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Neural Network Modeling and Sorption of As III with Zinc Oxide Nanoparticle Bounded on Activated Silica using *Ocimum Sanctum*

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Abstract

A three-layered Artificial Neural Network (ANN) model was more advanced to foretell the removal efficiency of As (III) ions from aqueous solution with Ocimum Sanctum. Batch experiments resulted into standardization of optimum conditions of adsorbent dosage (3 g), As (III) concentration (0.007N) volume (20 ml)at pH 6 and agitation speed of 250 rpm. A time of thirty minutes was found sufficient to achieve the equilibrium. The ANN model was premeditated to forecast adsorption efficiency of Zinc oxide nanoparticle ingrained on activated silica using Ocimum Sanctum (ZnO-NPs-AS-Os) by combining back propagation (BP) with prime component analysis using neuron (R2011a) solution. A tangent sigmoid axon was used as transfer function for input to hidden layer whereas a linear purelin function is used at output layer. The Levenberg–Marquardt Algorithm (LMA) was applied to give minimum Mean Squared Error (MSE) for training, testing and cross validation. Comparison between the model results and experimental data gives a high degree of correlation (R2 = 0.9846) indicating that the model is able to predict the sorption efficiency with reasonable accuracy.

Keywords: 1. Neurons 2.Back Propagation 3.Network Architecture 4.Kinetics.

Introduction

Arsenic is naturally present at high levels in the groundwater of a number of countries. Arsenic is highly toxic in its inorganic form. Contaminated water used for drinking, food preparation and irrigation of food crops poses the greatest threat to public health from arsenic. Long-term exposure to arsenic from drinking-water and food can cause cancer and skin lesions. It has also been associated with developmental effects, cardiovascular disease, neurotoxicity and diabetes. The most important action in affected communities is the prevention of further exposure to arsenic by provision of a safe water supply. Arsenic is a natural component of the earth's crust and is widely distributed throughout the environment in the air, water and land. It is highly toxic in its inorganic form. People are exposed to elevated levels of inorganic arsenic through drinking contaminated water, using contaminated water in food preparation and irrigation of food crops, industrial processes, eating contaminated food and smoking tobacco. Long-term exposure to inorganic arsenic mainly through drinking of contaminated water, eating of food prepared with this water and eating food irrigated with arsenic-rich water can lead to chronic arsenic poisoning [1]. Skin lesions and skin cancer are the most characteristic effects. The greatest threat to public health from arsenic originates from contaminated groundwater. Inorganic arsenic is naturally present at high levels in the groundwater of a number of countries, including Argentina, Bangladesh, Chile, China, India, Mexico and the United States of America. Drinking-water crops irrigated with contaminated water and food prepared with contaminated water are the sources of exposure. People who smoke tobacco can also be exposed to the natural inorganic arsenic content of tobacco because tobacco plants essentially take up arsenic naturally present in the soil. Also in the past the potential for elevated arsenic exposure was much greater when tobacco plants used to be treated with lead arsenate insecticide [2]. The immediate symptoms of acute arsenic poisoning include vomiting, abdominal pain and diarrhea. These are followed by numbness and tingling of the extremities, muscle cramping and death in extreme cases. The first symptoms of long-term exposure to high levels of inorganic arsenic (e.g. through drinking-water

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and food) are usually observed in the skin and include pigmentation changes, skin lesions and hard patches on the palms and soles of the feet (hyperkeratosis). These occur after a minimum exposure of approximately five years and may be a precursor to skin cancer. In addition to skin cancer long-term exposure to arsenic may also cause cancers of the bladder and lungs. The International Agency for Research on Cancer (IARC) has classified arsenic and arsenic compounds as carcinogenic to humans and has also stated that arsenic in drinking-water is carcinogenic to humans. Other adverse health effects that may be associated with long-term ingestion of inorganic arsenic include developmental effects, neurotoxicity, diabetes and cardiovascular disease. The most important action in affected communities is the prevention of further exposure to arsenic by the provision of a safe water supply for drinking, food preparation and irrigation of food crops [3]. There are a number of options to reduce levels of arsenic in drinking-water. The present study is aimed at selection of a low cost adsorbent which can adsorb arsenic from aqueous solution. Detailed batch studies with the selected adsorbent, *Azadirachta indica, Corriandrum sativum, Acalypha indica, Emblica officinalis* have been carried out in the earlier investigation. The tribulations of our bionetwork are increasing with the encroachment in technology. Techniques used for deduction of heavy metals like lime coagulation, reverse osmosis, chemical precipitation, ion exchange and solvent extraction are expensive and non-environmental friendly as compared to adsorption. Adsorption is one of the easiest safest and most expenditure effective methods for the removal of metals. The foremost advantage of an adsorption system of silica embedded zinc oxide nanoparticle are less investment in terms of both initial cost and simple designed easy operation and has no effect of toxic substance compared to conventional chemical treatment process. There is a vital requirement for development of innovative but low cost processes by which heavy metals can be removed. Adsorption technique is quite trendy due its simplicity and high efficiency, as well as the ease of use of a wide range of adsorbents [4].Increasing awareness towards green chemistry and biological processes has led to the efficacy and feasibility of an eco-friendly approach for the synthesis of ZnO nanoparticle entrenched on activated silica as proficient adsorbent for removal of As (III) using Artificial Neural Network.

Materials and Methods

Adsorbent preparation and Characterisation

Aqueous leaf extract of *Ocimum Sanctum* was stirred for 30 min to that 1g of Zinc acetate dihydrate was added under vigorous stirring. After 1hr stirring 10 g of activated silica was introduced into the above solution followed by the addition of aqueous NaOH resulted in a white aqueous solution at pH 12. This was then sited in a magnetic stirrer for 2hr. The activated silica supported ZnO nanoparticle were then filtered and washed with double distilled water. The synthesized ZnO-NPs-AS-*Os* was maintained at 60ºC for 12 hrs. ZnO-NPs-AS-*OS* structure was primed by green synthesis method. A mortar was used to homogeneously ground ZnO-NPs-*Os*. The proposed sorbent were stored in air at room temperature. The X-Ray powder diffraction pattern of the as- synthesized sample was recorded on an X-ray diffractometer (XRD, PW 3040/60 Philips X'Pert) using Cu (Kα) radiation (λ =1.5416 A˚) operating at 40 kv and 30 mA with 2θ ranging from 10- 90˚.The external morphology of the sample were characterized by scanning electron microscope (SEM) (LEO 1530FEGSEM).

Biosorption studies

Sorption studies using standard practices were carried out in batch experiments (duplicate) as a function of adsorbent dosage (0.5-5.5g), contact time (10-80 min), volume of the test solution (20-250 ml), metal concentration (0.005-0.1N), with pH (1-8) and agitation speed of (50-350rpm). The details of methodology have been described in our earlier publications [2]. Percent metal uptake by the adsorbent has been computed using the following equation % Sorption = Co – Ce x 100 / Co, where Co and Ce were the initial and final concentration of metal ions in the solution.

Definition of the ANN model

ANN utilizes interconnected mathematical neurons to form a network that can model complex functional relationship [5]. In recent years ANN have been used as a powerful modeling tool in various processes such as membrane filtration, gas separation, ultra filtration, reverse osmosis etc. [6-7]. In continuation of our work on modeling of As^{3+} adsorption from aqueous solution using *Ocimum sanctum* [8], the present paper describes the abatement of As (III) ions from aqueous system using(ZnO-NPs-AS-*Os*) .Pursuing benchmark

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comparisons of BP algorithms, a study was conducted to determine the optimization study to determine the optimal network structure .Experimental data were initially distributed to three subsets as training, validation and testing. Finally, output obtained from the ANN modeling was compared with the experimental data. The present work highlights the prospect for the metal ions from aqueous solution in the range of metal concentration with which lab experiments indicates that the model is able to predict the sorption efficiency with sensible accuracy. Neuro Solution 7(R2011a) mathematical software was used to predict the sorption efficiency. Ninety five experimental sets were used to develop the ANN model. A three-layered ANN with sigmoid axon transfer function and linear purelin was used for input hidden and output layers. The data gathered from batch experiments were divided into five input matrix. The three layered sigmoid and purelin network represents functional relationship between inputs and output, provided sigmoid and purelin layers has enough neurons. Levenberg-Marquardt algorithm is fastest training algorithm for network of moderate size used in the present. Feed-Forward Back Propagation (FFBP) neural networks were utilized for training of experimental data set in some application works related to drying technology [9-11]. A typical FFBP neural network consists of the following layers: input layer, hidden layer and output layer as illustrated in Figure 1. For studying by FF network, back propagation (BP) learning algorithm is normally used. In this BP algorithm, the first output layer weights were updated. A desired value (target) exists for each neuron showing as the output layer. During training the FFBP, calculations were conducted from input of network toward output and then error values were propagated to previous layers. The neurons from which the network is built are the data processing units. The ANN calculates the outgoing values on the basis of the information given at the incoming side of the network. Determining the number of hidden layers is not easy. There are arbitrary and less arbitrary methods by which to select them correctly. Typically the best results are obtained when these values are selected empirically. After determining the number of layers the weight and threshold values for all the neurons should be selected. They should be selected to provide the minimum error of network activity that gives the highest prediction. Most often the network error of the configuration of some weight and threshold parameters for the correct solution is assessed by comparing the output values with those given as examples (learning data). The basis for the automatic modification of weights and ranges allowing for minimal error for collected cases, i.e., input data with a solution fraction of the correct learning data that are typically historical cases. These data are involved in the network's learning a process which is equivalent to adjusting the parameters of the model in the discussed cases. Prediction operations are made on the basis of examples of input data of testing series and are performed by so called learning algorithms. In order to evaluate the assumed parameters of the neural networks model, the RMSE was determined using the following function:

$$\mathbf{RMSE} = \sqrt{\frac{1}{N}\sum_{1}^{N}(\mathbf{SP}-\mathbf{SR})^{2}} \quad (1)$$

$$\mathbf{R} = \frac{\sum_{i=1}^{N}(\mathbf{SPi}-\mathbf{SP})(\mathbf{SRi}-\mathbf{SR})}{\sqrt{\sum_{i=1}^{N}(\mathbf{SPi}-\mathbf{SP})\sum_{i=1}^{N}(\mathbf{SRi}-\mathbf{SR})}} \quad (2)$$

The second evaluation criterion was to define the correlation factor, *R*, between the projected and the observed values. The value of the linear correlation on the basis of the element sample was calculated from the formula.

Where *R is* the observed value and *P S* the predicted value of the solution. The correlation coefficient *R* expresses a linear dependence between the two variables. The closer the correlation coefficient is to 1, the stronger the linear dependence [12-13]. Usually, the following dependence of the correlation coefficient is adopted:

- R < 0.2 – no linear dependence,
- R = 0.2 – 0.4 – weak dependence,
- R = 0.4 -0.7 – moderate dependence,
- R = 0.7 – 0.9 – strong dependence,
- R > 0.9 – 1very strong dependence.

Results and Discussions

Characterization of ZnO-NPs-AS - Os

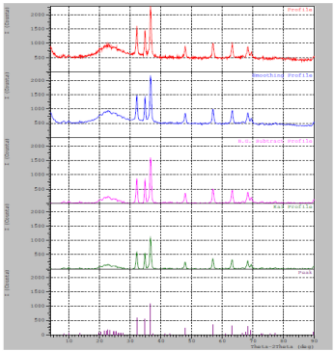

Figure.1A XRD Out line

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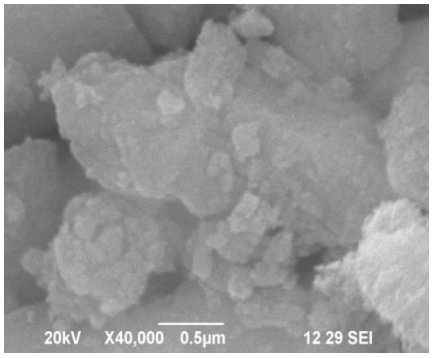

Figure.1B SEM Micrograph

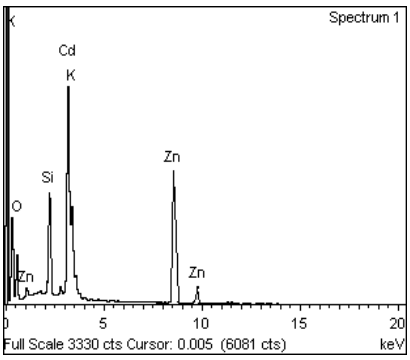

Figure.1C EDX Spectrum

Figure 1A shows the unique XRD outline of ZnO-NPs-AS prepared by the green synthesis method at 60˚C for 12 hours for aqueous leaf extract of *Ocimum Sanctum*. Corresponding emblematic peak primarily at about 12˚, 20˚ for silica and 32˚, 34˚, 36˚ for ZnO NPs are indicative of nano-crystalline nature of ZnO-NPs-AS-Os in combination. It can be seen that all of these peaks [14] are well matched with that of Zincite phase (JCPDS CARD NO: 36-1451).Figure 1B shows the SEM micrograph of homogeneous shape and size for ZnO-NPs-AS-*Os* at X40,000 obtained using aqueous leaf extract of *Ocimum Sanctum*. The size and morphology of the obtained ZnO-NPs-AS-*Os* is found to be 100 nm. The EDX spectrum (Figure 1C) shows the peak only for the presence of Zinc, Oxygen silicon elements in the as-prepared ZnO-NPs-AS -*Os*. The percentage of zinc, silica and oxygen is found to be 100 which proves that it is completely free from impurities.

Sorption Studies

Sorption studies led to the standardization of the optimum conditions as As(III) concentration (0.07N) ,contact time (30 min),sorbent dosage of (3g) and volume (250 ml) at pH(6) at an agitation speed (250 rpm) for maximum (98.08%)As (III) removal. The results indicate that ZnO-NPs-AS-*Os* has considerable potential to be used as sorbent for metal removal from waste water.

Effect of the System	Concentration (N)	Adsorption dosage (g)	Contact time (min)	pH	Agitation (rpm)	Percentage removal (%)
Concentration (N)	0.005, 0.0075, 0.01, 0.02, 0.03, 0.04, 0.05, 0.06,**0.07**, 0.08,0.09,0.10	3	30	6	250	98.08
Adsorption dosage (g)	0.07	0.5, 1, 1.5, 2, 2.5, **3**, 3.5, 4, 4.5,5,5.5	30	6	250	98.08
Contact time (min)	0.07	3	10, 20, **30**, 40, 50, 60, 70, 80	6	250	98.08
pH	0.07	3	30	1, 2, 3, 4, 5, **6**, 7,8	250	98.08
Agitation (rpm)	0.07	3	30	6	50, 100, 150, 200, **250**, 300, 350	98.08
Percentage removal (%)	0.07	3	30	6	250	92.38,95.30, 97.45,**98.08**, 96.02

Table.1 Sorption Studies at Optimum Conditions of As(III) Removal Data Statistics for NN Architecture

Studies, therefore, have been planned to predict the efficiency of ZnO-NPs-AS-*Os* for the removal of As (III) using ANN

Optimization of the ANN structure

The prediction and removal efficiency of As (III) ions from aqueous system using ZnO-NPs-AS-*Os* are made in the range of metal concentration with which experiments have not been conducted. A training set of ninety five experimental data sets was selected to develop the model. ANN model based on three layers recurrent Back Propagation Algorithm (BPA) for the experimental data was applied to train the neural network. During training, the output vector is computed by a forward pass in which the input variable (concentration, dosage, time, pH and agitation) is propagated forward through the network to compute the output (percentage removal) of each unit (Table.1). The output is then compared with the desired vector which resulted into error signal for each output unit [15]. To minimize the error appropriate adjustments were made for each of the weights of the network. After several such iterations, the network was trained to give the desired output for a given five input variable (Table.2). The best possible network structure was determined as three layer with 32 hidden neurons (Figure.2) and 13epochs (Figure.3) with unsurpassed validation performance and least mean squared error describing the dynamics of As (III) in aqueous solutions with output one. The available 95 data set are divided into 67 data set for training and 14 data set for testing and validation. When the training for 67 data set has been

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completed testing for 14 data has done to examine the performance of the derived network as testing is the primary approach for verifying that inputs produced the appropriate output as 1. The iteration continues until the error sum is converged.

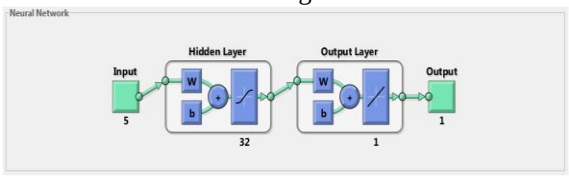

Figure.2 Optimum Three Layered LMFFBP Neural Network Architecture

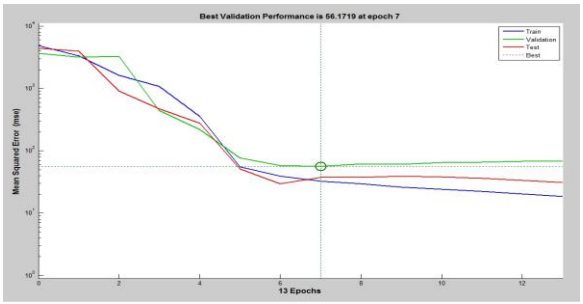

Figure.3 Best validation Performance with least MSE

In the field of machine learning a confusion matrix also known as a contingency table or an error matrix is a specific table layout (Figure.4) that allows visualization of the performance of an algorithm typically a supervised learning. Each column of the matrix represents the instances in a predicted class, while each row represents the instances in an actual class. This demonstrates how a confusion matrix can be used to assess the performance. All dominant diagonal elements represented in the green and blue cells (67,14,14) on the confusion matrix represents the classified data with 100 percentage performance while the off-diagonal (red cells) represents the misclassified data .

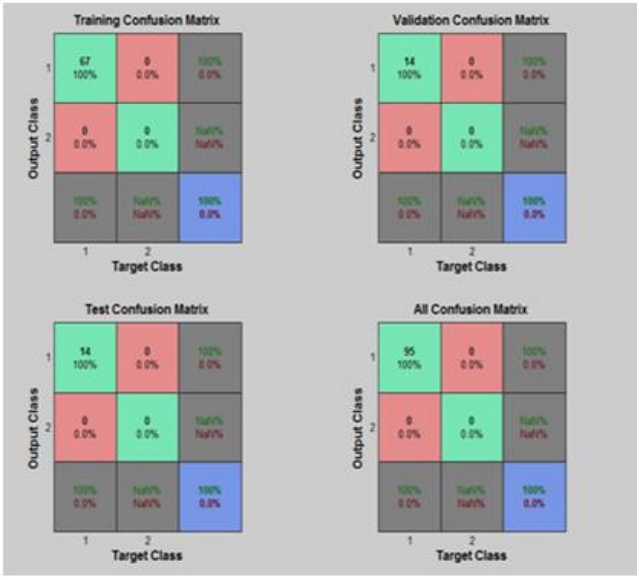

Figure.4 Contingency Table or an Error Matrix Specific Table Layout

This error histogram graphically represents (Figure.5) the distribution of data and a plot of the number of data points in each 20 bin. Experimental histogram depicts zero error after repeated trials of 20 instances.

For the present study a total of 95 points have been used to train the neural network, of which 67 points are chosen for training and 14 points are chosen for validation and 14 points for testing. Total iteration number was set as 1500 at 13 epochs for the learning algorithms and the performance goal is set at 10^4. A Feed-Forward BP was used for modeling the experimental design for predicting the removal capacity of As (III). The experimental design used in this research work was based on one factor experiment at a time. The data and their related statistics are given in Table2. The network is tested with different number of neurons to find the optimal number of neurons at the hidden layer by observing the mean squared error. Thirty two neurons are selected in the hidden layer when mean square error starts decreasing. Learning and momentum parameters are set at 0.20 and 0.10 respectively during the training phase. During training phase the output vector is computed by a forward pass in which the input is propagated forward through the network to compute the output value of each unit [16-18]. The output vector is then compared with the desired vector which resulted into error signal for each output unit.

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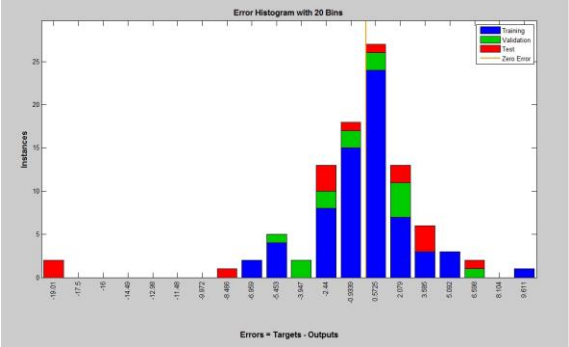

Figure 5 Error Histogram

In order to minimize the error appropriate adjustments were made for each of the weights of the network. After several such iterations the network was trained to give the desired output for a given five input vector. Then network is trained till minimum root mean square error is observed (Table.2) A root mean square error of 0.0012 is observed at epoch number 13 (Figure 3). Training was stopped at this point and weights have been frozen for network to undergo testing phase. A high degree of correlation between actual and predicted sorption efficiency of 100% observed is shown in Figure.4. Coefficient of determination (Figure 6) R^2 = 0.9846 is obtained for training data set. When the network is well trained testing of the network with testing data set is carried out. The prediction ability of the developed network model for responses of experimental data not forming part of the training set. During testing phase output of the data is not presented to the network. The distribution of output of testing data is shown in Figure 6. A high degree of correlation (R^2 = 0.9853) between actual and predicted sorption efficiency (mg/g) is observed as shown in Figure 6 for validation data set. The mean least absolute relative percentage error for training data is observed in Figure 3.

Table 2 The performance of LMFFBP Network for 5-32-1 type of ANN

Training Algorithm	Max (RMSE)	Min (RMSE)	Max (R^2)	Min(R^2)
Levenberg-Marquardt	0.0507	0.0012	1	0.9846

The correlation coefficient (Figure.6) confirms the degree of linear dependence of two random variables. An added variable plot illustrates the incremental effect on the response of percentage removal by removing the effects of all other terms. The slope of the fitted line is the coefficient of the linear combination of the specified terms projected onto the best-fitting direction(R=0.9846). The adjusted response includes the constant intercept terms and averages out all other terms. So we conclude from the Figure 6 there is little noise and there exist linear dependence.

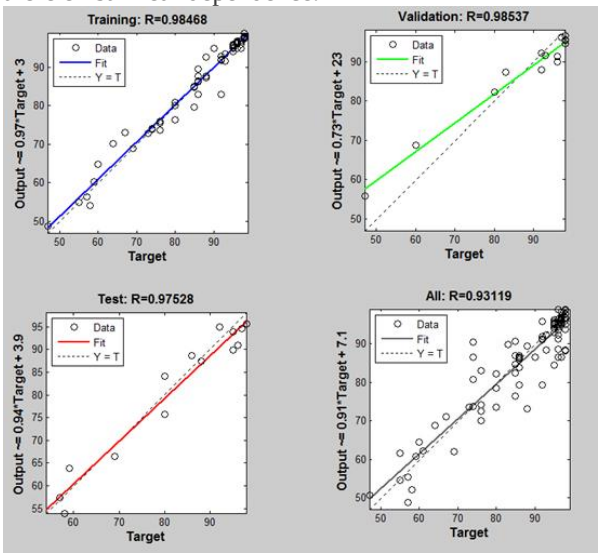

Table 3 Empirical and Mathematical Correlations in the Prediction of Adsorption

The studies conclude that ANN approach is quite efficient in modeling complex adsorption phenomenon. This new paradigm is rapidly opening possibilities in research to combine biological principles with chemical procedure and modeling techniques to spawn ZnO-NPs-AS-*Os* particle with precise function as adsorbent with neural network modeling. This imperative branch of biosynthesis of ZnO-NPs-AS-*Os* as adsorbent enable research to correct chemical defects in unhealthy environment.

Conclusion

- Introduction of this knowledge-based systems is efficient and this green approach confirmed the prediction of percentage adsorption efficiency for the removal of As (III) ions.
- The present piece of work demonstrates the successful removal of As (III) ions from the aqueous solutions using *Ocimum sanctum (*ZnO-NPs-As-*Os)* with maximum removal efficiency (98.08%).
- The three layered ANN modeling technique was applied to optimize this process.
- The Levenberg–Marquardt algorithm (LMA) was found best of BP algorithms with a minimum mean squared error (MSE) for training and cross validation as0.0012 and 0.0017respectively.
- The maximum removal As (III) ions of 98% is obtained at concentration of 0.07N, absorbent

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dosage to 3g , contact time 30 min and agitation speed of 250 rpm at pH 6 .

- The correlation coefficient (R=0.9846) confirms the degree of linear dependence of two random variables.
- The present outcome recommends that ZnO-NPs-AS-*Os* synthesized in a inventive green method may be used as an economical and effectual adsorbent for the confiscation of As (III) ions from aqueous solutions.
- The power of the proposed neural isotherm models lies in the universality of its application. The ability of a single unifying model capable of representing data for all recognized types of adsorption isotherm models is an achievement that classic adsorption isotherm models cannot attain individually.

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