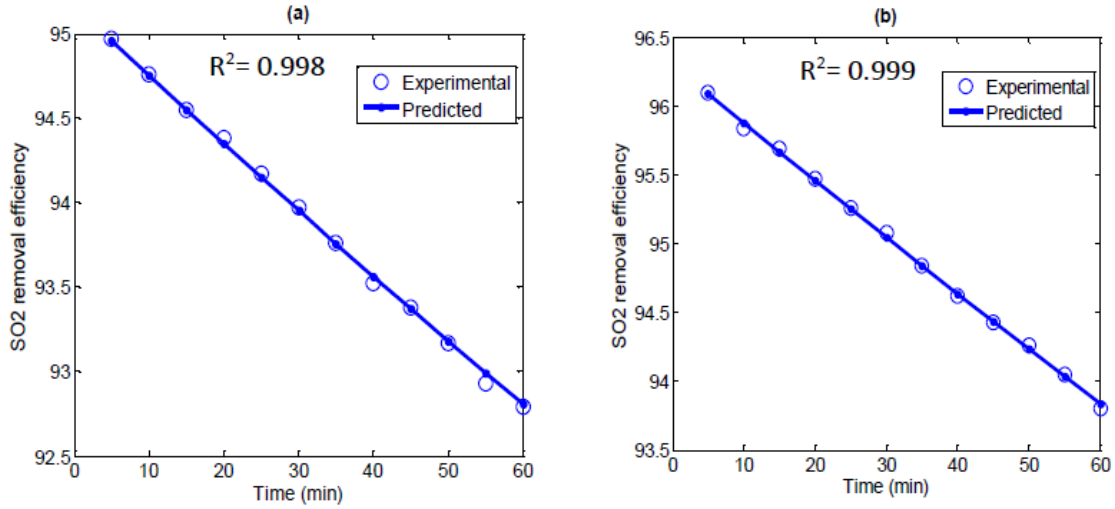

Fig.(6) (a,b) SO_2 removal predicted by the neural network vs. experimental of the SO_2 removal efficiency for extrapolating data.

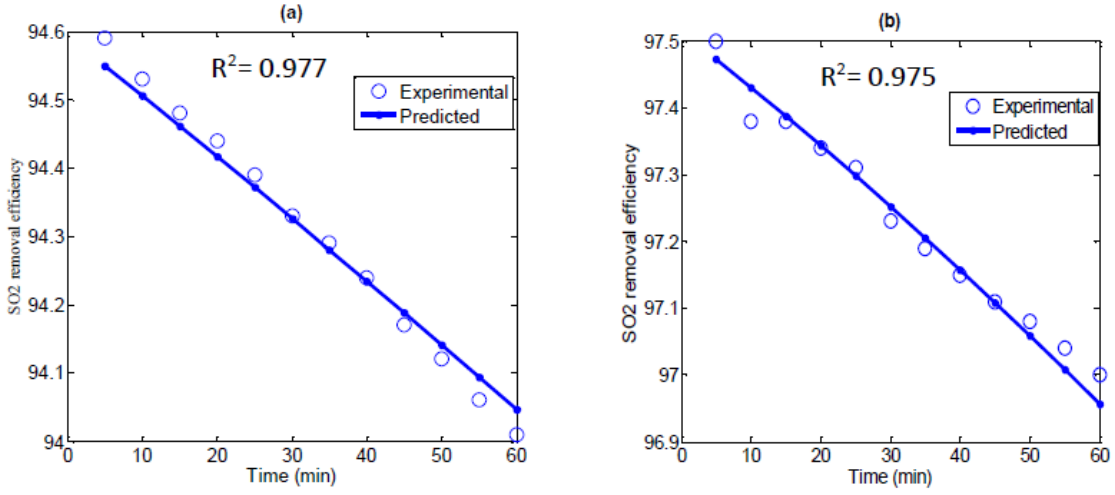

Fig.(7) (a,b) SO_2 removal predicted by the neural network vs. experimental of the SO_2 removal efficiency for extrapolating data.

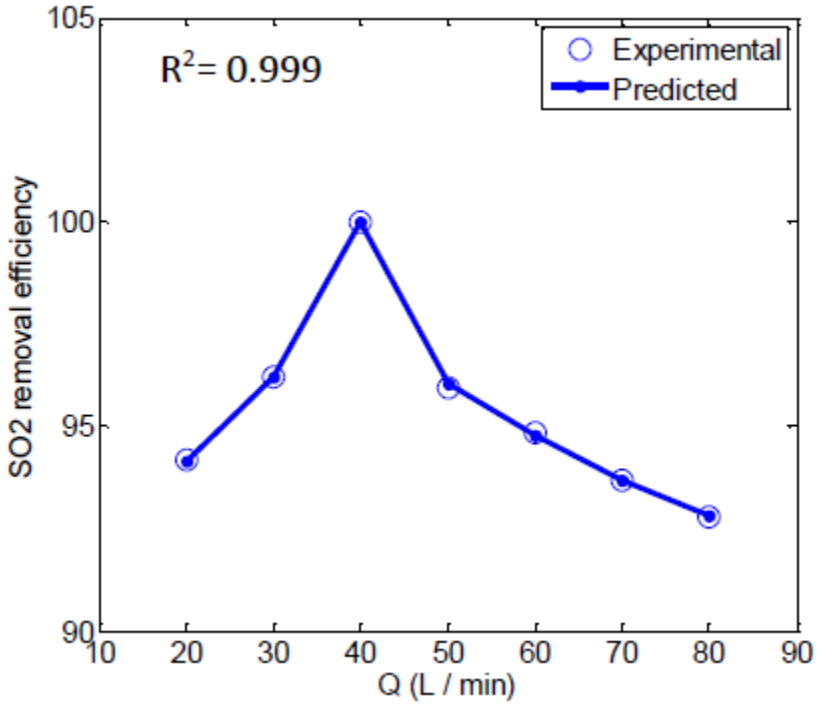

Fig.(8) The effect of gas flow rate on the SO_2 removal efficiency.

نمذجة الشبكات العصبية في ازالة ثاني اوكسيد الكبريت باستخدام الكربون المنشط كمادة مازة

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الخلاصة

في هذا البحث تم تطوير الموديل الرياضي باستخدام نظام الشبكات العصبية لتقييم طريقة ازالة ثنائي اوكسيد الكبريت من خليط غازي من (الهواء وثنائي اوكسيد الكبريت) باستخدام الكاربون المنشط كمادة مازة في مفاعل الطبقة الثابتة. النتائج العملية تم الحصول عليها بتاثير ستة عوامل رئيسية, وهي التركيز الاولي لثاني اوكسيد الكبريت و درجة حرارة التفاعل و ومعدل تدفق الغاز العادم وقطر الجزيئة الممتصة وارتفاع العمود وزمن التفاعل.

تم استخدام المعطيات العملية لتدريب واختبار موديل الشبكة العصبية وقد تم استخدام اسلوب التغذية المرتدة ذات الطبقتين المخفية لتدريب واختبار الشبكة العصبية. النتائج النظرية لازالة ثنائي اوكسيد الكبريت المستحصلة يتطابق مع المعطيات العملية حيث اعطى مقدار مربع الخطأ بنظام التدريب والاختبار بقيمة $0.112*10^{-4}$ و $0.817*10^{-3}$ على التوالي.

الكلمات الدالة: الشبكات العصبية الاصطناعية, الامتصاص , كفاءة الازالة.