

Artificial Neural Networks for Modeling of Adsorption

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Abstract

Artificial Neural Networks are applied to the literature data pertaining to adsorption batch studies to develop and validate a model that can predict the Pollutant Removal Efficiency (PRE). Back-Propagation Network (BPN) is used with one hidden layer. The BPN model is systematically trained with 440 data points and is validated with 73 data points from the database. Three different combinations of learning parameters for number of neurons in hidden layer, learning rate, number of epochs, and error tolerance are attempted. Standard Deviation based on the test data is calculated to validate the accuracy of the output. The optimum values of learning parameters that are giving encouraging results for the present problem are identified using Elimination Technique.

1. Introduction

Adsorption is one of the established unit operations used for the treatment of contaminated water i.e., raw water and/or wastewater [1]. Adsorption studies are usually conducted over batch studies and column studies. The batch studies are aimed at determining the kinetics and isotherm constants while column studies are performed for determining the breakthrough curve. The Pollutant Removal Efficiency (PRE) is the most significant output of adsorption studies with respect to wastewater treatment. The variation of PRE depends on several factors such as adsorbent characteristics, contact time, adsorbate concentration etc. Babu and Ramakrishna [2] studied some of the above factors *vis-à-vis* the PRE and found that such attempts are leading to contradictory conclusions. If a same adsorbate (Pb^{+2}) is considered for example, the adsorbent with a surface area $5.36 \text{ m}^2/\text{g}$ recorded [3] a PRE of 72.72% - 90.12% whereas another adsorbent with a surface area of $168 \text{ m}^2/\text{g}$ recorded [4] a PRE of 78.4%-85%.

Babu and Ramakrishna [5] reported the following observations pertaining to the characteristics that are being reported in literature to that prescribed in the relevant IS codes (IS 2752:1995 & IS 8366: 1989):

- inconsistency in the characteristics of several adsorbents that are being reported
- insufficiency in the information to completely understand the adsorption mechanism when a database is generated for similar characteristics/trends in adsorption

This is evident from the above illustration showing comparison of PRE with respect to adsorbent characteristics. This illustration shows that the results that are reported by an investigator may be accurate and adequate for his study (i.e., Micro level) but when they are compared on a common basis (i.e., Macro level), with respect to the adsorbate/adsorbent/specific characteristic property etc., inconsistent trends in the results are observed. It is observed by the authors that it is extremely difficult to compare any indigenous data with that already published in literature, especially after observing the inconsistent and contrasting trends that are available in literature. This led to development of a model to predict the PRE from batch studies database of adsorption from literature assuming that the database reported in the literature is accurate at Micro level.

Babu and Ramakrishna [2] formulated the functional relation between the PRE and the above-mentioned factors. The factors considered are material of adsorbent, pollutant (adsorbate), pH, equilibrium and contact times, initial concentration of adsorbate, and adsorbent dosage. There are both dimensional variables (equilibrium and contact times, initial concentration of adsorbate, & adsorbent dosage) and dimensionless groups [material of adsorbent, pollutant, & pH] in these factors. The attempts made in proposing the empirical models both with dimensional variables and dimensionless groups could not fit the experimental data well, which may be due to the complexity of non-linear relationship and incomplete understanding between the input and output variables. Artificial Neural Networks (ANNs) have the ability to relate the input and output variables without having any knowledge on physics of the system provided an accurate and large amount of data on the system variables to train the networks is available. The neural networks can yield solutions to complex phenomena where the relationships and rules are not known [6, 7, 8]. The Back-Propagation Network (BPN) is one of several networks that is widely used for predicting the output and is successfully applied to a wide range of problems [9-19].

Regression technique is widely used for any Physical Modeling purposes. Babu and Ramakrishna [20] proved using a case study that Regression technique fails to represent the mechanism of adsorption. They concluded that the ANN model is giving better results for predicting results (output) from adsorption database. Babu and Ramakrishna [21] studied extensively on the Modeling of Adsorption Isotherm constants. They also observed

the inconsistent data that is being reported in literature pertaining to adsorption studies. They analyzed a compiled database from literature pertaining to adsorption isotherm constants using Regression analysis and Neural Networks. They observed that a very wide range of values for adsorption isotherm constants is being reported in literature. They concluded that the conventional analysis using Regression technique is not suitable for adsorption experimental data since the technique fails to understand the physics of the system. The BPN model attempted in that study to predict the adsorption isotherm constants gave relatively better results compared to that of Regression technique.

The ANN is hence applied to predict the PRE in adsorption studies in the present study keeping in view of the following aspects:

- Reported contradictory trends for PRE *vis-à-vis* adsorbent characteristics
- Discouraging results obtained for the Regression models in predicting the PRE in adsorption studies.

The prediction of PRE using ANN is studied for a large amount of data consisting as many as eight different pollutants and twenty-five different adsorbents [3, 4, 22-35] The database thus comprised of a heterogeneous data, which is systematically trained and tested using a BPN of ANN. The results are presented in this paper.

2. Problem Formulation And Data Generation

For batch studies of adsorption, the PRE (η) is considered a function of

$$\eta = f(\text{material, pollutant, pH, } t_{eq}, t_c, C_d, C_o)$$

where,

t_{eq} = equilibrium time of adsorption, T

t_c = contact time of adsorption, T

C_d = adsorbent dosage, ML^{-3}

C_o = initial concentration of adsorbate, ML^{-3}

A database (refer Annexure) of 513 data points comprising of the above variables is collected and compiled. The seven variables listed in the right-hand side of above equation are considered as input variables while the PRE is considered as the output variable. Thus, the ANN is having seven input variables and one output variable. The database is normalized to suit the input requirements of BPN of ANN using the formula:

$$Y_{norm} = \frac{Y - Y_{min}}{Y_{max} - Y_{min}}$$

where, Y_{norm} is the normalized value
 Y is the original value
 Y_{max} is the maximum value
 Y_{min} is the minimum value

This normalized data is used for training the network such that, the data will lie in the range of 0 to 1.0. Separate numbers are assigned to all eight pollutants for which the data pertaining to pollutant removal efficiency is available. This facilitates in normalizing the data available for the pollutants. The twenty-five materials attempted for the removal of above eight pollutants mentioned above are also assigned separate numbers for facilitating the normalization of the data for the materials. The objective here is to include as many different materials as possible that are employed for the removal of one or more of the eight pollutants under study. The range of pH for all the pollutants is taken as 0 to 14. The software code is written in C++. From the normalized database, the ANN is trained with 440 data points and tested (or validated) with 73 data points. The variables adopted for the learning sequence are:

- Error tolerance
- Learning rate
- Number of epochs
- Number of hidden layers
- Number of neurons in the hidden layer

For the present problem, there are seven inputs (as described earlier) and one output (PRE). The BPN is systematically trained and tested using three combinations of learning parameters and one hidden layer (Refer Table-1). A systematic approach is adopted in the present study to identify the optimum learning parameters for the Network using *Elimination Technique* as per the following priority:

- Number of neurons in the Hidden layer
- Learning rate
- Number of epochs

Table-1: Combinations of learning parameters studied

Learning parameter	Combinations		
	I	II	III
Number of epochs	10000, 15000, 20000, 40000	10000, 15000, 20000, 40000	10000, 15000, 20000, 40000
Number of neurons in hidden layer	5, 7, 9, 10, 12, 14, 15, 20, 25	5	5
Learning rate	0.3, 0.5, 0.7	0.3, 0.5, 0.7	0.3, 0.5, 0.7
Error tolerance	0.01	0.001	0.1

The results obtained from each of the steps adopted are carried forward in the subsequent analysis.

3. Results And Discussion

The percentage errors in the results are calculated using the formula:

$$Error(\%) = \frac{Testdata - Obtaineddata}{Testdata} \times 100$$

The Standard Deviation (SD) is calculated using the following formula [36]:

$$SD = \sqrt{\frac{\sum_{i=1}^n \left[\frac{y_{expected} - y_{calculated}}{y_{expected}} \right]^2}{n-1}}$$

The tests are performed using the combinations given in Table-1. The learning rate is varied as 0.3, 0.5, & 0.7 for all the above combinations. The other learning parameters are retained. The results are discussed in detail as follows:

3.1 Identification of Optimum number of neurons in the Hidden layer

The variation of Standard Deviation (SD) calculated using the above formula is compared with respect to number of neurons in hidden layer *vis-à-vis* each of the three learning rates. It is noticed that,

- As the learning rate is increased, the SD value is becoming lesser. In other words, the learning and prediction of the ANN is effective.
- Among the number of neurons attempted in the hidden layer, the SD value for 14 neurons is consistent (range: 2.5-3.0) for three epochs viz., 10000, 15000, & 20000. The SD value is even low (1.58) for 40000 epochs for the 14 neurons.

If the number of neurons in the hidden layer is more, the network becomes complicated. Results probably indicate that, the present problem (predicting the PRE of adsorbents) is not too complex to have a complicated network routing. Hence, the results can be satisfactorily achieved by keeping the number of neurons in the hidden layer at an optimum value of 14.

3.2 Identification of Optimum value of Learning rate

The combinations-II & -III are attempted in order to study the effect of Error Tolerance (ET) on the Learning rate. For this purpose, the number of neurons in the hidden layer are kept constant at 5 while the variation of the SD value is studied for each of the three learning rates viz., 0.3, 0.5, & 0.7 respectively. The accurate prediction of the output depends on ET. In general, the lesser the ET, the more accurate is the prediction. Four different epochs are attempted for understanding the variation of SD with respect to ET. The number of times the SD value reduced for a specific learning rate, for an ET from 0.01 to 0.001 is recorded. Inferences drawn from this observation are as follows:

- For a learning rate of 0.7, the SD value is reduced for an ET from 0.01 to 0.001 on one-out-of-four occasions (Epochs: 20000) and is found to be almost constant on two occasions (Epochs: 10000 & 40000).
- For a learning rate of 0.5, the SD value is reduced for an error tolerance from 0.01 to 0.001 on one-out-of-four occasions (Epochs: 40000) and is found to be constant on one occasion (Epochs: 20000).

- For a learning rate of 0.3, higher values of the SD are observed for an ET from 0.01 to 0.001 on three-out-of-four occasions (Epochs: 10000, 20000 & 40000).

Based on the results obtained for the reduction of SD value pertaining to a reduction in error tolerance from 0.01 to 0.001, the following conclusions are drawn:

- The learning rates of 0.3 & 0.5 can be ignored.
- The learning rate of 0.7 can be adopted.

Interestingly, similar observation is also drawn from Table-2. It may be noted that, three different learning rates viz., 0.3, 0.5, & 0.7 are considered in the present study. The learning rates of 0.3 and 0.7 are chosen such that they can represent the conditions above and below the learning rate of 0.5. Keeping the SD values obtained for the learning rate of 0.5 as *zero condition*, the percentage changes of the other two learning rates are calculated with respect to that of 0.5. A positive value indicates a high SD value compared to that obtained for a learning rate of 0.5. Similarly, a negative value indicates a low SD value compared to that obtained for a learning rate of 0.5. A large number of negative values (Refer Table-2) for the learning rate change from 0.5 to 0.7 indicate that the learning rate 0.7 is giving encouraging results in terms of low SD values compared to that obtained for a learning rate of 0.5. Similarly, a series of negative values ranging from -36% to -62.73% are observed for 14 neurons in the hidden layer for all the four epochs. Results given in Table-2 also confirm that a learning rate of 0.7 and 14 neurons are giving encouraging results. This inference confirms that there is a maximum decrease of SD value up to 62.73% at 40000 epochs. This shows that a higher learning rate given to the ANN in the present case is resulting in lesser SD values.

3.3 Identification of Optimum number of epochs

Epochs are usually increased in ANN to make the network repeatedly understand the trends of the data. There can be a point beyond which even ANN cannot understand the trends despite increasing the epochs. This may even result in higher error values. From the earlier discussions, it is observed that, a learning rate of 0.7 is giving encouraging results. Hence the analysis is focused exclusively for the results obtained for the learning rate of 0.7 to understand the variation of SD value with regard to the number of epochs and given in Fig. 1. In order to understand how many epochs each set of neurons is consuming before converging to a minimum SD value, 5 sets of neurons viz., 5, 7, 10, 14, & 15 are selected based on the results obtained and discussed earlier.

The following observations are made from Fig. 1:

- The SD values of 14 neurons follow a consistent range (2.67-2.74) for the first three sets of epochs (i.e., 10000, 15000, & 20000) while it is even lower (1.58) for 40000 epochs.

- The SD values of 5 neurons follow a consistent range (2.66-2.92) for all the four different epochs.
- The SD value of 14 neurons at 40000 epochs (1.58) is lower than that of 5 neurons (2.66) at the same set of epochs.
- The SD values for other neurons considered are relatively not encouraging.

Table-2: Percentage change of Standard Deviation for learning rates of 0.3 & 0.7 over 0.5 with respect to number of nodes in hidden layer

(a): NNHL vs. 10,000 & 15,000 epochs

NNHL	10,000 epochs		15,000 epochs	
	0.3	0.7	0.3	0.7
5	41.65	-2.81	54.33	5.06
7	4.81	-31.26	5.26	-8.17
9	55.73	8.22	136.51	47.46
10	-0.38	-35.51	-0.40	-31.50
12	2.95	-34.62	13.13	11.63
14	-4.0	-37.48	-0.65	-36.0
15	2.14	-3.87	-1.35	-37.31
20	-6.94	-7.30	8.45	7.02
25	114.36	33.53	2.63	2.74

NNHL: Number of Nodes in Hidden Layer

(b): NNHL vs. 20,000 & 40,000 epochs

NNHL	20,000 epochs		40,000 epochs	
	0.3	0.7	0.3	0.7
5	42.62	-8.10	-0.57	-37.79
7	1.87	-35.87	1.58	-35.70
9	2.81	-34.11	-2.31	-35.03
10	-2.47	-36.76	-0.85	-3.46
12	4.49	-29.98	0.02	0.28
14	0.16	-37.18	0.58	-62.73
15	-5.20	-1.29	-0.94	2.52
20	2.28	-36.51	-2.52	-47.93
25	2.42	-38.33	99.86	100.31

NNHL: Number of Nodes in Hidden Layer

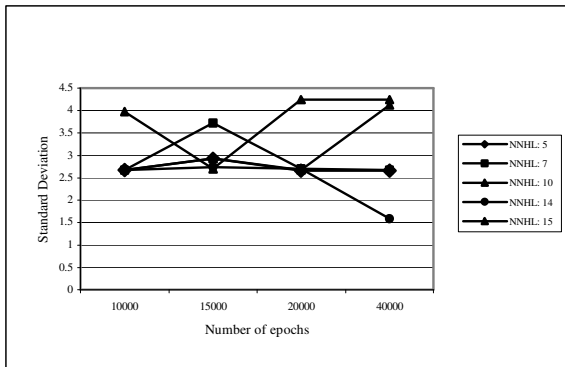


Fig. 1: Effect of number of epochs on Standard Deviation for number of neurons in hidden layer at a learning rate of 0.7

As is evident from the above results, the successful predictions are obtained by using 14 neurons in the hidden layer of the BPN for the present problem.

It can thus be concluded that, the following learning parameters are found to give optimum results for the present problem:

- Number of hidden layers: 1
- Error tolerance: 0.001
- Number of neurons in the hidden layer: 14
- Learning rate: 0.7
- Number of epochs: 40000

4. Summary And Conclusions

ANNs are attempted to predict the PRE by adsorption for a heterogeneous data collected from 25 different materials tested over eight different pollutants. Back-Propagation Network (BPN) is used with one hidden layer. Seven input parameters are identified and the network is systematically trained with 440 data points and tested with 73 data points. Three different combinations of learning parameters are attempted. Standard Deviation based on the test data is calculated to validate the accuracy of the output. Elimination Technique is used and the optimum values of the learning parameters for the present problem are obtained.

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Annexure

Details Of The Data Points Used For Training And Testing The Network

Data points used for training the network: 440

Data points used for testing the network: 73

Range of the operating parameters for each of the eight pollutants

Chromium			Mercury		
<i>Parameter</i>	<i>Minimum</i>	<i>Maximum</i>	<i>Parameter</i>	<i>Minimum</i>	<i>Maximum</i>
Equilibrium time, hours	1.5	24	Equilibrium time, hours	3	3
Contact time, min	30	1440	Contact time, min	30	180
Adsorbate concentration, mg/L	2	150	Adsorbate concentration, mg/L	50	500
Adsorbent dose, g/L	0.2	150	Adsorbent dose, g/L	20	20
Zinc			Cadmium		
<i>Parameter</i>	<i>Minimum</i>	<i>Maximum</i>	<i>Parameter</i>	<i>Minimum</i>	<i>Maximum</i>
Equilibrium time, hours	0.5	24	Equilibrium time, hours	0.5	24
Contact time, min	10	1440	Contact time, min	30	1440
Adsorbate concentration, mg/L	10	200	Adsorbate concentration, mg/L	20	70
Adsorbent dose, g/L	10	100	Adsorbent dose, g/L	0.1	10
Copper			Lead		
<i>Parameter</i>	<i>Minimum</i>	<i>Maximum</i>	<i>Parameter</i>	<i>Minimum</i>	<i>Maximum</i>
Equilibrium time, hours	0.5	24	Equilibrium time, hours	0.5	24
Contact time, min	20	1440	Contact time, min	10	1440
Adsorbate concentration, mg/L	10	100	Adsorbate concentration, mg/L	20	1000
Adsorbent dose, g/L	10	100	Adsorbent dose, g/L	0.1	40
Color			COD		
<i>Parameter</i>	<i>Minimum</i>	<i>Maximum</i>	<i>Parameter</i>	<i>Minimum</i>	<i>Maximum</i>
Equilibrium time, hours	3	4	Equilibrium time, hours	8	8
Contact time, min	30	240	Contact time, min	480	480
Adsorbate concentration, mg/L	89.1993	1000	Adsorbate concentration, mg/L	400	1500
Adsorbent dose, g/L	3.33	50	Adsorbent dose, g/L	20	30