Hybrid artificial neural network with imperialist competitive algorithm approach for prediction of adsorption efficiency of Ni(II) and Cd(II) from wastewater by perlite nanoparticles

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Abstract

Contamination of water by heavy metals is a global problem. Nowadays everybody knows that heavy metal ions consist of iron, lead, manganese, zinc, copper, cadmium, and nickel and so on they are common contaminants in wastewater and known to be toxic and carcinogenic that lead to many problems for human and water environment. In this research, experiments have been performed in the batch system to obtain equilibrium data of the individual adsorption of cadmium and nickel ions by perlite nanoparticles. The experiments have been carried out for the chosen temperature of $25\,^{\circ}C$ and operational conditions such as constant agitation and pH 4 and 6 for cadmium and nickel respectively. The results of isotherm show that the Langmuir isotherm showed better correlation with the experimental data. Also, in this paper, the model based on a multilayer perceptron artificial neural network (MLP-ANN) optimized by imperialist competitive algorithm (ICA) to predict of heavy metals removal process (Cd^{2+} and Ni^{2+}) is proposed. ICA is used to decide the initial weights of the neural network. The ICA–ANN model is trained using an experimental data set to approximate the relation between C_0 and C_e are the equilibrium concentration of the cadmium and nickel in solution, and time contact as inputs and adsorption efficiency as output. The performance of the ANN-ICA model is compared with multiple linear regressions (MLR). Coefficient of determination (R^2) and mean square error (MSE) were calculated for the models to compare the results obtained. For the ANN-ICA model to predict of heavy metals, R^2 and MSE are equal to (0.9297 and 0.0141 for Ni^{2+}) and (0.9539 and 0.012 for Cd^{2+}). The results demonstrate the effectiveness of the ANN-ICA model.

Keywords: Adsorption efficiency, Artificial neural network, Imperialist competitive algorithm, Multiple linear regression, Langmuir isotherm.

1- Introduction

The contamination of the heavy metal is an ambient problem of word-wide interest (Fagundes-Klen *et al*., 2007). The heavy metals such as cadmium and nickel are the common contaminants found in the different industrial sewages. The presence of toxic metals in streams and lands, from the release of many industrial sources, such as mine water, metal plating, picking baths, electroplating, tanneries, plastic manufacturing, fertilizers, pigments and metallurgical processes, has received extensive attention throughout the world in recent Decade (Shanmugaprakash and Sivakumar, 2013). Heavy metals are persistent and cannot be degraded or decayed and can be accumulated in biotic organisms (Turan *et al*., 2011a).

Cadmium and Nickel are metals of major concern because of their common uses in developing countries and their persistent nature. Removal and recovery of Heavy metals are very important with respect to environmental and economic considerations (Nurbaş Nourbakhsh *et al*., 2002). Industrial Wastewaters usually contain Cadmium. The maximum level

permitted in wastewater is 0.01 mg/l for cadmium. Cadmium may cause adverse effects such as cancer, cardiovascular disturbances and liver and kidney damage (Hajdu *et al*., 2010; Mashitah *et al*., 2008). Nickel has been identified as one of the toxic heavy metals commonly used in mining, acid battery manufacturing, metal plating etc. (Periasamy and Namasivayam 1995; Ronteltap *et al*., 2007). Higher concentration of Nickel causes cancer of kidneys, lungs, renal edema, pulmonary fibrosis, nausea, vomiting, gastrointestinal distress and skin dermatitis (Akhtar *et al*., 2004). These harmful effects of Nickel require that it removal from wastewaters before it release into streams.

Strict legislation on the discharge of this toxic heavy metal makes it necessary to develop a variety of efficient technologies for the removal of contaminants from wastewater (Turan *et al*., 2011b). Several conventional treatment technologies for heavy metal remove linclude chemical precipitation (Martins *et al*., 2010), evaporation (Sahoo *et al*., 2014), ion exchange (Yuan *et al*., 2010), filtration membrane technology (Song *et al*., 2011), adsorption process (Chiban *et al*., 2011) and electrochemical processes. However, these processes are economically non-feasible, especially when the heavy metal concentration is upper than 100 mg/l (Miretzky *et al*., 2006; Seifpanahi Shabani *et al*., 2013). Among all the techniques mentioned above, the adsorption process is very popular and efficient due to simplicity and low cost. Several sorbent materials which are capable of absorbing metal ions from aqueous solutions such as zeolites (Ardejania *et al*., 2008), clays (Sadeghi *et al*., 2012), activated carbon (Ahn *et al*., 2009), microbial biomass (Ahluwalia and Goyal 2007), diatomite (Niu *et al*., 2007), perlitee (Ghassabzadeh *et al*., 2010), red mud (Lopes *et al*., 2013), chitosan (Kyzas *et al*., 2013), sawdust (Pehlivan and Altun 2008), clay (Sheikhhosseini *et al*., 2013) and agricultural residues (Sud *et al*., 2008), but less researches have focused on heavy metal ions adsorption by natural mineral adsorbents with nano size particles. Nanoparticles present good adsorption efficiency especially, because of particle size reduction and increase number of active sites for interaction with metallic ions (Diallo *et al*., 2005; Kardam *et al*., 2013). In the present study used perlitee as nanoparticles for removal of two heavy metal ions Ni(II) and Cd(II) ions, because of their environmental significance.

The applications of mathematical models to describe the biosorption process can give a quantitative its evaluation as well as will help to optimize its operational conditions. The mechanism of biosorption is highly complex and is difficult to model and simulate using conventional mathematical modeling. This is mainly due to the interaction of number adsorption process variables, and hence the resulting relationships are highly non-linear (Van Deventer *et al*., 1995). Adsorption isotherms are inadequate to accurately predict the extent of adsorption and reproduction of results.

During the last years, artificial neural network (ANN) modeling was used often in different separation and technological applications, mainly due to its powerfulness for solving complex multiple regression problems. ANN's ability for mapping non-linear relationships between the inputs and outputs of a system has extended the field of applications of ANN modeling (Simpson 1990).The main goal of this paper is to extend the ANN's approach in order to finding appropriate initial values for optimization main parameters such as weighting and Biases.

Some evolutionary algorithms such as Particle Swarm Optimization (Price and Bauer 1985) and Genetic Algorithm (Hajihassani *et al*., 2014) can be used for this determination. Recently, a new evolutionary algorithm has been proposed by Atashpaz-Gargari and Lucas (2007), which has inspired from a socio-

political evolution, called Imperialist Competitive Algorithm (ICA). This evolutionary optimization strategy has shown great performance in both convergence rate and better global optima achievement (Biabangard-Oskouyi *et al*., 2008; Gargari *et al*., 2008; McCulloch and Pitts 1943; Niu *et al*., 2014).

In the present work, we propose ICA for optimizing the weights of feed-forward neural network. Then simulation results demonstrate the effectiveness and potential of the new proposed network for heavy metals removal process (Cd^{2+} and Ni^{2+}) prediction compared with multiple linear regression (MLR) using the same data.

2- Materials and Methods

2.1- Sorption Studies

Max effects of particle size (70-120 nm), adsorbent dose (0.19g for Cd, 0.21g for Ni), initial pH value (pH 4 for cadmium, 6 for nickel), initial solution concentration (5–100 ppm) and contact time (0–120 min) on the sorption were investigated. These studies were carried out in 25 °C.

2.2- Batch Nano Sorption Experiments

Stock solutions of 1000 mg/L were prepared by dissolving appropriate quantities of $FeCl_2*4H_2O$, $MnSO_4*4H_2O$, $CuCl_2*2H_2O$, $NiSO_4.\ 6H_2O$, $3CdSO_4.\ 8H_2O$ and $K_2Cr_2O_7$, respectively in a liter of double distilled water. Working solutions were prepared by diluting each stock solution to give the desired concentration and followed by batch adsorption studies at 25 on a multi stirrer hot plate to investigate the sorption processes. Known mass of modified perlitee nanoparticles was added to a 25 mL of the each metal ion solution, thoroughly mixed, and allowing sufficient time for equilibrium. Fast filtration followed and remaining metal ion concentrations were determined. The percentage adsorption of heavy metals from aqueous solution was computed using following formulae:

$$R\% = (C_0 - C_e) * 100 / C_0 \quad (1$$

where, C_0mg/Land C_e mg/Lare the equilibrium concentration of the cadmium and nickel in solution, respectively. Experimental study base on Seifpanahi Shabani *et al.,* (2013) research as details were done.

2.3- Adsorption Isotherm

In this research, Langmuir isotherm was fitted to experimental data for modeling of the equilibrium adsorption of Ni(II) and Cd(II) ions onto perlite nanoparticles. The Langmuir and isotherm given by the following equation:

$$\text{Langmuir: } q_e = \frac{q_0 K_L C_e}{1 + K_L C_e} \quad (2$$

where q_e (mg/g) is the adsorption capacity at equilibrium of cadmium and nickel, C_e (mg/Lis) the equilibrium concentration of the cadmium and nickel in solution, q_0 (mg/g) is the monolayer adsorption capacity and K_L (L/mg) is the Langmuir constant associated to the free energy of adsorption. The linearized forms of Langmuir equation can be written as follows:

$$\frac{1}{q_e} = \frac{1}{q_0} + \frac{1}{q_0 K_l}\frac{1}{C_e} \quad (3$$

3- Hybrid ANN-ICA Model

3.1- Artificial Neural Network

ANN firstly was introduced by McCulloch and Pitts (1943) who presented ability of this technique to calculate any logical functions. Processing of the information is executed with the help of many interconnected simple elements known as neurons which are placed in distinct layers of the network. Multi-layer perceptron, the most famous type of ANNs, consists of at least three layers: input, intermediate or hidden layers and output. Difficulty level of the problem determines the number of the hidden layers and neurons

(Simpson, 1990). The neurons are linked from a layer to the next one, but this connection is not within the same layer. Once a series of inputs presents to the network, the input values are transmitted through the links to the second layer. In every link, the transmitted value is multiplied to the weight of the link. The weighted values are come together at a node in the hidden layer and a bias is summed to the weighted values in that particular node. Consequently, the achieved value transfer to an activation function and a signal is created. Using the departing links of hidden nodes, the results are transmitted to the output layer. Similar to hidden nodes, the input values of the output nodes are weighted, biased, summed and transferred to the activation function. The created values of activation functions in output layers are the outputs of the network. Performance of an ANN is dependent on architecture of the network which is the pattern of the connections existing between the neurons. The network should be trained with sufficient input–output patterns that are known as the training data (Meulenkamp and Grima, 1999). As the error reached specified error goal, training is finished and the optimum model is determined.

3.2- Imperialist Competitive Algorithm

The Imperialist Competitive Algorithm (ICA) is a new global heuristic search in the Evolutionary Computation field based on the imperialistic competition process as a source of inspiration.

ICA is a new socio politically motivated global search strategy that has recently been introduced for dealing with different optimization tasks. In this paper, we have applied this algorithm for optimizing the weights of MLP-ANN.

After dividing all colonies among creating the initial empires and imperialists, these colonies start moving toward their relevant imperialist state which is based on assimilation policy (Tsoulos *et al.,* 2008). Figure 1 shows the movement of a colony towards the imperialist. In this movement, θ and x are random numbers with uniform distribution as showed in following formulae) and d is the distance between the imperialist and colony.

$$x \approx U(0, \beta \times d), \theta \approx U(-\gamma, \gamma) \quad (4$$

where γ and β are parameters that modify the area that colonies randomly search around the imperialist. In our implementation β and γ are considered as 2 and 0.5 (Radian) respectively.

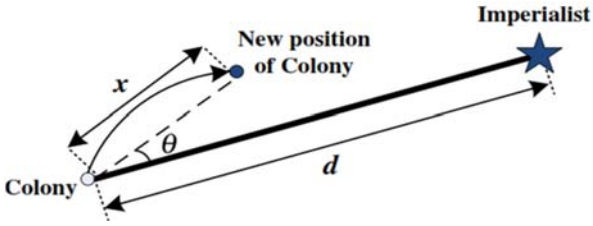

Figure 1) Movement of colonies toward their relevant imperialist (Atashpaz-Gargari and Lucas, 2007).

In ICA, revolution causes a country to suddenly change its socio-political characteristics. That is, instead of being assimilated by an imperialist, the colony randomly changes its position in the socio-political axis. The revolution increases the exploration of the algorithm and prevents the early convergence of countries to local minimums. The total power of an empire depends on both the power of the imperialist country and the power of its colonies which is shown in following formulae:

$$T.C._n = Cost(imperialist_n) + \xi mean\{Cost\ colonoes\ impire_n)\} \quad (5$$

In imperialistic competition, all empires try to take possession of colonies of other empires and control them. This competition gradually brings about a decrease in the power of weaker empires and an increase in the power of more powerful ones. This is modeled by just picking some of the weakest colonies of the weakest empires and making a competition among all empires to possess these colonies. Figure 2 shows the flowchart of the ICA. More detailed mathematical formula andinformation of ICA

was presented in Gargari *et al*., (2008) and Atashpaz-Gargari and Lucas (2007).

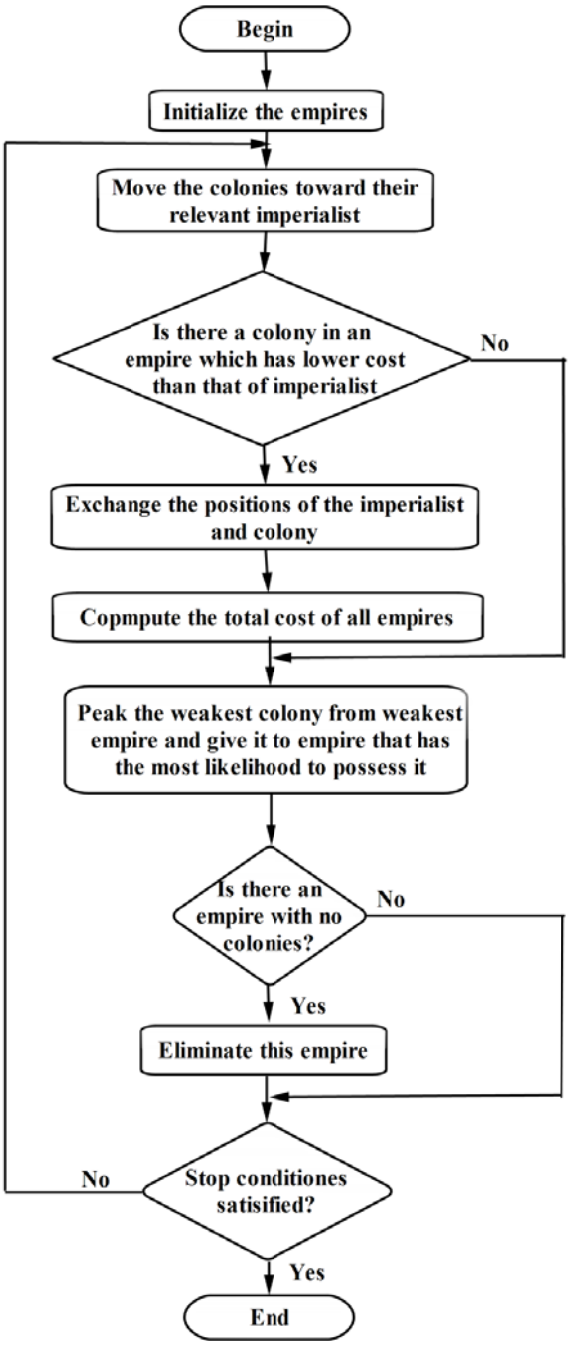

Figure 2) The flowchart of the ICA.

4- Prediction of Adsorption Efficiency Of Ni(Ii) And Cd(Ii) Using Hybrid ANN-ICA Model

To simulate adsorption efficiency of Ni(II) and Cd(II) from wastewater by perlite nanoparticles using hybrid ANN-ICA model, all relevant parameters should be determined, due to the fact that ANNs work based on given data and do not have previous knowledge about the subject of prediction. The following sections describe the inputs and output parameters and simulation of adsorption efficiency of Ni(II) and Cd(II) using hybrid ANN-ICA model.

4.1. Inputs and Output Data

In ANN-ICA modeling, any type of input can be used as long as they have effects on output results. To train and verify the accuracy and ability of the ANN-ICA model, a total of 70 experimental data set (35 data set for Cd^{2+} and 35 data set for Ni^{2+}) were used in this research. In total, three input parameters including C_o, C_e and time output including adsorption efficiency were used. A few samples of the training data sets are shown in Table 1. Also, descriptive statistics of the data sets used for modeling are shown in Table 2.

Table 1) A few samples of the training data sets for Cd^{2+} and Ni^{2+}, ANN-ICA model.

Cd^{2+} (PH=4, temperature=25^0C and Nano Perlite=0.19)				
No.	Input			Output
	C_0 (ppm)	Time (min)	C_e (ppm)	Adsorption (%)
1	5	120	0	100
2	100	10	83.12	3.173
3	25	90	7.3325	73.4
4	50	50	31.195	30.47
5	100	70	83.12	18.88
6	50	70	31.195	38.5
7	100	50	83.12	16.64
8	100	30	83.12	12.65
9	25	70	7.3325	64.71
10	25	120	7.3325	70.67
Ni^{2+} (PH=6, temperature=25^0C and Nano Perlite=0.21)				
No.	Input			Output
	C_0 (ppm)	Time (min)	C_e (ppm)	Adsorption (%)
1	25	50	0.825	54.69
2	25	70	0.825	74.73
3	50	10	32.575	8.188
4	50	30	32.575	20.76
5	100	0	90.46	0
6	5	50	0	94.21
7	100	30	90.46	16
8	15	50	0	90.8
9	15	70	0	95.87

10	5	30	0	87.8

Table 2 Descriptive statistics of the data sets for Cd^{2+} and Ni^{2+}

Cd^{2+} (PH=4, temperature=25^{0}C and Nano Perlite=0.19)			
Parameter	Maximum	Minimum	Mean
C_0(ppm)	100	5	39
Time (min)	120	0	52.85
C_e(ppm)	83.12	0	24.35
Adsorption (%)	100	0	44.98
Ni^{2+}(PH=6, temperature=25^{0}C and Nano Perlite=0.21)			
Parameter	Maximum	Minimum	Mean
C_0(ppm)	100	5	39
Time (min)	120	0	52.85
C_e(ppm)	90.46	0	24.77
Adsorption (%)	100	0	47.7

4.2. Pre-Processing of Data

In data-driven system modeling methods, some pre-processing steps are usually implemented prior to any calculations, to eliminate any outliers, missing values or bad data. This step confirms that the raw data retrieved from database is perfectly proper for modeling. In order to softening the training procedure and improving the accuracy of prediction, all data samples are normalized to adapt to the interval [0, 1] according to the following linear mapping function:

$$x_M = \frac{x - x_{\min}}{x_{\max} - x_{\min}} \quad (6$$

where x is the original value from the dataset, x_M is the mapped value, and x_{min} (x_{max}) denotes the minimum (maximum) raw input values, respectively. It is to be noted that model outputs will be remapped to their corresponding real values by the inverse mapping function ahead of calculating any performance criterion.

4.3. Network Design

In this paper, after building several ANN-ICA models based on trial and error, the best result of each model, listed in Tables 3 and 4, are compared and the one with maximum of coefficient of determination (R^2) and minimum of mean square error (MSE) is chosen. The best ANN architecture for Ni^{2+} was: 3-3-5-1 (3 input units, 3 hidden neurons in first layer, 5 hidden neurons in second layer, 1 output neuron) and for Cd^{2+} was 3-5-4-1 (3 input units, 5 hidden neurons in first layer, 4 hidden neurons in second layer, 1 output neuron). ICA is used as neural network optimization algorithm and the MSE used as a cost function in this algorithm. The goal in proposed algorithm is minimizing this cost function. In these simulations, the number of imperialists and the colonies is considered 30 and 200, respectively; parameter β is set to 2. Figure 3 shows architecture of best ANN-ICA model for prediction of adsorption efficiency of Ni(II) and Cd(II) from wastewater by perlite nanoparticles.

Table 3) Comparison between the best results of some ANN-ICA models for Adsorption efficiency (for Ni^{2+})

No.	Model architecture	Correlation coefficient (R^2)	MSE
1	3-7-5-1	0.564	0.0418
2	3-4-5-1	0.8541	0.0407
3	3-10-14-1	0.4206	0.0615
4	3-24-12-1	0.6888	0.045
5	3-5-2-1	0.8055	0.0298
6	3-6-5-1	0.7886	0.03
7	3-5-5-1	0.4913	0.0341
8	3-18-13-1	0.7616	0.0747
9	3-16-18-1	0.7456	0.0667
10	3-5-4-1	0.8653	0.0332
11	3-5-3-1	0.7867	0.0426
12	3-5-6-1	0.5407	0.628
13	3-2-5-1	0.7849	0.023
14	3-17-22-1	0.8217	0.0373
15	**3-3-5-1**	**0.9297**	**0.0141**

Table 4) Comparison between the best results of some ANN-ICA models for Adsorption efficiency (for Cd^{2+})

No.	Model architecture	Correlation coefficient (R^2)	MSE
1	3-7-5-1	0.8697	0.0171
2	3-4-5-1	0.4747	0.0498
3	3-10-14-1	0.6435	0.0389
4	3-24-12-1	0.8367	0.0366
5	3-5-2-1	0.888	0.0128
6	3-6-5-1	0.752	0.0430
7	3-5-5-1	0.9012	0.030
8	3-18-13-1	0.5982	0.0448
9	3-16-18-1	0.6698	0.0546
10	3-3-5-1	0.6977	0.0426
11	3-5-3-1	0.7816	0.0272
12	3-5-6-1	0.7453	0.0551
13	3-2-5-1	0.9004	0.0191
14	3-17-22-1	0.9207	0.0182
15	**3-5-4-1**	**0.9539**	**0.012**

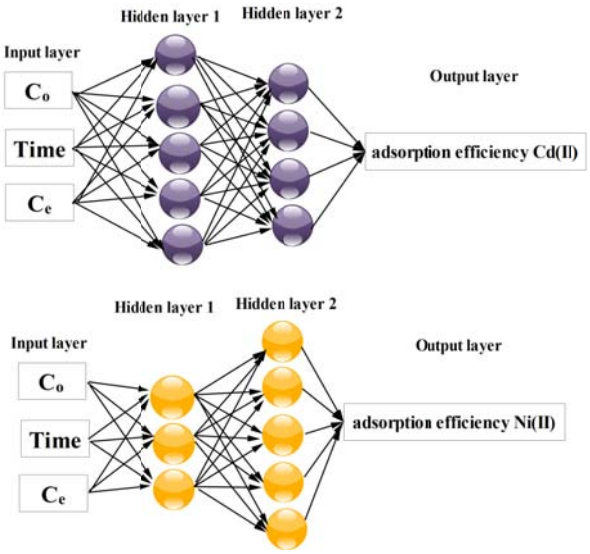

Figure 3) Architecture of best ANN-ICA model for prediction of adsorption efficiency of Ni(II) and Cd(II) from wastwater by perlit nanoparticles.

Also, in this study, tansig was used as transfer function between input and hidden layer, as well as was used as transfer function between hidden and output layer, shown by the following equation:

$$\text{tansig} = \frac{2}{(1+\exp(-2x))} - 1 \quad (7$$

The simulation performance of the ICA–ANN model was evaluated on the basis of MSE and R^2, shown by the following equations:

$$MSE = \frac{1}{N}\sum_{i=1}^{n}(y_i - y_i')^2 \quad (8$$

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(y_i - y_i')^2}{\sum_{i=1}^{n} y_i^2 - \frac{\sum_{i=1}^{n} y_i'^2}{n}} \quad (9$$

where, y and y' are the measured and predicted values respectively and N is the number of samples.

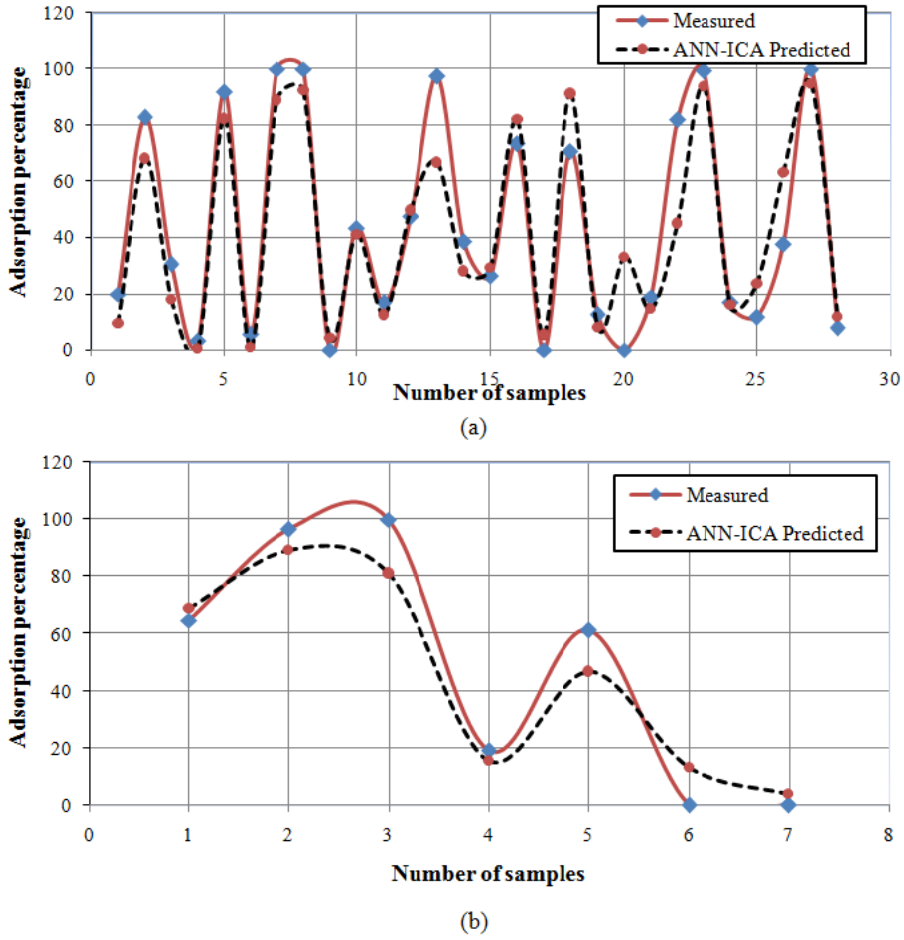

Figure 4) Comparison between measured and predicted Adsorption efficiency (for Cd^{2+}) a) trainingdatasets b) testing datasets.

where R^2 efficiency criterion represents the efficiency of the initial uncertainty explained by the model. The best fitting between measured and predicted values, which is unlikely to occur, would have MSE = 0 and $R^2 = 1$.

A comparison between predicted values of Adsorption efficiency (for Cd^{2+} and Ni^{2+}) by the ANN-ICA model and measured values for 70 data sets at training and testing phases is shown in Figs. 4 and 5. As shown in Figs. 4 and 5, the results of the ANN-ICA model in comparison with actual data show a good precision of the ANN-ICA model (see Table 5).

Table 5) Performance of the model for predicting Adsorption efficiency (for Cd^{2+} and Ni^{2+})

Description		R^2	MSE
Cd^{2+}	Training datasets	0.8505	0.0207
	Testing datasets	0.9539	0.0120
Ni^{2+}	Training datasets	0.8873	0.0204
	Testing datasets	0.9297	0.0141

Performance analysis of the ANN-ICA model for predicting Adsorption efficiency (for Cd^{2+} and Ni^{2+}) is shown in Table 5. Furthermore, correlation between measured and predicted values of Adsorption efficiency (for Cd^{2+} and Ni^{2+}) for training and testing phases are shown in Figs. 6 and 7. In all two states of using training and testing data, the performance indices obtained in Table 5 indicate the high performance of the ANN-ICA model that can be used successfully for the prediction of Adsorption efficiency.

5- Multiple Linear Regression

In this paper, regression analysis was performed using the training and test data employed in ANN-ICA model. Adsorption efficiency (for Cd^{2+} *and* Ni^{2+}) was considered as the dependent variable and C_o, time and C_e were considered as the independent variables. A computer-based package called SPSS (Statistical Package for the Social Sciences) was used to carry out the regression analysis. The estimated regression relationships for Adsorption efficiency (for Cd^{2+} *and* Ni^{2+}) are given as below:

$$\text{for } Ni^{+2} \quad \text{Adsorption percentage} = 72.631 + 0.512 \times \text{time} - 2.949 \times Co + 2.475 \times Ce \qquad (a)$$

$$\text{for } Cd^{+2} \quad \text{Adsorption percentage} = 73.749 + 0.467 \times \text{time} - 2.116 \times Co + 1.356 \times Ce \qquad (b)$$

The statistical results of the model are given in Table 6. Adsorption efficiency (for Cd^{2+} and Ni2+) were estimated according to the Eqs. a and b. Figs. 8 and 9 shows the correlation between measured adsorption efficiency and those predicted using MLR with three inputs for Cd^{2+} and Ni^{2+}.

Table 6) Statistical characteristics of the multiple regression models

Model	Method	Independent variables	Coefficient	Standard error	Standard error of estimate	t value	F ratio	Sig. level	Determination coefficient (R^2)
Eq. a	Enter	Constant	72.631	11.312	19.62693	6.420	31.872	0.000	0.755
		C_o	-2.949	0.736		-4.005		0.000	
		Time	0.512	0.082		6.211		0.000	
		C_e	2.475	0.793		3.120		0.004	
Eq. b	Enter	Constant	73.749	10.393	18.550075	7.096	35.719	0.000	0.781
		C_o	-2.116	0.517		-4.092		0.000	
		Time	.467	0.080		5.821		0.000	
		C_e	1.356	0.496		2.736		0.010	

Tables 7 and 8 compares the correlation coefficient R2 and MSE associated with two methods for both training and test data. It is well illustrated in Tables 7 and 8 that a close agreement can be seen between the predicted and measured data when the ANN-ICA method is used. Low correlation values between the model predictions and measured data using MLR method describes its low capability in prediction adsorption efficiency.

Table 7) The comparison of the results (R^2, MSE) of two methods in training and testing data for adsorption efficiency (for Ni^{2+})

Method	R^2 (train data)	R^2 (test data)	MSE (train data)	MSE (test data)
ANN-ICA	0.8873	0.9297	0.0204	0.0141
MLR	0.6294	0.9826	0.0698	0.0336

Table 8) The comparison of the results (R^2, MSE) of two methods in training and testing data for adsorption efficiency (for Cd^{2+})

Method	R^2 (train data)	R^2 (test data)	MSE (train data)	MSE (test data)
ANN-ICA	0.8505	0.9539	0.0207	0.0120
MLR	0.7542	0.8202	18.29	0.0252

6- Conclusions

A new method to predict adsorption efficiency (for *Cd^{2+} and Ni^{2+})* by experimental data has been presented using ANN-ICA method. The predictions for adsorption efficiency (for *Cd^{2+} and Ni^{2+})* using ANN-ICA method with MLR method are presented and compared with the measured data. The input data for the ANN-ICA and MLR models have been selected based on the high values of the correlation coefficients between adsorption efficiency (for *Cd^{2+} and Ni^{2+})*. It was found that a close agreement was achieved between the predicted and measured concentrations for adsorption efficiency (for *Cd^{2+} and Ni^{2+})* when the ANN-ICA method was used. Low correlation values between the model predictions and measured data using MLR method describes its low capability in prediction adsorption efficiency (for *Cd^{2+} and Ni^{2+})*.

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