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Neural network prediction of fluidized bed bioreactor performance for sulfide oxidation

Varsha Midha*,†, Mithilesh Kumar Jha*, and Apurba Dey**

*Department of Chemical Engineering, National Institute of Technology, Jalandhar 144 011, India
**Department of Biotechnology, National Institute of Technology, Durgapur, India
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Abstract−Sulfide oxidation rate of a fluidized bed bioreactor was predicted using ANN, with upflow velocity, hydraulic retention time, reactor operation time and pH given as input. The reactor was fed with 100 mg/L synthetic sulfide wastewater after biofilm formation on nylon support particles. Feedforward neural network model was prepared using 81 data sets, of which 63 were used for training and 18 for testing in a three-way cross validation. Prediction performance of the network was evaluated by calculating the percent error of each data set and mean square error for test data set in three partitions. The mean square error for test data set was 5.55, 4.08 and 2.30 for partition 1, partition 2 and partition 3, respectively. The predicted sulfide oxidation values correlated with the experimental values and a correlation coefficient of 0.96, 0.97 and 0.98 was obtained for partition 1, partition 2 and partition 3, respectively.

Key words: FBBR, Mean Square Error, Neural Network, Sulfide, Sulfur

INTRODUCTION

Sulfide is one of the most harmful compounds in the effluent generated by various industries like tanneries, petrochemical plants and anaerobic treatment of sulfate containing waste water. Sulfide is emitted into the environment in two forms: dissolved sulfide in the form of S^{2-} and HS^-, and as hydrogen sulfide (H_2S) in waste gases. The presence of dissolved sulfide in wastewater causes severe odor and corrosion problems. Even at low concentrations of dissolved sulfide, it is possible to generate a large amount of hydrogen sulfide gas, depending on the pH and temperature of the wastewater. Moreover, when oxygen becomes available, hydrogen sulfide is converted into sulfuric acid by sulfate oxidizing bacteria causing corrosion.

Sulfide can be removed from waste streams by a number of physicochemical methods but the high energy requirements or the high chemical and disposal costs besides environmental problems constitute important drawbacks of these methods. Biological processes, which break down the organic and inorganic compounds by the action of microorganisms, have been extensively applied for wastewater treatment due to their low cost, high efficiency, low energy requirements and minimal secondary waste generation. Some of the biological processes investigated so far involve the use of colorless sulfur bacteria that derive energy from the oxidation of reduced sulfur compounds, but they are rather sensitive to the concentration of sulfide and only survive if the sulfide concentration is low [1]. Usually, colorless bacteria completely oxidize the sulfide to sulfate, yielding more metabolically useful energy compared to the partial oxidation. To obtain S^o as a product, sulfide oxidation must proceed in the direction of sulfur production by using high sulfide loading rate or low oxygen concentrations. The following (biological) overall reactions occur in an aerobic sulfide removal system [2]:

$$2HS^-+4O_2 \rightarrow 2SO_4^{2-}+2H^+ \qquad \Delta G^o=-732.58\ \text{kJ/mol} \tag{1}$$

$$2HS^-+O_2 \rightarrow 2S^0+2OH^- \qquad \Delta G^o=-169.35\ \text{kJ/mol} \tag{2}$$

The formation of sulfur as the end product is preferred over sulfate formation for a variety of reasons. The insoluble sulfur can be removed from the treated effluent, leading to reduction of the total sulfur content of the wastewater. The recovered sulfur is a valuable compound which can be reused for the production of sulfuric acid or in the bioleaching process [3].

Phototrophic sulfur bacteria have been proposed as an alternative method for the treatment of sulfide containing effluents which avoid the requirement of constant oxygen supply. The major disadvantages in the use of photosynthetic bacteria on field scale lie in their anaerobic nature and the requirement of radiant energy, and hence extremely large, transparent surface area.

Surface attached accumulations of microbial cells encased in extracellular polymeric substances (known as biofilms) can be used in various types of reactors such as continuous stirred tank reactors (CSTR), packed bed reactors (PBR), fluidized bed reactors (FBR), airlift reactors (ALR) and expanded granular sludge bed (EGSB) reactors etc. Dissolved sulfide can be treated in biofilm reactor using pure or mixed culture to form sulfur, the immediate metabolic product in the biological oxidation process. Many materials have been successfully used as support particles for biomass immobilization, including sand, stone, glass and activated carbon. Use of high density support materials (sand, activated carbon etc.) requires high flow rates in order to achieve bed fluidization. These flow rates produce a "shearing" effect, leading to biofilm detachment, which is a basic process required for proper operation once the system is stabilized. However, when this process takes place during the development phase it becomes undesirable since the biofilm stabilization is delayed. On the contrary, when trying to prevent biofilm detachment by reducing flow rates, the biomass loss process is usually reversed, to the extent of turning the reactor performance into a fixed bed regime and, therefore, undergoing a bed clogging behavior in a shorter period of time [4-6]. Lertpocasombut et al. suggested the use of lower density support particles to avoid entrainment of particles from the reactor

†To whom correspondence should be addressed.
E-mail: midhav@rediffmail.com

[7]. Synthetic polymers have been successfully used as support media [8-12]. Polymer particles present a large surface area for microbial colonization and densities close to biofilm density, which assures homogeneous particle distribution in the fluidized-bed reactor. Several researchers studied the performance of bioreactor for sulfide oxidation [1-12] but none of them tried to establish a model for estimating the performance of fluidized bed bioreactor for sulfide oxidation.

It is very difficult to estimate the performance of such a bioprocess, as the performance depends on several factors, such as wastewater composition, operational parameters of the reactor, and the microbial community. The complex physical, biological and chemical processes involved in wastewater can be better controlled by developing a robust model for predicting the sulfide oxidation (%) from given inputs [13,14]. The operational parameters can be carefully selected so that an acceptable level of sulfide in the effluent is achieved after treatment.

Intelligent control systems, such as neural networks and fuzzy logic, have proved to be able to model nonlinear systems. Neural networks map a set of input patterns onto a corresponding set of output patterns after learning a series of past process data from a given system. Moreover, a neural network model has distinctive ability of learning non-linear functional relationships without requirement of the structural knowledge of the process to be modeled. In various studies [15-20], neural network models have been effectively applied to various chemical process controls, including wastewater treatment. Rangasamy et al. developed a multilayer perceptron neural network model for starch wastewater treatment in anaerobic tapered fluidized bed reactor. Vinod et al. used feedforward neural network to simulate the biodegradation process in a fluidized bed bioreactor. Wang et al. applied ANN to predict the steady state performance of an extended granular sludge bed bioreactor for nitrite denitrification and the complete DSR process. Delnavaz et al. observed that ANN based models provided an efficient and a robust tool in predicting MBBR performance for treating aromatic amine compounds [21]. There are several other similar applications of ANN in the field of environmental engineering, but none of them predict the sulfide oxidation in a bioreactor. ANN models have been reported to be better in their prediction performance as compared to statistical models [22].

In this paper, a neural network model is developed to predict the sulfide oxidation in a fluidized bed bioreactor with nylon support particles, using four input parameters:. upflow velocity, hydraulic retention time, reactor operation time and effluent pH.

MATERIALS AND METHODS

1. Fluidized Bed Bioreactor

The fluidized bed bioreactor was fabricated with glass, having 0.38 m length and 0.045 m diameter (Fig. 1). The total volume of the reactor is 0.6 L. The reactor is equipped with feed tank, peristaltic pump, recirculation pump and an aeration tank. A separate 2 L bottle was used as aeration tank to avoid turbulence in the reactor. The reactor has three uniformly located sampling ports used to extract liquid and bioparticles from the reactor. The reactor was filled with nylon supporting material to give 16 cm initial bed height from the supporting mesh. The working volume of the reactor was 0.25 L with packed bed height of 16 cm. The reactor was inoculated with

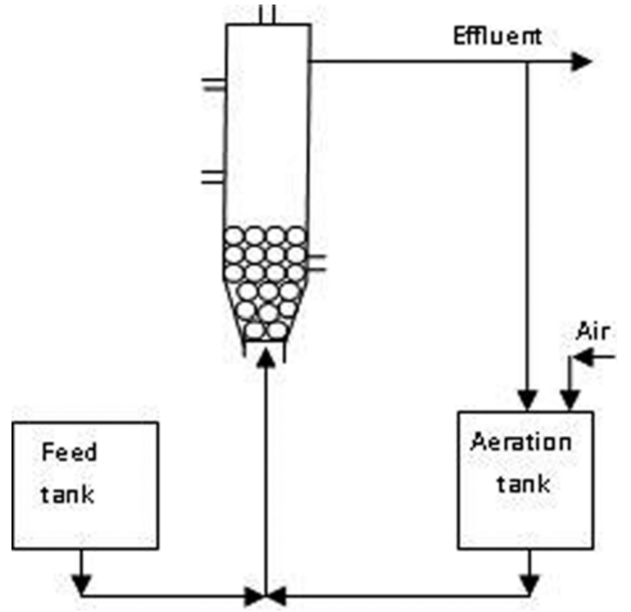

Fig. 1. Schematic view of fluidized bed bioreactor.

inoculum, having initial count of 4×10^7 CFU/mL to start growth of microorganisms on support particles. A heterogeneous population of microorganisms was obtained from the effluent treatment plant of a tannery. Inoculum was prepared by cultivating the mixed culture of microorganisms in shake flasks using growth medium with glucose 10 g/L, yeast extract 0.34 g/L, ammonium chloride 0.84 g/L, potassium dihydrogen phosphate 0.134 g/L, dipotassium hydrogen phosphate 0.234 g/L and magnesium chloride 0.084 g/L [23]. 200 mL of the inoculum was added to 2,000 mL of growing medium in the fluidized bed reactor. 10% inoculum prepared over the last 24 hr was added into new culture medium every day to keep sufficient nutrients for growth of microorganisms. After the completion of biofilm formation (42±3 μm), the synthetic wastewater containing sulfide concentration of 100 mg/L was fed from the bottom of the reactor with a peristaltic pump. The treated effluent coming from the reactor was passed through the aeration tank and was partly recirculated back to the reactor using another peristaltic pump. The air was continuously supplied to the aeration tank so that the dissolved oxygen was maintained between 2-5 mg/L in the aeration tank. The temperature was maintained at 30 °C. The dissolved oxy-

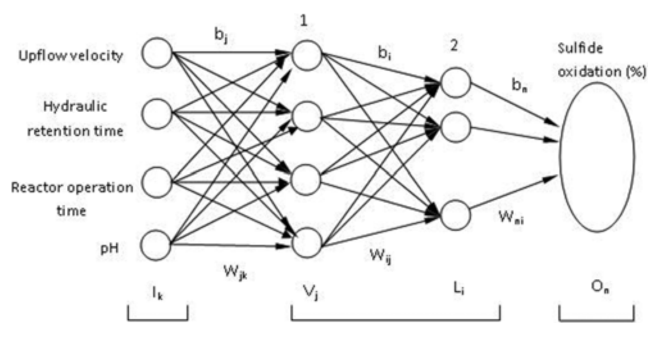

Fig. 2. Architecture of neural network used in the study: W_{jk} is the weight matrix, b_j is the bias, and V_j and L_i are the number of nodes in the hidden layers.

gen in the reactor was found to be less than 1 mg/L.

The samples were drawn at 24 hr and analyzed for pH, DO, temperature and sulfide. The experiments were carried out at different hydraulic retention times of 25 min, 50 min and 75 min and upflow velocity of 14 m/hr, 17 m/hr and 20 m/hr. All the analytical determinations were done according to APHA standard methods.

Inlet and outlet sulfide concentration was measured by iodometric titration method [24]. 2-4 mL of 0.025 N iodine solution was added to 10 mL sample, 3-5 drops of starch indicator and 2-4 drops of 6 N HCl were added in 250 mL conical flasks. This was then titrated against 0.025 N sodium thiosulfate solution (Na_2SO_3) until the sample turned colorless from typical blue color. Sulfide was calculated using the following equation:

$$\text{mg } S^{2-}/l = \frac{[(A\times B)-(C\times D)]\times 16000}{\text{ml sample}} \tag{3}$$

Table 1. Data set for neural network model

S. no.	Upflow velocity (m/hr)	Hydraulic retention time (min)	Reactor operation time (days)	pH	Sulfide oxidation (%)	S. no.	Upflow velocity (m/hr)	Hydraulic retention time (min)	Reactor operation time (days)	pH	Sulfide oxidation (%)
1	14	25	1	6.45	72	42	17	50	6	5.98	77
2	14	25	2	6.32	72	43	17	50	7	5.76	79
3	14	25	3	5.51	76	44	17	50	8	5.71	83
4	14	25	4	6.42	78	45	17	50	9	5.82	83
5	14	25	5	6.21	80	46	20	50	1	5.87	61
6	14	25	6	5.45	84	47	20	50	2	5.65	65
7	14	25	7	6.61	86	48	20	50	3	6.02	67
8	14	25	8	6.5	88	49	20	50	4	6.25	70
9	14	25	9	6.34	90	50	20	50	5	6.14	74
10	17	25	1	6.18	68	51	20	50	6	6.26	74
11	17	25	2	6.11	72	52	20	50	7	6.37	70
12	17	25	3	6.24	76	53	20	50	8	6.24	76
13	17	25	4	5.78	78	54	20	50	9	6.04	76
14	17	25	5	5.71	80	55	14	75	1	6.51	73
15	17	25	6	6.12	84	56	14	75	2	6.43	77
16	17	25	7	6.23	86	57	14	75	3	6.37	81
17	17	25	8	5.78	84	58	14	75	4	6.28	85
18	17	25	9	5.7	86	59	14	75	5	6.54	85
19	20	25	1	6.25	67	60	14	75	6	6.32	88
20	20	25	2	6.21	69	61	14	75	7	6.26	90
21	20	25	3	6.02	69	62	14	75	8	6.11	92
22	20	25	4	5.96	71	63	14	75	9	6.35	92
23	20	25	5	6.18	75	64	17	75	1	6.01	68
24	20	25	6	5.94	79	65	17	75	2	6.12	72
25	20	25	7	5.78	77	66	17	75	3	6.11	76
26	20	25	8	5.53	79	67	17	75	4	5.89	78
27	20	25	9	6.01	79	68	17	75	5	6.1	80
28	14	50	1	6.41	67	69	17	75	6	6.23	84
29	14	50	2	6.1	71	70	17	75	7	6.34	86
30	14	50	3	5.44	79	71	17	75	8	6.11	88
31	14	50	4	6.1	77	72	17	75	9	6.14	88
32	14	50	5	5.64	79	73	20	75	1	5.95	67
33	14	50	6	5.51	83	74	20	75	2	5.72	69
34	14	50	7	5.46	88	75	20	75	3	6.13	69
35	14	50	8	5.98	90	76	20	75	4	6.25	71
36	14	50	9	5.56	90	77	20	75	5	6.16	75
37	17	50	1	6.1	62	78	20	75	6	6.18	80
38	17	50	2	6.36	77	79	20	75	7	6.26	77
39	17	50	3	6.42	75	80	20	75	8	6.31	80
40	17	50	4	6.02	71	81	20	75	9	6.25	80
41	17	50	5	6.14	79						

Where, A-iodine solution (mL),
B-normality of iodine solution,
C-$Na_2S_2O_3$ solution (mL) and
D- normality of $Na_2S_2O_3$ solution

2. Artificial Neural Network Model

Artificial neural networks (ANN) are potent data-modeling tools that are able to capture and represent any kind of input-output relationship. This knowledge based system acquires, represents and uses knowledge for a specific purpose. The neuron is the processing element that takes a number of inputs, weights them, sums them up, adds a bias and uses the results as the argument for a singular valued function, which results in the neuron's output [14,15].

The problem of predicting the sulfide oxidation (%) during wastewater treatment from reactor process parameters can be viewed as one of function approximation. The sulfide oxidation is a function of the process parameters, and the goal is to approximate that function from a set of measurements. A feedforward neural network, which has been widely used for a variety of function approximation tasks, is used in the study. Fig. 2 shows the feedforward neural network using four nodes in the input layer (equal to the number of input parameters), two hidden layers and one node in the output layer (corresponding to the output, i.e., sulfide oxidation). Multilayer networks can be trained using various algorithms, but *back-propagation* is most widely used [15,25]. Back-propagation is a powerful, flexible training algorithm, though its training speed is low. It is an iterative gradient descent algorithm that minimizes the sum of squared error between the desired output and the actual output of a set of patterns. In addition, it is capable of handling noisy data [26,27].

To train a feedforward neural network properly, various network parameters have to be optimized, including the number of hidden layers, the number of neurons in the hidden layers, the learning rate and the number of training cycles (also known as epochs). Although some researchers suggest that one hidden layer is usually sufficient, the introduction of additional hidden layers allows the fit of a larger variety of target functions and enables approximations of complex functions with fewer connection weights [28,29]. In the present study, two hidden layers are used and different number of neurons in the hidden layers is tried to optimize the network. The starting point for number of neurons in the hidden layer was chosen by a thumb rule [30]:

$$n_{hidden} > 2\times[\max\text{ (input neurons, output neurons]} \quad (4)$$

Using too few neurons starves the network of the resources it needs to solve the problem. Using too many will increase the training time and may cause the problem called over-fitting. The error on the training set is driven to a very small value, but when new data is presented to the network, the error becomes very large. Although the network is able to map the training set, it cannot generalize to new situations. Hence, some of the test data points give very high errors. One method for improving network generalization is to use a network that is just large enough to provide an adequate fit. But it is difficult to know beforehand how large a network should be for a specific application. Network generalization can be improved by regularization or early stopping. In the present study, regularization is carried out to avoid over fitting, by modifying the performance function of mean square error, i.e., 'mse' to 'msereg'.

$$\text{msereg} = \gamma\,\text{mse} + (1-\gamma)\text{msw} \quad (5)$$

$$\text{msw} = \frac{1}{n}\sum_{j=1}^{n} W_j^2 \quad (6)$$

Where msw is mean square weights: γ is the performance ratio (default value 0.5). the value of 'msereg' becomes lower than 'mse'

Table 2. Experimental and predicted sulfide oxidation for test data set in three different partitions

Partition 1				Partition 2				Partition 3			
S. no.	Experimental	Predicted	Error (%)	S. no.	Experimental	Predicted	Error (%)	S. no.	Experimental	Predicted	Error (%)
1	72	70.44	2.17	2	72	73.28	−1.78	4	78	79.10	−1.41
5	80	78.24	2.20	6	84	82.25	2.08	9	90	90.37	−0.41
11	72	71.19	1.13	12	76	77.97	−2.59	10	68	70.72	−4.00
15	84	84.60	−0.71	16	86	84.89	1.29	14	80	80.65	−0.81
21	69	68.35	0.94	22	71	69.68	1.85	20	69	69.19	−0.27
25	77	80.88	−5.04	26	79	75.82	4.03	24	79	75.68	4.20
31	77	73.17	4.97	32	79	79.95	−1.20	30	79	78.91	0.12
35	90	89.64	0.40	36	90	92.59	−2.87	34	88	86.36	1.87
41	79	73.69	6.72	37	62	65.85	−6.22	40	71	70.39	0.86
45	83	85.80	−3.38	42	77	79.03	-2.63	44	83	81.88	1.35
46	61	61.84	−1.38	47	65	63.25	2.70	50	74	71.43	3.48
51	74	72.56	1.95	52	70	73.87	−5.53	54	76	76.63	−0.83
56	77	76.99	0.01	57	81	81.32	−0.39	55	73	72.69	0.42
61	90	90.45	−0.50	62	92	90.59	1.53	60	88	88.24	−0.27
66	76	77.82	−2.39	67	78	78.28	−0.36	65	72	69.50	3.48
71	88	89.23	−1.40	72	88	89.49	−1.69	70	86	85.36	0.75
76	71	66.66	6.11	73	67	68.32	−1.97	75	69	68.50	0.72
81	80	79.32	0.85	77	75	75.97	−1.30	80	80	78.36	2.05

hence the total error will be reduced.

3. Network Training and Cross Validation

A total of 81 data sets for sulfide oxidation (Table 1) were taken for the neural network model, of which 63 (about 78%) were used for training and 18 for testing the network. The prediction performance of the neural network was evaluated by testing with eighteen data sets which were not used during the training. To reduce the dependency of the results on a specific partition of the data into training and testing sets, a three-way cross validation test was performed; total data was divided into training and testing set in three different ways.

RESULTS AND DISCUSSION

Table 1 shows the sulfide oxidation values during the experiments at different levels of process parameters. During the experiments sulfate production was quite low in the range of 5 mg/L to 25 mg/L. No thiosulfate formation was observed during the experiments. The detailed values of sulfate, thisulfate and sulfur production during the experiments have been discussed and presented elsewhere [31]. The sulfide oxidation data was used to develop the neural network model. When the neural network was trained by training set, the sulfide oxidation was predicted for both training and testing sets. The percent error of each data set and the mean square error were calculated. Since mean square error (MSE) is the average of the square of the difference between the experimental and the predicted sulfide oxidation, this value can be used to estimate the prediction power of the ANN model. The predicted sulfide oxidation was correlated to the experimental sulfide oxidation, and the correlation coefficient between the experimental and predicted sulfide oxidation was obtained.

The input parameters--upflow velocity, hydraulic retention time, reactor operation time and effluent pH--are considered to predict sulfide oxidation in a fluidized bed bioreactor using nylon support particles. Table 2 shows the experimental and predicted sulfide oxidation in three different partitions.

1. Prediction Performance

Table 3 shows the prediction performance of the network for three ways cross validation data.

1-1. Partition 1

There are six neurons in the first hidden layer and four neurons in the second hidden layer. After 1028 iterations of training, the network converged to a minimum error level. The mean square error (MSE) in the training set is 0.26 and the coefficient of determination is 1. Maximum error is 2.53% and minimum error is 0.01%. In the test set, the MSE is 5.55 and the coefficient of determination is 0.92. The maximum and minimum errors are 6.11% and 0.01%, respectively.

1-2. Partition 2

There are five neurons in the first hidden layer and four in the second hidden layer. After 968 iterations of training, the network converged to a minimum error level. The MSE in the training set is 0.10 and the coefficient of determination is 1. Maximum error is 0.92% and minimum error is 0%. In the test set, the MSE is 4.08 and the coefficient of determination is 0.95. The maximum and minimum errors are 6.22% and 0.36%, respectively.

1-3. Partition 3

There are 13 neurons in the first hidden layer and six neurons in

Table 3. Performance parameters of the neural network model

	Partition 1	Partition 2	Partition 3
Network architecture	4-6-4-1	4-5-4-1	4-13-6-1
Performance goal	0.01	0.01	0.01
Epochs	1028	968	311
Performance ratio	0.95	0.95	0.95
TRAINING SET			
Mean square error (MSE)	0.26	0.10	0.39
Maximum error (%)	2.53	0.92	3.91
Minimum error (%)	0.01	0.00	0.01
Coefficient of determination (R^2)	1	1	1
TEST SET			
Mean square error (MSE)	5.55	4.08	2.30
Maximum error (%)	6.11	6.22	4.20
Minimum error (%)	0.01	0.36	0.12
Coefficient of determination (R^2)	0.92	0.95	0.97

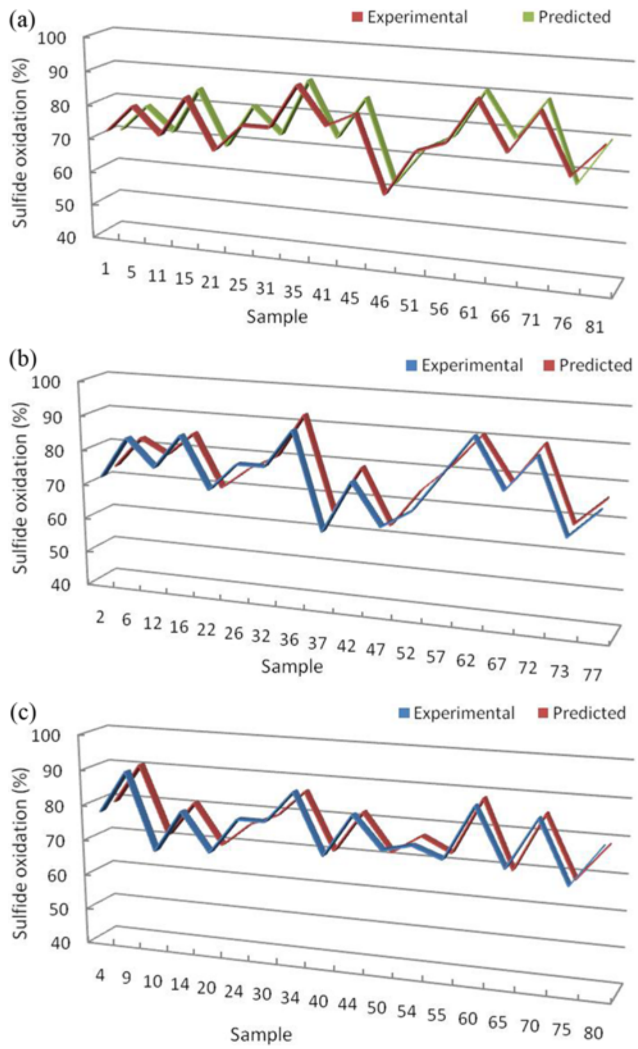

Fig. 3. Experimental and predicted sulfide oxidation (%) using nylon bioparticles: (a) Partition 1; (b) Partition 2; (c) Partition 3.

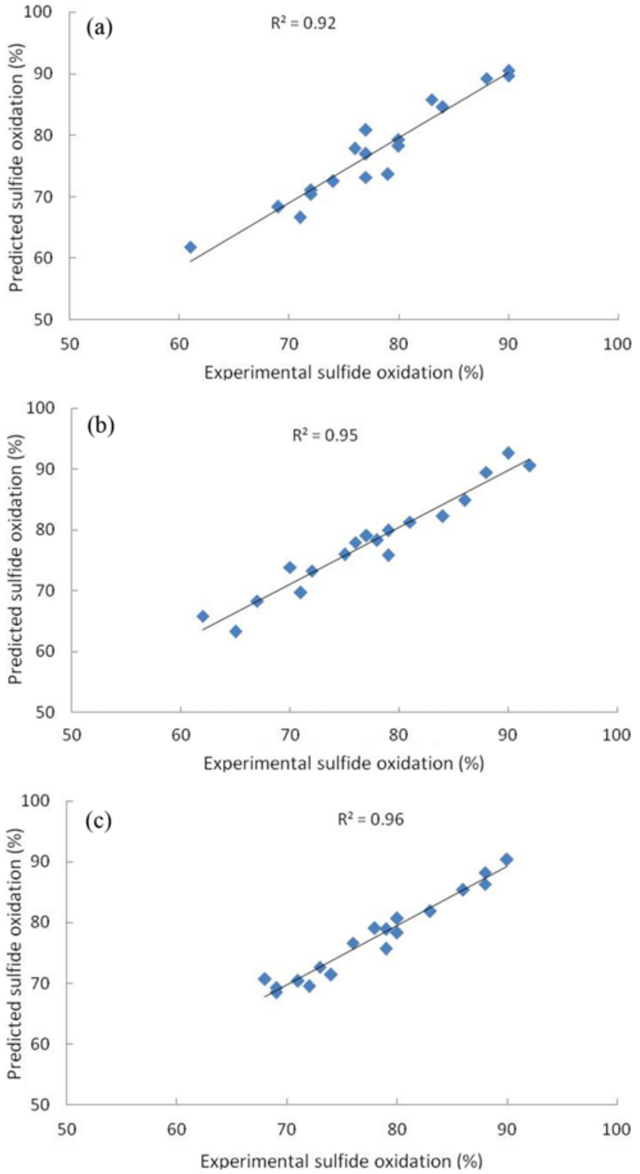

Fig. 4. Correlation between experimental values and predicted values of sulfide oxidation using nylon bioparticles for test sets using ANN: (a) Partition 1; (b) Partition 2; (c) Partition 3.

the second hidden layer. After 311 iterations of training, the network converged to a minimum error level, with mean square error (MSE) of 0.39 and the coefficient of determination of 1 in the training set. Maximum error is 3.91% and minimum error is 0.01% in the training set. In the test set, the MSE is 2.30 and the coefficient of determination is 0.97. The maximum and minimum errors are 4.20% and 0.12%, respectively.

Fig. 3 shows the experimental and predicted sulfide oxidation values for three way cross-validation tests. The proposed neural network approximates the sulfide oxidation values with a very low output error, regardless of the specific data partition into training and test sets. Fig. 4 shows the correlation between the experimental and predicted sulfide oxidation.

The values of correlation coefficient for three partitions are 0.96, 0.97 and 0.98, respectively. A good correlation between the experimental and predicted sulfide oxidation is obtained. The neural network system can be accurately used in the prediction of sulfide oxidation and optimizing the operational conditions.

According to the fluid mechanics laws of similarity, similar type of neural networks can be applied to reactors of the same design built at different scales or for different pollutants and their concentrations. Larger size reactors are expected to give higher removal efficiencies.

CONCLUSIONS

The feedforward neural network model predicted the performance of a sulfide oxidation fluidized bed bioreactor. The input parameters--upflow velocity, hydraulic retention time, reactor operation time and effluent pH--are considered to predict the sulfide oxidation in FBBR using nylon support particles. The developed model gave satisfactory results with minimum and maximum error of 0.01% and 6.22% among all the partitions. The correlation coefficient between the experimental and predicted sulfide oxidation values is 0.96, 0.97 and 0.98 for partition 1, partition 2 and partition 3 respectively. Hence, an ANN based model can be used to predict the fluidized bed bioreactor performance and control the operational parameters for improved process performance.

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