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# Prediction and simulation of Chromium (VI) ions removal efficiency by riverbed sand adsorbent using Artificial Neural Networks

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## Abstract

In the present work removal of chromium from aqueous solution using riverbed sand as adsorbent was studied. The initial Cr (VI) concentration was varied from 10 mg/L to 100 mg/L with varying amount of riverbed sand (0.025 – 0.2 gm) in laboratory batch adsorption experiment. The maximum adsorption efficiency was found at initial Cr (VI) concentration of 10 mg/L, adsorption dose of 0.2 g/L and pH of the solution of 2.0. The equilibrium contact time was found at 90 min. A three layer feed forward artificial neural network (ANN) with back propagation training algorithm was developed to model the adsorption process of Cr (VI) in aqueous solution using riverbed sand as adsorbent. The neural network architecture consisted of tangent sigmoid transfer function (tansig) at hidden layer with 10 hidden neurons, linear transfer function (purelin) at output layer and Lavenberg-Marquardt (LM) backpropagation training algorithm. The neural network model predicted values are found in close agreement with the batch experiment result with correlation coefficient (R) of 0.995 and mean squared error (MSE) 0.0043975.

**Keywords**:Artificial neural network, adsorption, Chromium, back propagation algorithm, Lavenberg-Marquardt..

## Introduction

Heavy metal pollution occurs in many industrial wastewater such as those produced by metal-plating facilities, dyeing operations, mining and metallurgical engineering, electroplating, nuclear power plants, aerospace industries, battery manufacturing processes, the production of paints and pigments, glass production, and municipal and storm water runoff. This wastewater commonly includes Ni, Cu, Cd, Cr, and Pb [1]. Chromium compounds are widely used in industries such as leather, textile, metal plating, battery and pigments. In the aqueous media chromium exists mainly in two states: Cr (III) and Cr (VI) [2, 3]. Cr (VI) exists as extremely soluble and highly toxic chromate ions ($HCrO_4^-$ or $Cr_2O_7^{2-}$) which can transfer freely in aqueous environments [4]. Persistent exposure to Cr(VI) causes cancer in the digestive tract and lungs, and may cause other health problems for instance skin dermatitis, bronchitis, perforation of the nasal septum, severe diarrhea, and hemorrhaging [5, 6]. Therefore, it is essential to remove Cr (VI) from wastewater before disposal. The permitted limit for Cr (VI) in potable water is 0.05 mg /L in both of Iran and USA [7].

The uptake of heavy metal ions from wastewater has attracted a great attention in recent years for global awareness of the underlying detriment of toxic metals in the environment. Application of traditional processes for the uptake of heavy metals has enormous cost, and they cause further environment damage because of the continuous input of chemicals. Hence, easy, effective, economic and ecofriendly techniques are required for fine-tuning of effluent wastewater treatment [8].

Several methods have been employed to remove high amounts of metal ions from aqueous solutions. These methods include coagulation, flotation [9], and chemical precipitation [10], ion exchange [11], and membrane processes [12]. Some of these methods have disadvantages and limitations. Adsorption process provides an attractive alternative treatment to other removal methods because it is more economical and readily available. A lot of nonconventional, low cost and easily obtainable

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adsorbents have been tested for heavy metal removal such as clay minerals [13–15], biomaterials [16] and industrial solid wastes [17, 18].

The mechanism of adsorption is highly complex and is difficult to model and simulate using conventional mathematical modeling. This is mainly due to the interaction of more number of adsorption process variables, and hence the resulting relationships are highly non-linear. Application of artificial neural networks (ANN) has been considered as a promising tool because of their simplicity towards simulation, prediction and modeling. The advantages of ANN are that they require less time for development than the traditional mathematical models, the need for extensive experimentation is avoided as limited numbers of experiments are sufficient to predict the degree of non-linearity and their ability to learn complex relationships without requiring the knowledge of the model structure [19]. ANN models could describe adsorption systems better than general rate models [20]. Even the adsorption isotherms can be represented by neural networks [21]. So, it is preferable to use a non-parametric technique such as a back-propagation neural network to represent such an equilibrium relationship [22].

In the present work, riverbed sand is used as an attractive adsorbent for the treatment of Cr (VI) from industrial leachate because it is cheap and locally available. The effects of various operational parameters, such as initial pH, adsorbent dosage, contact time, initial Cr (VI) concentration and agitation speed on the removal of Cr(VI) are also investigated. On the basis of batch adsorption experiments, a three-layer ANN model to predict the Cr (VI) removal efficiency of riverbed sand used as a low cost adsorbent is applied in this work. Adsorption of Cr (VI) ions from aqueous solution is optimized to determine the optimal network structure. The use of ANN based on back propagation algorithm is also investigated and proposed as an efficient network type for adsorption studies. Finally, outputs obtained from the models are compared with the experimental data.

**Materials and method**

**Materials:**

Stock solution (100 mg/L) of Cr (VI) ions was prepared by using $K_2Cr_2O_7$ purchased from Merck Ltd., India. The reagents NaOH and HCl were purchased from Merck Ltd., India. Chemicals and reagents used were of analytical reagent grade.

**Preparation of adsorbent and adsorbate:**

The riverbed sand was collected from Suruliriver named as Suruli Theertham, Theni District, Tamilnadu, India. The soil samples were initially sun dried for 7 days followed by drying in hot air oven at 383±1 K for 2 days. The dried soil was crushed and sieved to give a fraction of 150 mesh screen and then stored in sterile, closed glass bottles till further investigation [23]. A stock solution of chromium (VI) with concentration 100 mg/L was prepared by dissolving 0.2828 g of $K_2Cr_2O_7$ in deionised (D.I) water. The stock solution was further diluted to desired concentrations.

**Instrumentation techniques:**

The concentration of Cr (VI) ion in aqueous phase at equilibrium was measured by UV-Vis spectrometer (JASCO V530). Chromium concentration was measured by diphenylcarbazide method at 540 nm.

**Batch adsorption studies**

The batch tests were carried out in glass-stoppered, Erlenmeyer flasks with 100 mL of working volume, with a concentration of 10 mg/L. A weighed amount (0.2 g) of adsorbent was added to the solution. The flasks were agitated at a constant speed of 500 rpm for 90 minute in a magnetic stirrer at 313 K. The influence of pH (2.0–8.0), initial Cr (VI) concentration (10, 20, 30, 40, 50, 60, 70, 90, 100 mg/L), contact time (15, 30, 45, 60, 75, 90 min), adsorbent dose (0.05, 0.075, 0.1, 0.15, 0.2, 0.25, 0.3 g/100 ml) and agitation speed (100 – 800 rpm) were evaluated during the present study. Samples were collected from the flasks at predetermined time intervals for analyzing the residual chromium concentration in the solution. The residual amount of chromium in each flask was investigated using UV-VIS spectrophotometer. The amount of Cr (VI) adsorbed in mg/g at time *t* was computed by using the following equation:

$$q_t = \frac{(C_0 - C_t)V}{m} \qquad (1)$$

Where $C_0$ and $C_t$ are the Cr (VI) concentrations in mg/ L initially and at given time t, respectively. V is the volume of the Cr (VI) solutions in ml and m is the weight of riverbed sand in mg. The amount of Cr (VI) ions adsorbed in milligram per gram was determined by using the following mass balance equation:

$$q_e = \frac{(C_0 - C_e)V}{m} \qquad (2)$$

Where qe is the amount of Cr (VI) ion adsorbed onto per unit weight of the adsorbent in mg/g, $C_0$ is the initial concentration of metal ion in mg/L, $C_e$ is the equilibrium metal ion concentration in mg/L, V is the volume of adsorbate in ml and m is the

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weight of the adsorbent in mg. The percentage of removal of chromium ions was calculated from the following equation:

$$R\ (\%) = \frac{(C_0 - C_t)}{C_0} \times 100 \quad (3)$$

The effect of initial concentration of Cr (VI), contact time, initial pH, adsorbent dosage and agitation speed was investigated by varying any one of the parameters and keeping other parameters constant.

## Artificial neural network modeling

The progress of empirical models using numerical estimation techniques such as artificial neural network (ANN) can be considered as powerful alternatives for predicting of adsorption system. ANNs were formerly developed from the primary concept of AI that effort to simulate the process of human brain and nervous system [24, 25]. They contain a series of mathematical
correlation that are used to simulate the learning and memorizing operation. ANNs learn by example in which an actual measured set of input variables and corresponding output are presented to determine the rules that govern the relationship between the variable [24]. ANNs are considered to be powerful in capturing non linear effect and are practically applicable to every situation in which a relationship albeit highly non linear exist between the independent and dependent variables [26]. Networks consist of three main layers which are called input, hidden and output layers [27]. In this work a multilayer feed forward neural network have been used. In this set of networks information moves in only one direction forward from the input layer to the hidden layer and to the output. Running of neural network carried on in two stages, learning or training and testing. The network architecture is presented as l, m, n where l neurons are present at input layer (equal to the number of inputs in the network), m neurons at the hidden layer (optimized through experimentation) and n neurons at the output layer depending on number of outputs desired from model [25]. The all experimental data are divided into three sets: training (70%), validation (15%) and testing (15%). The complete data normalized in the 0–1. Therefore data ($X_i$) are converted to normalized value ($X_{normal}$) as follows [25, 26]:

$$X_{normal} = \frac{X_i - X_{min}}{X_{max} - X_{min}} \quad (4)$$

$X_{min}$ and $X_{max}$ are minimum and maximum actual experimental data. The input signals are modified by interconnection weight known as weight factor ($W_{ij}$), which represents the interconnection of $i^{th}$ node of the first layer to $j^{th}$ node of the second layer. The sum of modified signals (total activation) is then modified by a sigmoid transfer function and output is collected at output layer [25, 27]. Fig. 1 shows network architecture with three layers (a single hidden layer).

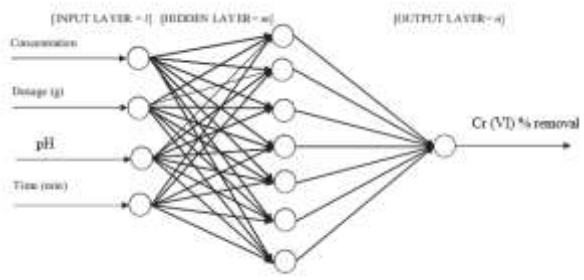


***Figure 1: Artificial neural network architecture***

## Model validation

In order to evaluate the integrity of the fit of experimental data and the prediction accuracy of the models utilized in the present work, the following statistical indices are employed [28]:

$$MSE = \frac{\Sigma\ (q_{exp} - q_{cal})/q_{exp} * 100}{N} \quad (5)$$

The network is tested with different number of neurons to find the optimum number of neurons at hidden layer by observing the mean squared error (MSE). The lowest mean square error is shown for 6 neurons and also maximum R-square is obtained for 6 neurons. Table 1 shows experimental condition for neural network which pH, initial concentration, contact time and dosage are input variable for network. The percentage removal efficiency was selected as targets.

***Table 1: Range of variables***

| S. No | Variables | Ranges |
|---|---|---|
| 1 | Amount of adsorbent dose (g/L) | 0.025-0.4 |
| 2 | Initial concentration of Cr (VI) (mg/L) | 10-100 |
| 3 | pH | 2-8 |
| 4 | Contact time (min) | 15-120 |
| 5 | Agitation speed (rpm) | 100-800 |
| 6 | Cr (VI) removal efficiency | 0-100 |

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| | | |
|---|---|---|
| | (%) | |

**ANN software**

The training, validation and testing of the ANN model was carried out using MATLAB 7 software with neural network tool box. A three layer feed forward neural network tansig transfer in the hidden layer and purelin transfer function in the output layer was used. The Levenberg-Marquart algorithm was used for the ANN model training.

## Results and discussion

**Effect of Dosage:**

The adsorbent dosage serves as an important parameter in adsorption process; this is because it determines the capacity of an adsorbent for a given initial concentration from the adsorbent. The batch experimental result revealed that the effect of adsorbent dosage on the percentage of Cr (VI) ions removal was observed for adsorbent dosage of 0.025 to 0.4 g/L. The effect of riverbed sand dosage on Cr (VI) adsorption was studied at an optimum pH of 2, 10 mg/L initial ion concentration of Cr (VI) ions, contact time of 90 min and agitation speed of 500 rpm. The percentage of adsorption increase from 0.025 to 0.2 g/L. Further increases of adsorbent amount beyond 0.2 g/L do not affect the adsorption significantly this observation can be explain in terms of availability of active sites on the adsorbent surface. The graph of percentage removal of Cr (VI) versus pH and initial adsorbent dosage was represented in 3D plots as represented in Figure 2.

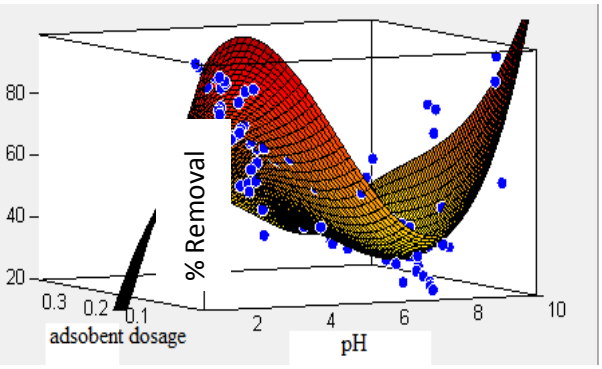


***Figure 2. Effect of pH and adsorbent dosage on Cr (VI) removal percentage***

**Effect of concentration:**

The principal driving force for overcoming the total mass transfer resistance of Cr (VI) ion between the solid phase and aqueous phase is the initial metal ion concentration. Effect of initial concentration of Cr (VI) ion was determined by mixing 0.2 g/L of adsorbent with 200 mL solution containing 10 to 100 mg/L. Increasing the initial Cr (VI) ion concentration significantly increased the uptake capacity of the riverbed sand. As the initial concentration of Cr (VI) ion increased from 10 to 100 mg L-1, the removal efficiency tended to decrease from 100 % to 61 % as shown in Figure 3. It was also clearly observed that, as the adsorbent dosage increases from 0.25 to 0.4 g, the removal percentage increased as well from 25 % to 100%. Finally, these results indicate that the adsorption process of different heavy metal ions on riverbed sand depends, to a certain extent, on the concentration of the adsorbate.

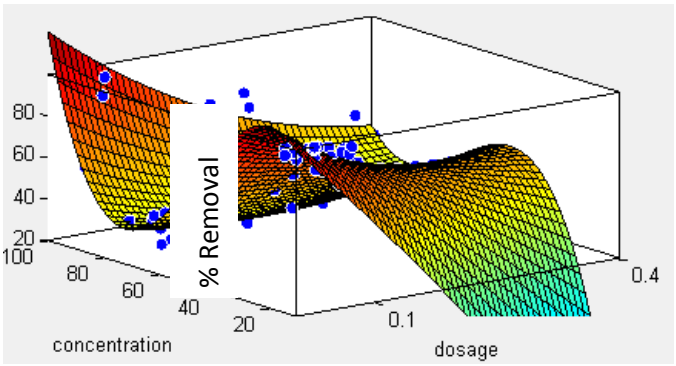


***Figure 3. Effect of concentration of metal ion and adsorbent dosage on Cr (VI) removal percentage***

***Effect of contact time***

The contact time was varied in the range 0-120 minutes between the adsorbate and adsorbent to study the time dependent behavior of adsorption of Cr (VI) onto riverbed sand. The initial concentration of Cr (VI) was kept at 10 mg/L, agitation speed at 500 rpm and riverbed sand dose was 0.2 g/L at pH 2. The result showed that the percentage removal of Cr (VI) increased when agitation time was increased and equilibrium was reached after 90 minutes. The adsorption of Cr (VI) on riverbed sand was found to be fast at the beginning and become slower as the contact time was increased until the equilibrium reached. The adsorption rate increased from 45 % to 100% and become constant after 90 minutes when the system reached equilibrium. The graph of percentage removal of Cr (VI) versus contact time and initial adsorbent dosage was represented in 3D plots as represented in Figure 4.

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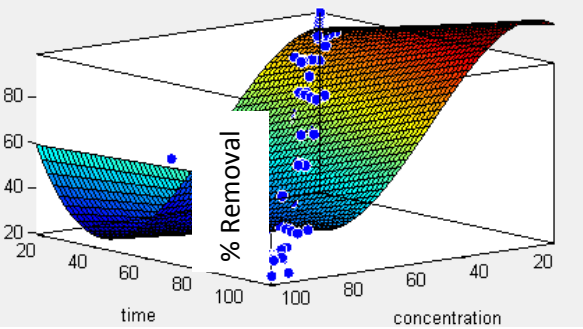


***Figure 4. Effect of contact time and concentration of metal ion on Cr (VI) removal percentage***

**Effect of agitation speed**

The effect of agitation speed rate on Cr (VI) adsorption is shown in Figure and it appears agitation speed has pronounced effect on the amount of Cr (VI) adsorbed. As the agitation speed increased from 100 to 500 rpm, the percentage removal increased from 69.5 % to 99.99 %. However beyond 500 rpm, the adsorption percentage remaind constant and the agitation speed of 500 rpm was selected in subsequent analysis. The increase in adsorption capacity at a higher agitation speed could be explained in terms of the reduction of boundary layer thickness around the adsorbent particles. The graph of percentage removal of Cr (VI) versus agitation speed and initial adsorbent dosage was represented in 3D plots as represented in Figure 5.

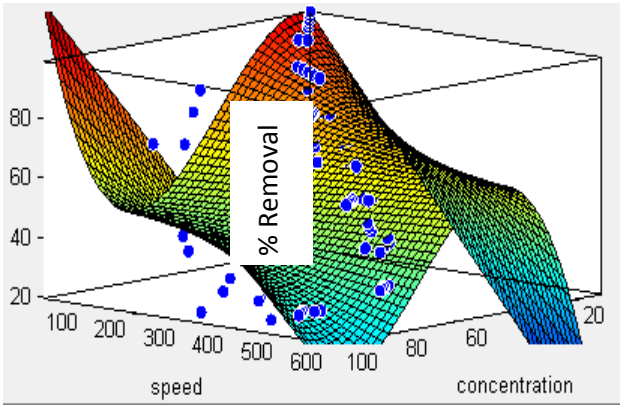


***Figure 5. Effect of agitation speed and concentration of metal ion on Cr (VI) removal percentage***

**ANN model developed for adsorption of Cr (VI) ions**

The feed forward backpropagation (BP) algorithm with Levenberg–Marquardt (LM) training was applied for development of ANN models. The BP is an approximate steepest descent algorithm with MSE used as performance function. In the neural network development, different number of hidden layers, number of neurons in each layer, and type of transfer function for each neuron were analyzed with a learning rate of 1.0 and training goal of $10^{-5}$. Then, the trained networks were tested using the testing data sets and MSE method by modifying the network weights. It was found that network with ten hidden layer of neurons is successful. The tansig transfer function was used in the hidden layer and linear transfer function in the output layer. The training of the network was carried out with different number of neurons in the hidden layer with training goal of $10^{-5}$. It was observed that the best network for all the models has 10 neurons in the hidden layer. Figure 6 shows the proposed optimum ANN type. The ANN consists of 3 layers: input, hidden, and output. Input layer has 5 neurons, which use a tangent sigmoid activation function. The hidden layer has 10 neurons, which use a tangent sigmoid activation function; and finally, the output layer has 1 neurons, which use a linear activation function.

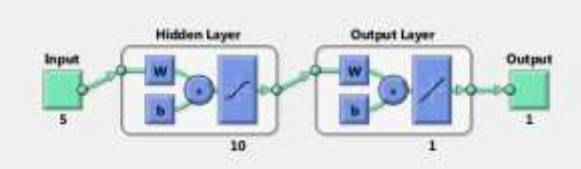


***Figure 6. proposed ANN structrue***

As an initial stages, two neuron was chosen in the hidden layer. With the increase of neuron numbers the MSE value was found decreasing. MSE values was much higher for 2(MSE= 0.4119) and 4(MSE=0.3742) neurons in the hidden layer. With the increase of hidden neurons from 5 to 8, the MSE value decreases from 0.23310 to 0.005917). With further increasing of hidden neurons the MSE value decreasing further and reached minimum value (MSE=0.0043975) at 10 hidden neurons. Hence the neural network containing 10 hidden neurons was selected as optimum case. As neuron number in the hidden layer was increased to 12, the MSE value was found slightly increased to 0.0044573. With further increase in neuron numbers in hidden layer resulted a sharp increase in the MSE value.

The LM training algorithm used was one of the fast training BP methods i.e., Levenberg–Marquardt algorithms. The LM algorithm is designed to approach second-order training speed without computing the Hessian matrix. This algorithm uses the approximation to the Hessian matrix as given in Eq. (6).

$$X_{k+1} = X_k - [J^T - \mu I]^{-1} J^T e \qquad (6)$$

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The LM algorithm is appeared to be the fastest method for training moderate-sized feed forward neural networks (up to several hundred weights). It has better performance than the other methods for function approximation problems. Figure 7 illustrated the training, validation and test mean squared error for the Lavenberg-Marquardt algorithm. The training was stopped after 51 epochs

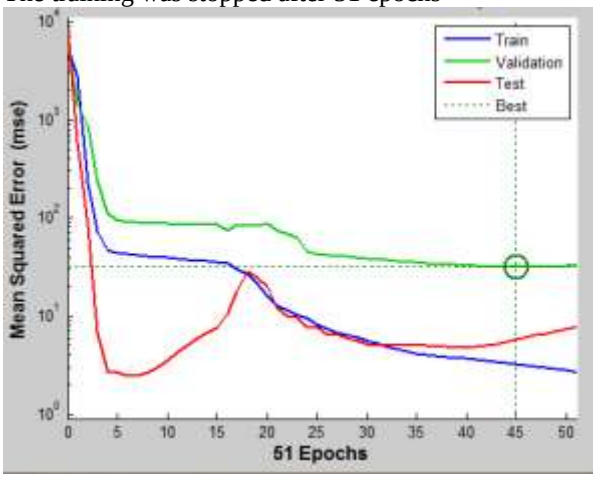


***Figure 7. ANN model training, validation and test mean squared error for Levenberg – Marquardt algorithm***

A regression analysis of the network response between ANN outputs and the corresponding targets was performed. The graphical output of the network outputs plotted versus the targets as open circles is illustrated in Figure 8. Taking into account the non-linear dependence of the data, linear regression shows a good agreement between ANN outputs (predicted data) and the corresponding targets (experimental data). The best linear fit was indicated by a solid red line and $R^2$ is almost 0.995. The performance control of ANN outputs was evaluated by estimating the correlation coefficient ($R^2$) which is defined as [29]:

$$R^2 = \frac{\sum_{p=1}^{N}(t_p - t_{\text{mean}})^2 - \sum_{p=1}^{N}(t_p - o_p)^2}{\sum_{p=1}^{N}(t_p - o_p)^2} \quad (7)$$

where $R^2$ is the correlation coefficient, $N$ is the number of the patterns, $p$ is the index number for pattern, $t_p$ is the target value for the $p^{\text{th}}$ pattern, $t_{\text{mean}}$ is the mean target value, $o_p$ is the output of the $p^{\text{th}}$ pattern which is produced by the ANN model. Test outputs showed a very small deviation in efficiency values from the experimental data.

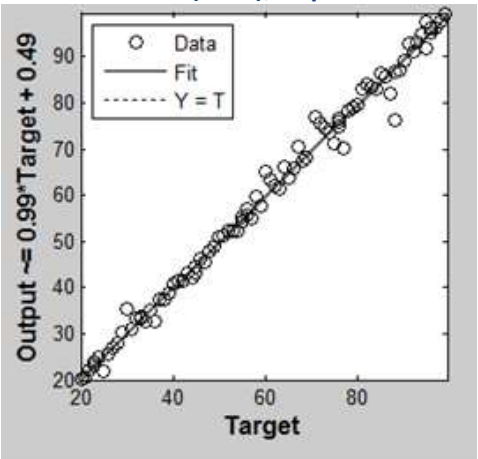


***Figure 8. Regression analysis for adsorption of Cr (VI)***

Figure 9 shows the relationshipmbetween experimental data and BP- ANN model outputs. It can be understood the there is best agreement between calculated and experimental data, which proves that BP – ANN is a most powerful fitting and predictive tool which can describe the adsorption behaviour.

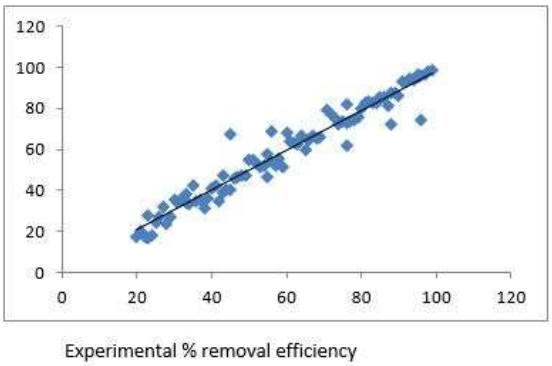


***Figure 9. Comparison between ANN simulated and experimental output data***

## Conclusion

In the present study, a three layer feed forward neural network was optimized to predict the Cr (VI) removal efficiency from aqueous solution using riverbed sand as adsorbent. The model consisted of Lavenberg-Marquardt back propagation training algorithm with tangent sigmoid transfer function (tansig) between input and hidden layer and linear transfer function (purelin) between hidden and output layer. The MSE value was found lowest (MSE=0.0043975) at 10 neurons in hidden layer. A regression analysis was performed between model predict value and experimental data. ANN predicted values are in close agreement with laboratory batch experimental data. The correlation coefficient (R) was

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found 0.995. The present studies showed that the ANN model can effectively simulate and predict Cr (VI) removal efficiency in complex adsorption process.

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