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Artificial Neural Network (ANN) Modelling for Removal of Arsenate from Groundwater by Impregnated Binary Oxide Adsorbent (IBOA)

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ABSTRACT:

In many developing and developed countries, presence of arsenite [As(III)] more than 10 ppb in groundwater is reported. Many people use the arsenic contaminated groundwater for drinking purposes, which is hazardous to human health. Hence need to be removed below the permissible level before use for drinking purpose. In this batch study, IBOA was used as an adsorbent to remove As(III) from water. From experimental data, optimal values for the dose of IBOA, time of contact and pH were found to be 1.0g/L, 24 hrs and 7.0, respectively. In this paper, ANN was applied to the various parameters and found that the results are satisfied in time study. Best fitting was found in dose study and initial concentration study where as in pH study, the ANN results and experimental results are in good agreement. In effect of coexisting ions study, ANN could not map in all cases and not shown the satisfactory results. From experimental data, it is proved that IBOA having good potential for removal of As(III) from groundwater.

Keywords: Arsenite Batch adsorption,ANN

INTRODUCTION

The presence of waste (particularly metal elements) in water bodies, generated by diverse sectors such as smelting, metallurgic and tanneries, is still a scientific and technological challenge worldwide. Arsenic is a metalloid element that is recognized as one of the toxic elements observed in the groundwater in many countries worldwide. Man-made activities such as waste generated from cities and industries, residues obtained from mines, and high doses of chemical fertilizer are the primary sources of arsenic, whereas natural processes such as volcanic eruptions, geochemical reactions, and disintegration of rock (FacundoBarraqu´ et al. 2021). The high level of arsenic in drinking water can results in intestinal, respiratory, pulmonary kidney, cardiac and neurological disorders (Rathi et al. 2021). The acuteness of arsenic effects depends upon the consumption of water contaminated with arsenic and its concentration. The various states of occurrences of arsenic in the environment are (-3, 0,+3 and +5) (Smedley P.L. and Kinniburgh G 2002). Arsenic is found in different forms in water, i.e. As(III), As(V), organic forms, monomethylarsenic acid (MMA) and diamethylarsinic acid (DMA). Arsenic has been categorized as a Class A human carcinogen (Ghosh et al. 2018). As(III) in water is more toxic than As(V) by 25-60 times and 70 times as toxic as methylated arsenicals (Pillewan et al. 2010, Dhoble et al. 2018). Death due to cancer (13out of 1000 population) is reported when the arsenic concentration in drinking water was more than 50ppb (Ghosh et al. 2018). For drinking water purposes, the permissible limit of arsenic in

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drinking water has been established by several countries. The maximum contaminated level(MCL) of arsenic in drinking water in Japan (10ppb), Canada (25ppb), Taiwan (50ppb) and the acceptable level for arsenic in India is 10 ppband permitted limit in the absence of alternative sources, 50ppb (Dhoble et al. 2018, BIS 10500- 2016). Arsenic is removed from water using a variety of processes, including Reverse osmosis (RO), Nano-filtration (NF), Electrodialysis (ED), Coagulation and Flocculation, Oxidation and

Filtration, Ion exchange process, Membrane Filtration (MF), Electrochemical Techniques (EC) and adsorption. In this study, the adsorption technique is used to removes arsenic from water.

Hence providing safe drinking water to the people is the major task for the researchers byadopting low-cost technology which will be eco-friendly and sustainable. With this view, the attempt has been made to develop a low-cost adsorbent (IBOA) to remove arsenic from groundwater and to reduce the level of arsenic less than the permissible level (10 ppb). In this paper, an attempt has also been made to simulate of the various parameters of the experimental data of batch study with ANN and discussed the results observed after simulation.

1 MATERIALS AND METHODS

1.1 Materials

1.1.1 Preparation of IBOA

In this chitosan was impregnated by iron and aluminum oxide and prepared IBOA. The details for the preparation of IBOA and all the materials used in this study is discussed in the published research paper (Dhoble et al. 2011).

1.2 Methods

The details of batch study is discussed in the published research paper (Dhoble et al. 2011).

1.2.1 Batch study

In this study, prepared IBOA was added in the As(III) of 50ml volume in a 100 ml capacity bottle and rotated in a rotary shaker at 150rpmat 27^{O}C at neutral pH. After a fixed interval of time, the samples were taken out and measured remaining the As(III) concentration in the samples by Atomic absorption spectrophotometer hydride vapour generator (AASHVG-1) 6300 Shimadzu Japan 2007.

1.2.1.1 Time study

Time of contact is an essential factor on which adsorption is dependent. In this, IBOA (50mg) was added in 50ml of As(III) water samples with neutral pH and kept in a rotary shaker at 27^{0}C, rpm 150. Samples were then removed at different time intervals (1-24 hrs), and the remaining As(III) concentrations were measured in the samples. This procedure was continued until the As(III) concentration level was less than 10ppb in the sample and finalised the contact time.

1.2.1.2 Dose study

In this study, at neutral pH, the dose of IBOA was adjusted from 0.01 g/L to 2.0 g/L, with an initial concentration of 1.0 mg/L and kept in a rotary shaker at 150 rpm. Samples were removed at a finalised contact time and measured the remaining As(III) concentration in samples. This procedure was continued until the As(III) concentration level was less than 10ppb and finalised the dose of IBOA.

1.2.1.3 pH study

In adsorption, pH plays a significant role in removing As(III) from water**.** It is reported pH of groundwater contaminated with arsenic is in the range of 7 and 9. With this view, a pH study wasconducted by varying pH from 3.0 to 11; initial concentration of 1mg/L, agitation speed 150rpm,and dose of adsorbent 1.0g/L.

1.2.1.4 *Initial concentration study*

To know the consequence of the initial concentration of As(III) on adsorption, the concentration of As(III) was varied from 0.1 to 2.0 mg/L with the IBOA dose (1.0g/L), 150rpm, pH (7.0) and contact time 24 hrs and measured the As(III) concentration present in samples after 24 hrs in each sample.

1.2.1.5 *Effects of coexisting ions*

In groundwater, various cations and anions are present, which may have positive and negative effects for As(III) removal from water depending upon the concentration of cations and anions present in water**.** The salts $Ca(NO_3)_4.H_2O$, $MgSO_4.7H_2O$, Na_2SO_4, Na_2HPO_4 and $NaNO_3$ (Merck India) were used. In this study for effect of Ca^{++} and Mg^{++} concentration was varied from (50- 600mg/L), SO_4 (50-800mg/L), PO_4 (100-800mg/L) and NO_3 (100-800mg/L) measured the effects of each for arsenite removal from water [conditions: As(III) 1mg/L, pH 7.0, Dose of IBOP (1.0 g/L), contact time 24 hrs, rotational speed (150rpm)] and measured the remaining concentration of As(III) at a fixed interval of time.

3.0 MODELLING

ANN is one of the most popular data-driven approaches developed in the pedigree of machine learning techniques (Palit A K and Popovic D, 2006). It works in a similar way the human brain neurons learn from the information sensed by human senses. It is widely applied as a tool in engineering applications, and it can map the non-linear relationship between the input and output data even in the presence of noise. The classical approaches do not have this feature, making ANN a popular tool among researchers (Tang et al. 2013, Kane & Andhare 2019, Alrashed et al. 2018). The architecture of ANN comprises the input layer, hidden layer and output layer. During training, the weights randomly assigned initially to the input layer are adjusted until the satisfactory relations among the input and output parameters are mapped. In this work, the multilayer feed-forward back propagation neural networks are used with the two hidden layers and ten nodes in each hidden layer in the MATLAB computing tool. (https://in.mathworks.com/discovery/neural-network.html; https://in.mathworks.com/help/deeplearning/ug/choose-a-multilayer-neural-network-training-function.html). This architecture detail can be seen in **Fig. 1** and **Fig. 2**. The activation function selected in the hidden layer is tan-sigmoid (tanh), and in the output layer is log-sigmoid. The training algorithm selected is '*trainlm*' (Levenber Marquardt) (Hagan et al. 1997). The satisfactory training performance in all cases is ascertained by observing the ANN training performance plot as shown in **Fig. 3**. **Fig. 4** indicates the correlation coefficient of 0.8531 between the predicted and target values After training ANN, the Pearson's correlation coefficient indicating the agreement between the experimental values and ANN predicted values is evaluated. Pearson's correlation coefficient obtains values based on the similarity of the ANN fitting functions predicted values and experimental values observed. For Pearson's correlation coefficient, a *'corr'* function in MATLAB is used, and it is expressed by equation (1)

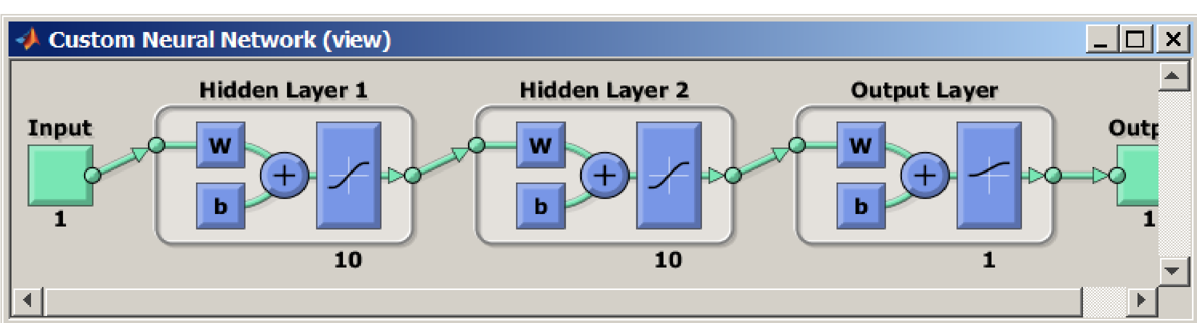

Fig.1 Architecture of ANN selected in Matlab

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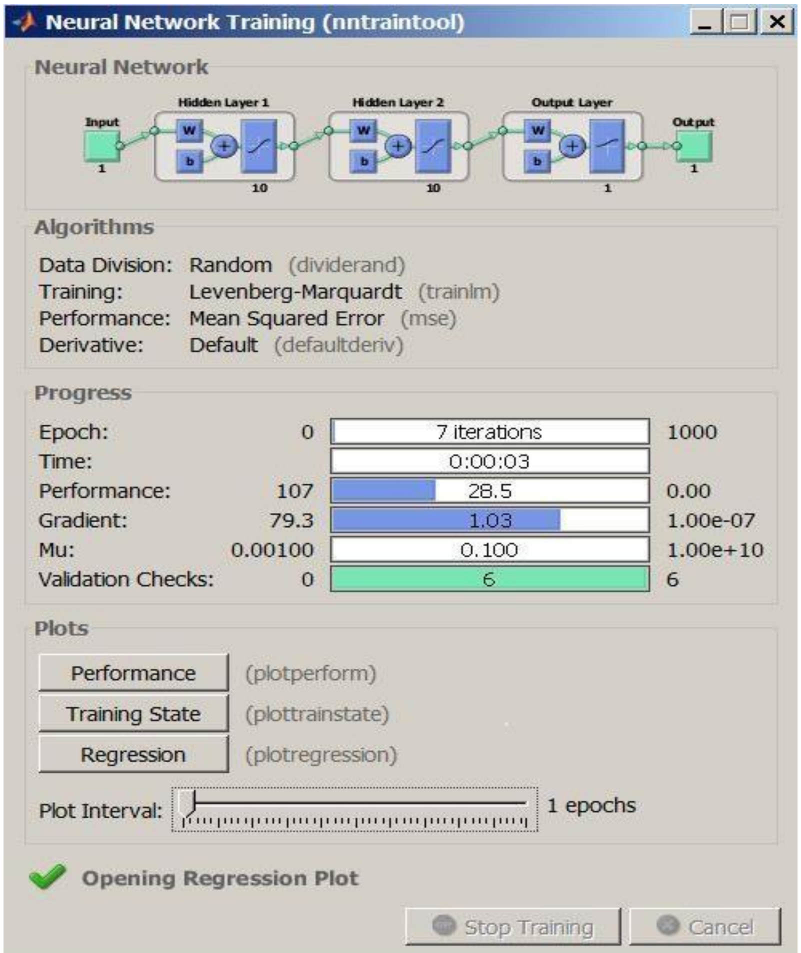

Fig.2 Architecture of ANN selected in Matlab

The outcomes of the experiments and the predictions of ANN are expressed in equation.(1).

$$R = \frac{\sum_{i=1}^{N}(t_p - \bar{t})(O_p - \bar{O})}{\sqrt{\sum_{i=1}^{N}(t_p - \bar{t})^2 \sum_{i=1}^{N}(O_p - \bar{O})^2}} \qquad (1)$$

Where N is the amount of data, t_p is the target values, O_p are the predicted values, $\bar{t}$ and $\bar{O}$ are the averages of the target and predicted values, respectively.

4.0 RESULTS AND DISCUSSION (ANN)

4.1 Time study

From the experimental data it is found that as the time increases, the percentage removal of As(III) also increases. After 22 hrs, it was almost constant with level of As(III) was less than 10 ppb (**Fig. 5**). Hence for practical purpose it is considered as 24 hrs and used same throughout the study. The ANN model architecture as

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https://publishoa.com
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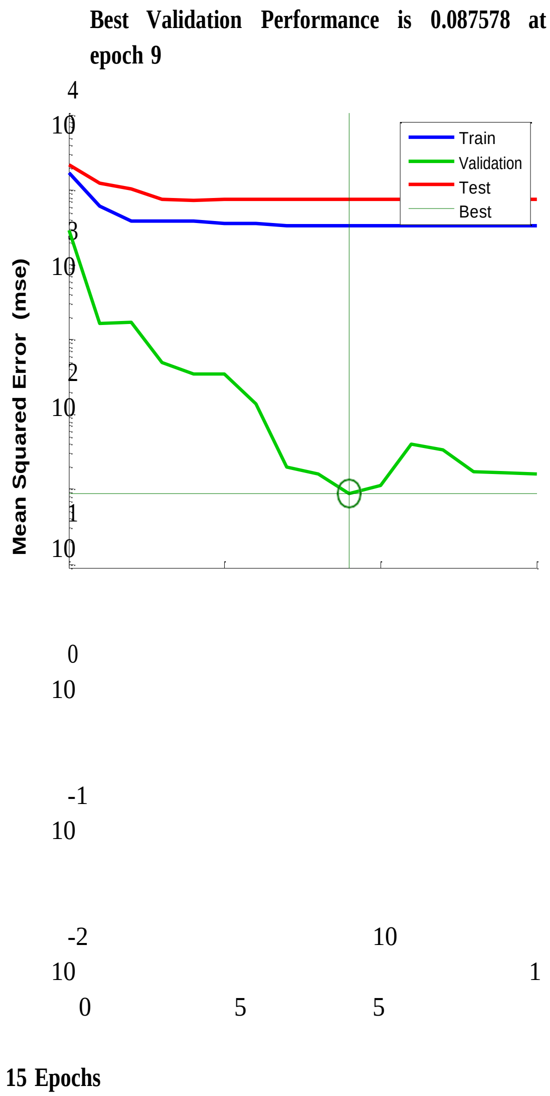
described above is trained with the dataset obtained from the experimental results, and the results of the experimental results and the results predicted by the trained model are compared.



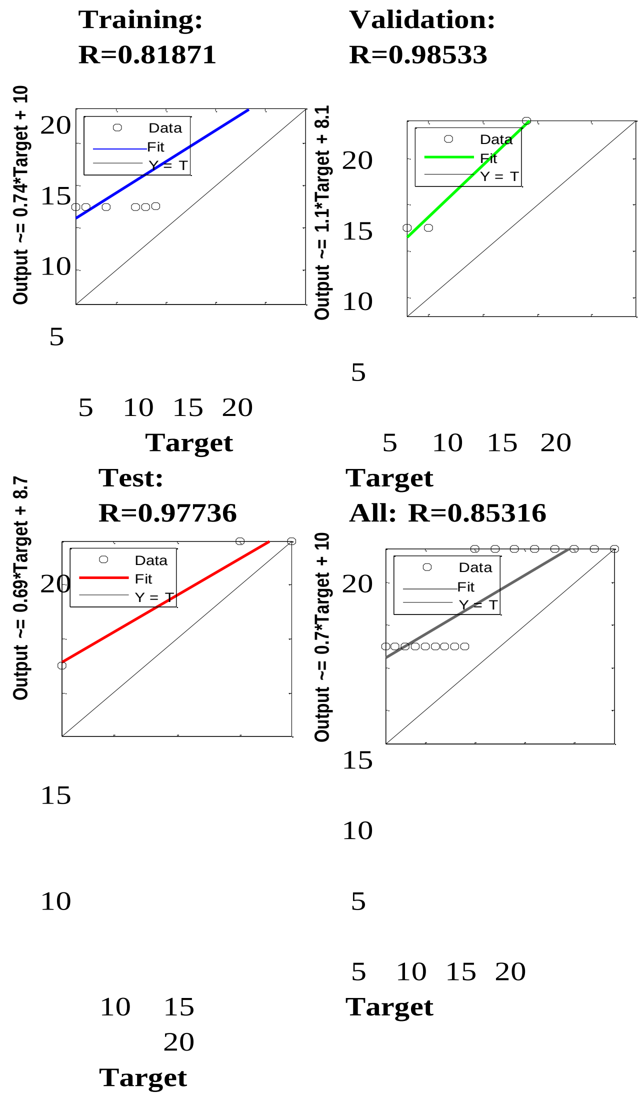


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Fig . 3 ANN Training performance plot

Fig. 4 ANN training regression plot

The training of the ANN is model is found to be satisfactory, as shown in **Fig.** 4 The regression correlation coefficient of 0.85316 is obtained as indicated in **Fig. 4** which shows the satisfactory mapping of the trained and predicted data. Similar plots are observed for other studies for other parameters mentioned below in other studies.

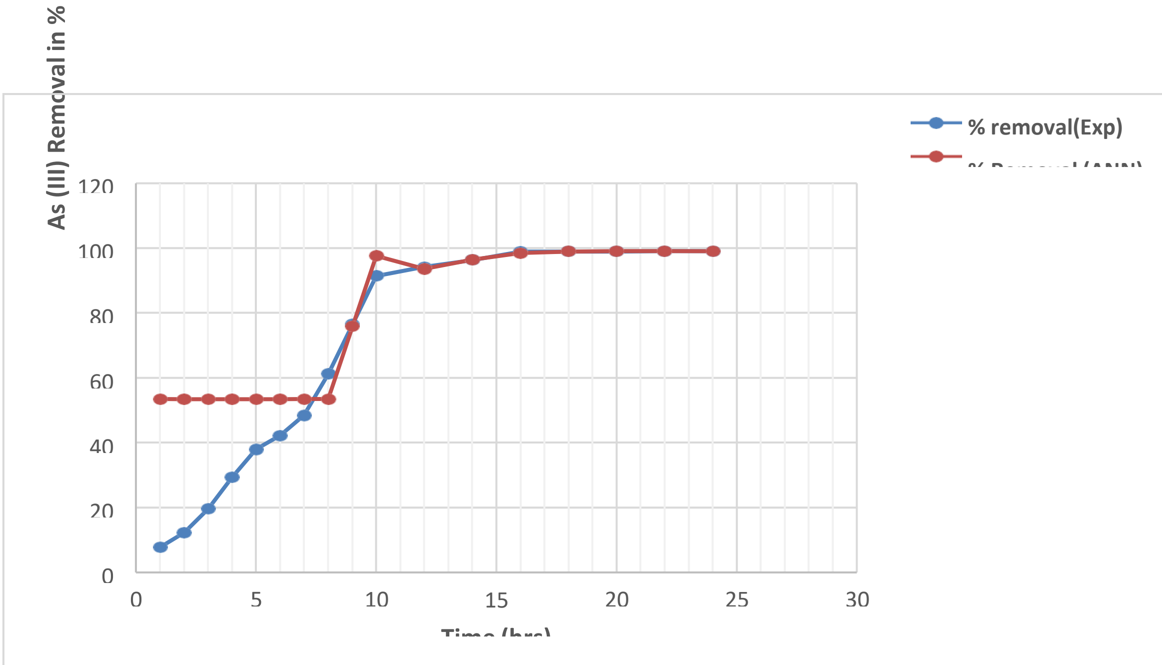

Fig. 5 Comparison of ANN and experimental results (time study)

Fig. 5 shows a satisfactory comparison of the ANN and experimental results. The Pearson's correlation coefficient obtained for the experimental results values and value predicted by ANN is found to be 0.9316, indicating a satisfactory correlation. With a sufficient sample size, a data- driven technique like ANN model shows good potential for predicting the results for different input values.

4.2 Dose study

In this study, at neutral pH, the dose of IBOA was varied from 0.01 g/L to 2.0 g/L with an initial concentration of 1.0mg/L, and from the experimental data, it was observed that removal of arsenite in percentage was enhanced from 27 to 99 (**Fig. 6**). From the obtained data, it was concluded that the optimum dose of IBOA was fixed 1.0 g/L as the concentration of As(III) in the sample was less than the permissible level of arsenic in drinking water (10 ppb) (WHO 2004) and used for further study.

The ANN was satisfactorily trained to obtain the fitting function, and the satisfactory value of the coefficient correlation of 0.8145 is obtained between the target and predicted values. As shown in **Fig. 6**, the ANN and experimental results are good fittings for the experimental results and results given by ANN. The Pearson's correlation coefficient obtained for the experimental percentage removal values and value predicted by ANN is found to be 0.8866, indicating the satisfactory correlation.

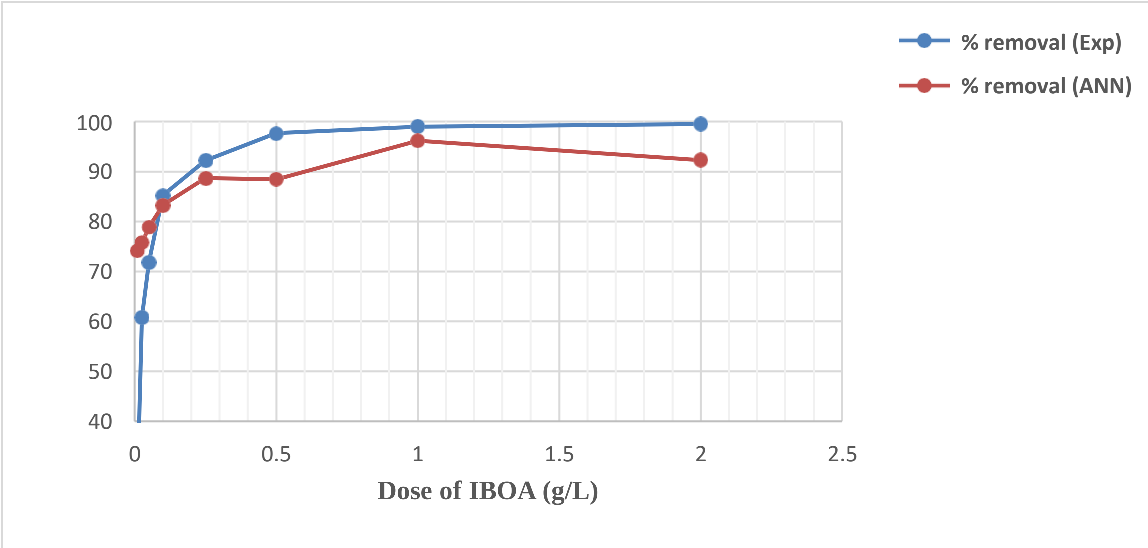

Fig. 6 Comparison of ANN and experimental results (dose of IBOA)

4.3 pH study

From **Fig. 7** it is observed that with increases of pH, the adsorption capacity of As(III) increases gradually upto 7.0 pH value and then it drops down suddenly. It is also observed that there is no significant effect of pH in the range of 3 to 7 on percentage removal of As(III) from water which is practically advantageous; hence the pH was fixed at 7.0 throughout the study.
Based on this pH study experimental data, the ANN was trained to obtain fitting function, and the satisfactory coefficient correlation of 0.9445 is obtained between the target and predicted values. As shown in **Fig.7**, the ANN results and experimental results are in good agreement. The Pearson's correlation coefficient obtained for the experimental % removal values of As(III) and values predicted by ANN is found to be 0.8866, indicating a satisfactory correlation.

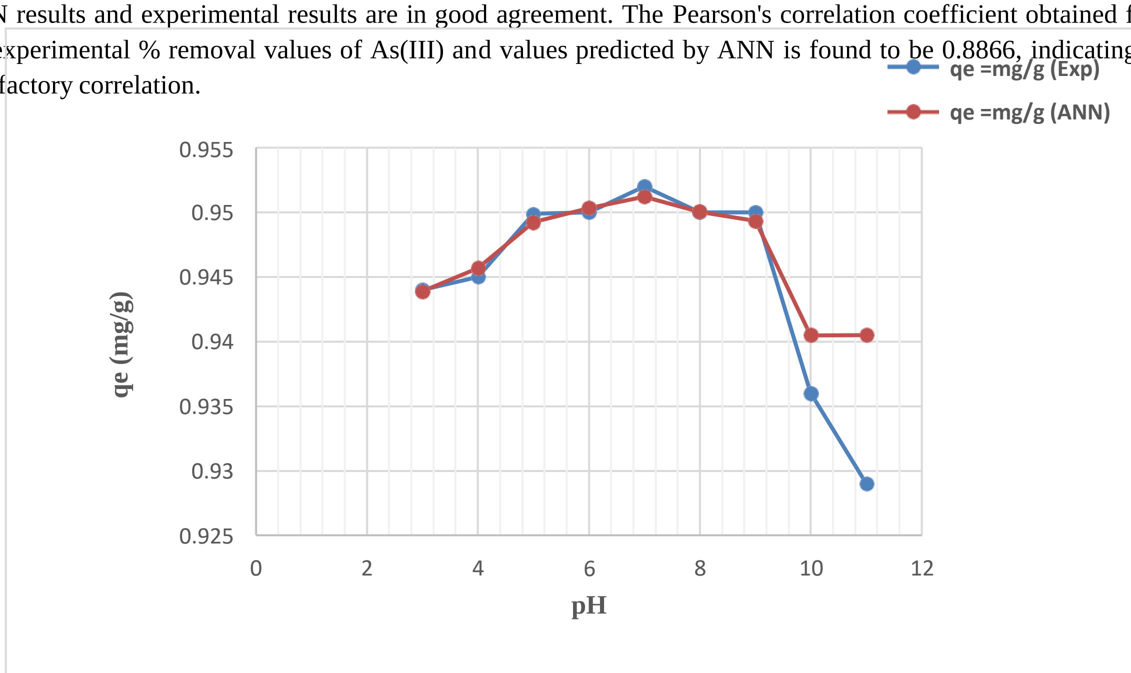

Fig. 7 Comparison of ANN and experimental results (pH)

JOURNAL OF ALGEBRAIC STATISTICS
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4.4 Initial concentration study

The experiments were performed to know the effects of the initial concentration of As(III) on As(III) adsorption by IBOA. From **Fig. 8** it is observed that the adsorption capacity decreased as the initial concentration increased. This could be owing to a lack of active sites on the adsorbent, to adsorb As(III) ions at higher As(III) concentration.

ANN is applied to the experimental data and revealed that the ANN could satisfactorily predict the % removal of As(III) for the given input based on the fitting function obtained. The training correlation coefficient of 0.8393 is obtained while training the ANN. The Pearson's correlation coefficient of 0.8314 is obtained between the experimental result, and ANN predicted values.

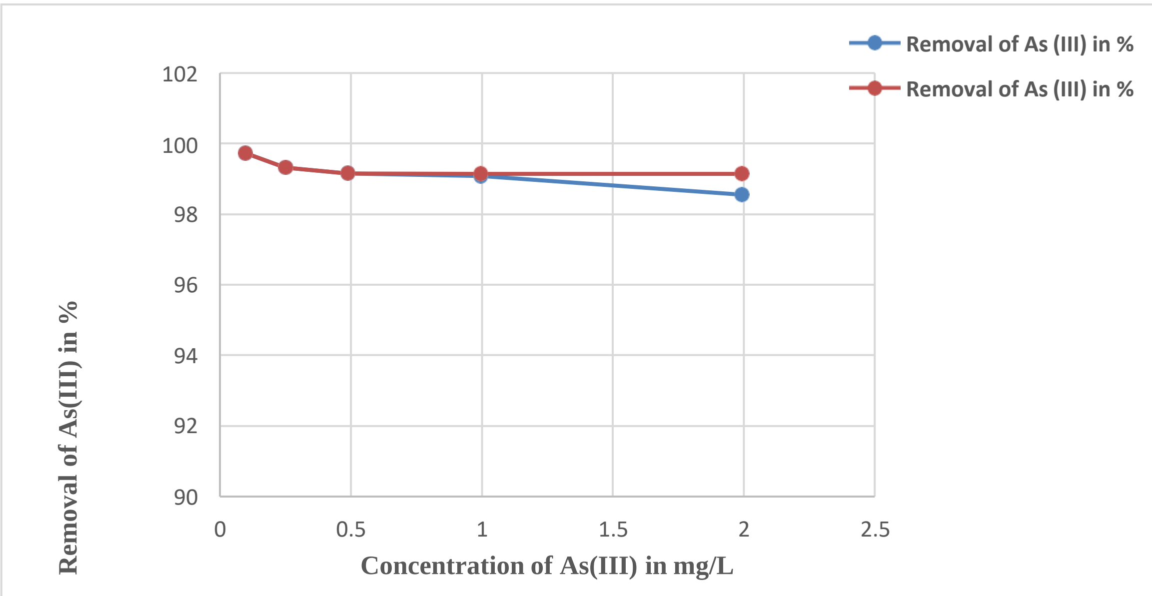

Fig. 8 Comparison of ANN and experimental results (initial concentration)

4.5 Effect of coexisting ions

The results obtained from the experimental data when applied to ANN, the satisfactory results could not be found except on the presence of Mg^{++} and the percentage removal of As(III). This can be attributed to the very small sample size for training the ANN. The Pearson's correlations coefficient found in the case of Mg^{++} for the experimental and ANN results is found to be satisfactory of 0.8492, while for all other cases, it was found to be less than 0.55. The results obtained from the ANN and experimental for each coexisting ions on percentage removal of As(III) results are shown in **Fig. 9 (a- e).**

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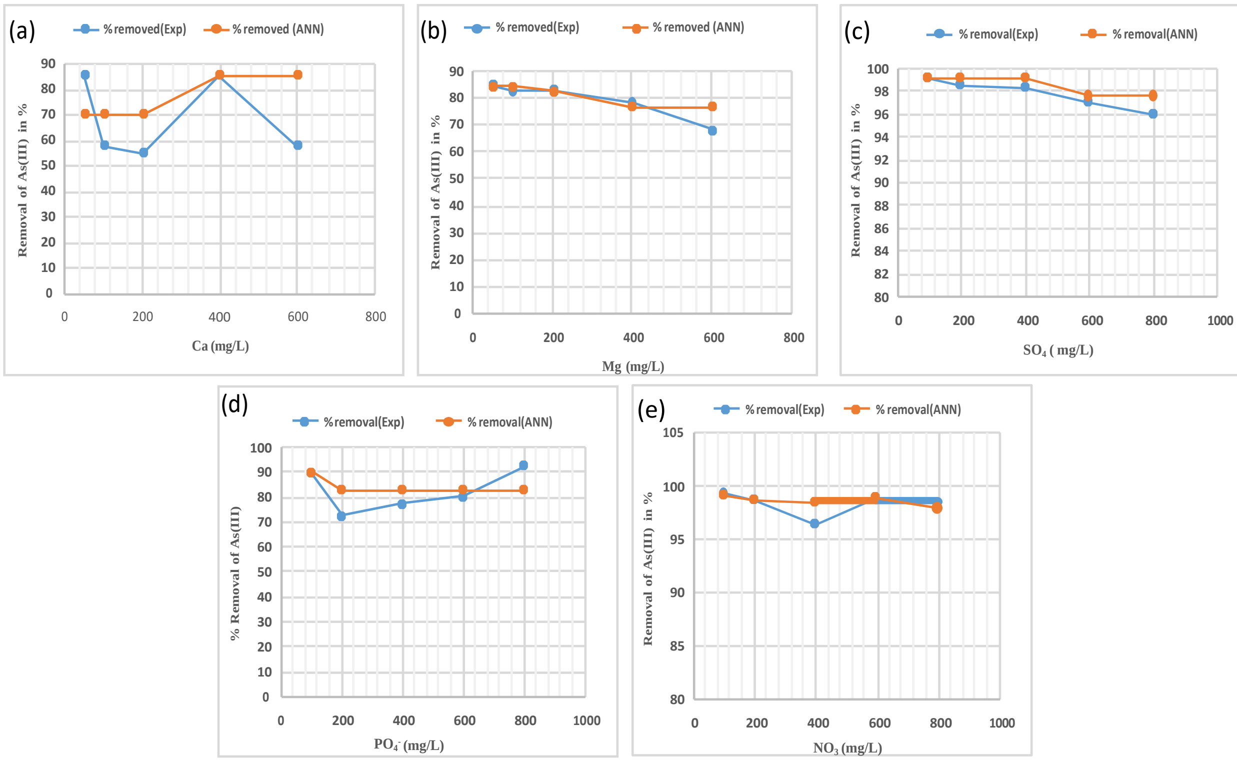

Fig.9 Comparison of ANN and experimental results of a) Ca^{++} b) Mg^{++} c) $SO4^{2-}$d) $PO4^{-}$e) $NO3^{-}$

5.0 CONCLUSION

In this study, ANN was compared with experimental data of time, dose, pH, initial concentration and coexisting ions present in water. The results obtained by applying the ANN fitting function to the experimental data set depicts the ability of ANN to map the non-linear relationship among the input and output values. In the time study, satisfactory results are observed in establishing this fitting function from 5 sec to 25 seconds for the percentage removal of As(III). In the case ofthe dose study, the best fit among the experimental values and the ANN predicted values for percentage removal of As(III) against the input values of dose of adsorbent is found. The pH study was also found to depict the ability of ANN to map the non-linear relationship between the increase in pH and the adsorption capacity of As(III) and the ANN results and experimental results are in good agreement. In the initial concentration study, ANN and experimental values are the best fit from the concentration of 0 to 1.0 mg/L. However, the ANN could not satisfactorily map the relationship between input and output values in all the cases of study of the effect of coexisting ions on percentage removal of As(III), which could be attributed to less sample size. Thus, it can be concluded that tools such as ANN can be effectively implemented to map the relationship among the different input and output parameters. These trained ANN can identify the output for any input value for analysis. IBOA is proved as a promising material to remove arsenite [As(III)] from groundwater below 10ppb with marginal dose of IBOA at neutral pH.

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