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Simulation of Ammonia Nitrogen Removal from Simulated Wastewater by Sorption onto Waste Foundry Sand Using Artificial Neural Network

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Abstract— The present study investigated the removal efficiency of ammonia nitrogen from simulated wastewater by waste foundry sand based on 120 batch experiments which were modeled by three-layer artificial neural network technique. Contact time (5-120 min), pH of the aqueous solution (3-10), concentration (400-600 mg/L), sorbent dosage (20-120 g/100 mL) and agitation speed (50-250 rpm) were studied. Results showed that the best values of the above parameters were time of 90 min, pH= 10, 400 mg/L, dosage of 90g/100 mL and 200 rpm respectively with removal efficiency equals to 95%. The sorption process was described in a good manner using ANN model which consisted of the tangent sigmoid and linear transfer functions at hidden and output layers respectively with 8 neurons and the maximum sorption capacity was 0.9 mg/g. The sensitivity analysis signified that the relative importance of contact time equal to 36.9% and it is the influential parameter in the sorption of ammonia nitrogen. However, the relative importance of other parameters was agitation speed of 27.43%, WFS dosage of 17.32%, pH of 9.86% and initial concentration of 9.39%.

Keywords—.

1. Introduction

Nitrogen can be risky in high concentrations while it is a fundamental nutrient for all living organisms. The nitrogen is considered one of the mainly important pollutants in water resources [7]. An inorganic ion form of nitrogen impurity is "ammonium nitrogen (NH3-N)" where its presence in high concentrations will enhance the growth of algae and plants. This will cause a reduction in the dissolved oxygen of the aqueous media and increasing the botulism of aquatic organisms [25]. The pollution with ammonium has several hazardous effects such as; mortal, water resources demolition, malady, increased corrosion rate of soil materials, and, in the aquatic life, convulsion, coma as well as even death [7] [28].

The NH3-N can be removed from wastewater by different techniques where the biological treatment technique (nitrification–denitrification) is the traditional method. However, this method reveals that the achieved removal efficiency is low in the high concentrations of NH3-N [39]. Accordingly, ion exchange or adsorption can be adopted as an effective and efficient technique for remediation of water contaminated with this pollutant [33]. Fertilizer factories, coke plant, farming activities, refrigeration systems, metal-finishing industries and sewage treatment plants are represented the frequently sources of NH3-N [33] [22] [19] [14].

For removal of NH3-N, several types of adsorbents were investigated such as zeolite [33] [29] [6] [20] [39] and granular activated carbon [5] [17] [30] [24] [36] [23]. Many attempts were directed to investigate the ability of using other types of adsorbents with lower cost and higher sorption capacity. In this concern, bentonite, clinoptiloliten and sepiolite were investigated [34] [8] [7] [25] [31] [17].

Waste foundry sand (WFS) is a waste produced from the manufacture of both non-ferrous and ferrous metal castings. Foundry activities are depended on the using of silica sand in the moulding and casting where this sand can be reused for many times. The recycled sand will reach to the point that it cannot be reused. The unsuitable sand usually is landfilled at significant cost. In Iraq, Nasr Company for Mechanical Industries produced 10 tons of WFS per 8 hours when worked with full capacity and this means that large quantities of this waste are banished to the environment. Thus, the reuse of this material would save valuable landfill space and reduced the cost of disposal. The chemical composition of this sand is consisted of 93–99% silica and 1–3% chemical binder [32]. Lee and Benson (2002) studied the potential use of WFS as an inexpensive medium for PRBs. Batch and column tests were conducted to evaluate the reactivity and sorption capacity of twelve foundry sands for four groundwater contaminants: Trichloroethylene (TCE), the herbicides alachlor and metolachlor, and zinc [21]. Faisal and Ahmed (2014) were studied the possibility of using waste foundry sand (WFS) as PRB in treating of groundwater contaminated with copper [12]. Artificial neural networks (ANNs) have been used to investigate the nature of relationship related between the input and output variables. This can be achieved through extracting the controlling features from input database [35] [38] [26]. Accordingly, the feasibility of using WFS in the treatment of water contaminated with ammonia nitrogen depended on batch experimental results and ANN model is the focal point of the present study.

2. Definition of the ANN Model

Natural network of biological neurons is represented the working principle of ANN model. The neuron (or node) is the basic processing element of a neural network. The neuron impulse or the output of a node is calculated as weighted sum of the input signals from the proceeding neuron, altered by the transfer function. The learning capability of a neuron is accomplished by adjusting the weights in conformity to chosen learning algorithm. Input, hidden and output layers are represented the basic elements of ANN structure. The number of input and output variables is the principle parameter that specified the number of neurons in the hidden layer. The specifying of this number is an important step and it is can be selected by trial and error where root mean squared error has the lowest value [26].

3. Experimental Work

3.1 *Materials*

The WFS was brought from Nasr Company for Mechanical Industries/ Iraq and its characteristics are inserted in **Table 1**. The particle size distribution of this sand ranged from 75 μm to 1 mm with effective and median particle sizes of 180 and 320 μm respectively.

Water sample contaminated with 1000 mg/L of NH_3-N was prepared by dissolving a 3.819 g of anhydrous ammonium chloride (NH_4Cl, manufactured by E. MERCK, Denmark) in one liter of deionized water. The solution was kept at room temperature and it is used to prepare the required concentration where the pH was controlled by HNO_3 or NaOH with concentration of 0.1 M.

Table1: Characteristics of WFS used in the present study.

Property	**Value**
SiO_2	94.36
Al_2O_3	2.82
Fe_2O_3	2.12
Na_2O	0.24
CaO	0.05
TiO_2	0.14
MgO	0.23
K_2O	0.039
Bulk density (g/cm^3)	1.44
Porosity	0.46
Hydrulic conductivity (cm/s)	1×10^{-6}
Surface area (m^2/g)	5.935
Cation exchange capacity (meq/100 g)	10.94

3.2 *Batch Tests*

The effects of contact time, initial pH, NH_3-N concentration, WFS dosage and agitation speed on the sorption performance of NH_3-N ion onto WFS material were studied by a set of batch tests. Sorbent dosages with values ranged from 20 to 120 g were introduced into 250 mL flasks containing of 100 mL of aqueous solution with different concentration of NH_3-N range from 400 to 600 mg/L. The flasks were agitated using an orbital shaker (Edmund Buhler SM25, German) with contact time of 120 min and agitation speed of 200 rpm. Samples were withdrawn at specified intervals ranged from 5 to 120 min and, then, the supernatant was separated by filtration. The NH_3-N concentrations in the supernatant with volume of 5mL were measured by distillation and titration method where used KJELTEC AUC 1030 Analyzer [17][14].

Batch tests were carried out with pH ranged from 3 to10where the striping of ammonia nitrogen is occurred at pH greater than 10 [5].

Removal efficiency of NH_3-N (R) was chosen as the output parameter of the ANN model and it is determined as follows:

$$R = \frac{(C_o - C_e)}{C_o} \times 100 \quad (2)$$

Where C_o and C_e are the initial and equilibrium concentrations of NH_3-N respectively.

4. Results and Discussion

4.1 *ANN Model Development*

Simulation the removal efficiency of NH_3-N from aqueous solution by sorption technique was developed using ANN model with Levenberg–Marquardt back propagation (LMA) training algorithm. The algorithm was written by Matlab program version 7.9 (R2009b). Structure of ANN is represented by specifying the number of layers and the number of nodes in each layer as well as the nature of the transfer functions. Optimization was done by dividing the original experimental data into training, validation and test subsets. The biggest set is represented by the training data and it is very important for specifying the trend of the data by updating the weights of the network. The quality of the network was evaluated through the testing data. The validation set, which is the final check, is reflected the ability of the trained network to describe the data. A linear transfer function (purelin) at output layer and tangent sigmoid transfer function (tansig) at hidden layer were adopted in the present model.

Contact time (min), initial pH, dosage of WFS (g/100 mL), NH_3-N concentration (mg/L) and agitation speed (rpm) are chosen as the input variables and the removal efficiency is the output variable. For optimization of the network, mean square error (MSE) is plotted as function of the neurons number as shown in **Fig.1**. It is clear that the 8 hidden neurons must be chosen because it corresponds to minimum MSE. The training was stopped after 4 epochs for the LMA because there is an increase in the differences between the validation and training errors (**Fig.2**). However, the optimum topology of ANN for removal of NH_3-N from aqueous solution was found to be 5:8:1 as shown in **Fig. 3** and the best regression for LMA training, validation and testing algorithm was plotted in **Fig.4** where the correlation coefficient for these data was greater than 0.99328.

4.2 *WFS Dosage and Initial Concentration*

Experimental results (**Fig.5**) elucidated that the removal efficiency of NH_3-N was increased from 43 to 95 % due to change of WFS mass from 20 to 90 g for initial concentration of 400 mg/L. This may be due to the presence of larger number of binding sites in the high dosage of sorbent [3]. A further increase in the WFS mass did not lead to any significant change in the removal efficiency due to reaching the sorption capacity. This figure certifies that increasing of initial concentration lead to decrease of contaminant uptake. In addition, the removal efficiency of NH_3-N decreased due to increase of the initial concentration of NH_3-N. A good matching between ANN predictions and experimental results can be recognized with correlation coefficient greater than 0.989.

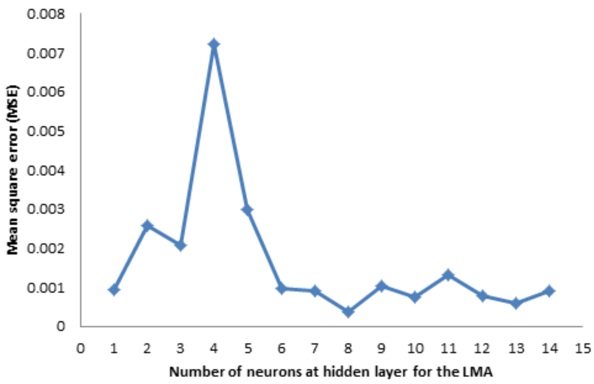

Figure 1: MSE as a function of neurons number at hidden layer for the LMA algorithm

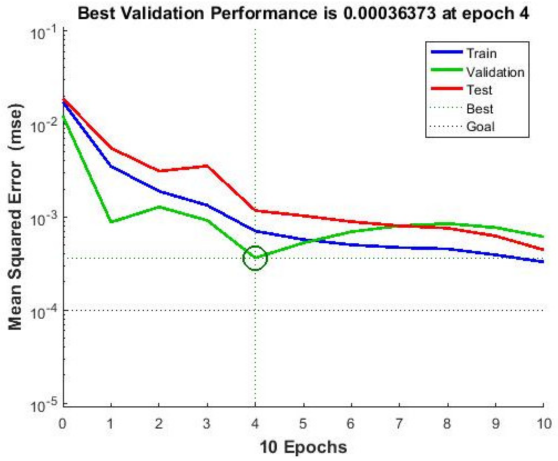

Figure 2: MSE for LMA training, validation and testing algorithm

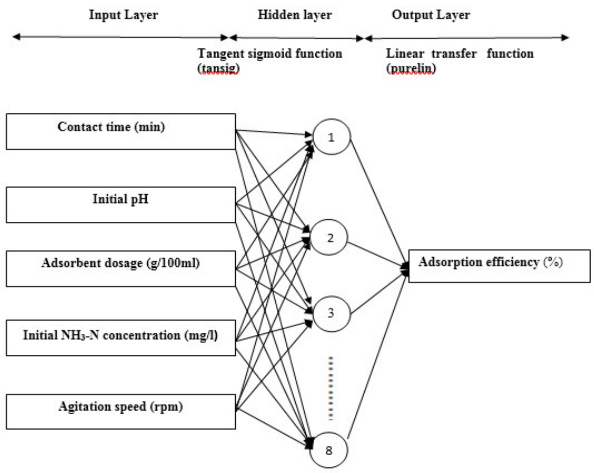

Figure 3: The optimal architecture of ANN

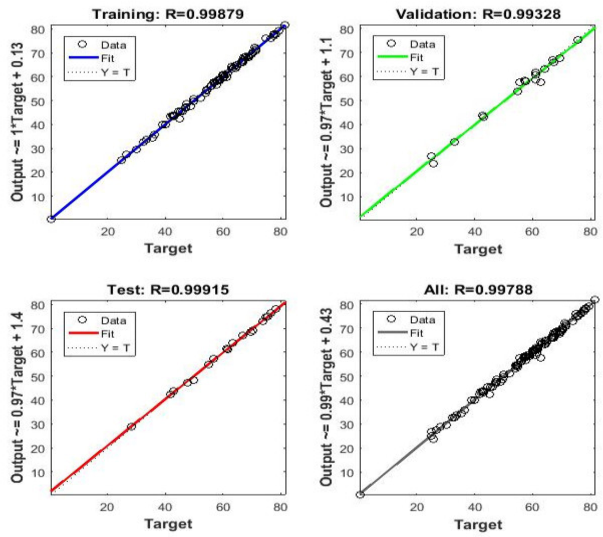

Figure 4: Training, validation and testing regression for the Levenberg-Marquardt algorithm

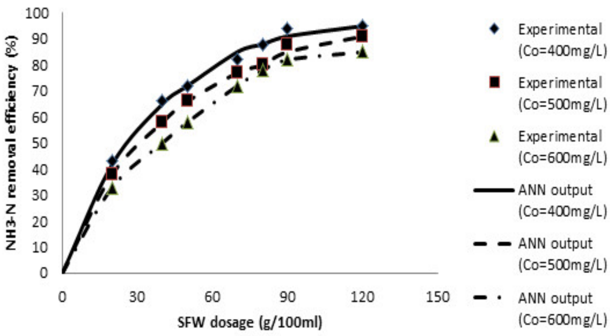

Figure 5: Experimental results and ANN outputs for removal efficiencies of NH_3-N for different values of WFS dosage and initial concentration (initial pH =8, contact time = 120 min and agitation speed = 200 rpm

4.3 *Contact Time and Initial pH of Solution*

The data obtained from the sorption of NH_3-N onto WFS signified that the time required to achieve the equilibrium is equal to 90 min and the sorption approximately stabilized with increasing of contact time (**Fig.6**). In the initial stages of time, the removal efficiency was increased rapidly and, then, remained constant with time until equilibrium time. This presence of large number of sorbent sites may be the cause for this behaviour [10] [13]. In spite that the equilibrium state can be achieved at time approximately of 90 min, all experiments are conducted with equilibrium time of 120 min for practical reasons and this consistent with Alkan et al. (2008) [2]. Increasing pH from 3to10 (**Fig.6**) will lead to the increase of the sorption to NH_4^+ by reducing the competition of H^+ and NH_4^+ on the sorption site. On the other hand, the ammonia nitrogen in solution is in two forms: NH_4^+ and NH_3. Under the neutral or acidic conditions, NH_4^+ is the main form of existence. When the pH value is high, NH_4^+ changes into NH_3, which tend to be transformed to ammonia gas according to the following equations [9] [11]:

$$NH_4^+ + OH^- \leftrightarrow NH_3 + H_2O \quad (2)$$

The correlation coefficient between the experimental data and ANN predictions is greater than 0.99 and this means that there is a good agreement between them.

4.4 *Agitation Speed*

The uptake of NH_3-N was increased with the increase of agitation speed as illustrated in **Fig.7**. It is clear that the uptake changed from 50 to 78% when the speed increased from 50 to 250 rpm at dosage of 90g/100mL. Improving the diffusion of contaminant towards the WFS (i.e. good contact with binding sites) can be the cause for this increment [4]. The same trend can be recognized for other values of dosage but with lower values of removal efficiency. This figure certifies that the agitation speed with value of 200 rpm is sufficient to achieve the required equilibrium. Also, **Fig.7** certifies that ANN model is a good tool for description of experimental results with correlation coefficient not less than 0.995.

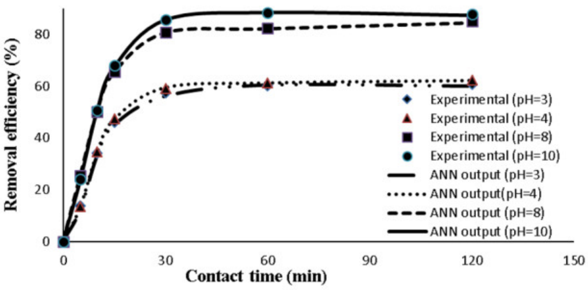

Figure 6: Agreement between ANN outputs and experimental data as a function of contact time for different values of initial pH (C_o = 600 mg/L, WFS dosage = 90 g/ 100 mL and agitation speed = 200 rpm)

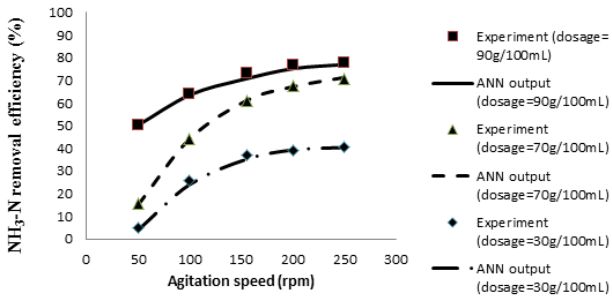

Figure 7: Experimental results and ANN outputs for removal efficiencies of NH_3-N for different values of agitation speed and WFS dosage (initial pH=9, C_o= 679 mg/L and contact time= 120 min)

4.5 *Sensitivity Analysis*

Garson (1991) equation illustrated below can be used to assess the relative importance (*I*) for each input parameter (*j*) depended on neural net weight (*W*) matrix [1]:

$$I_j = \frac{\sum_{m=1}^{m=Nh}\left(\left(\frac{|W_{jm}^{ih}|}{\sum_{k=1}^{Ni}|W_{km}^{ih}|}\right)\times|W_{mn}^{ho}|\right)}{\sum_{k=1}^{k=Ni}\left\{\sum_{m=1}^{m=Nh}\left(\frac{|W_{km}^{ih}|}{\sum_{k=1}^{Ni}|W_{km}^{ih}|}\right)\times|W_{mn}^{ho}|\right\}} \quad (3)$$

where Ni and Nh are the numbers of input and hidden neurons, respectively, the superscripts *i* (input), *h* (hidden) and *o* (output) layers, respectively. Subscripts k, *m* and *n* refer to input, hidden and output neurons, respectively. **Fig.8** presents the relative importance for each input variable and it is clear that the contact time is the most influential parameter in the sorption of NH_3-N onto SWF. However, many researchers proved that the influential variable and effect of each variable depended upon the experimental ranges adopted in the fitting model [37].

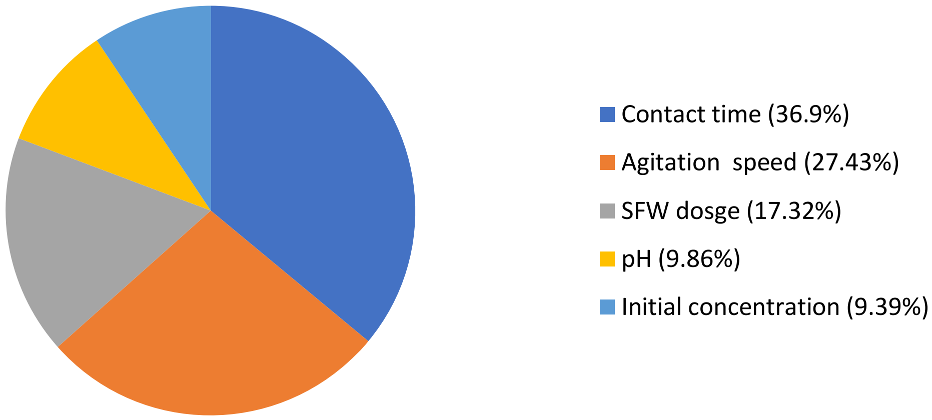

Figure 8: Sensitivity analysis using artificial neural network and Garson equation.

4.6 *Sorption Properties*

The sorption data were characterized by ANN model (**Fig.9**) which is related by the amount of NH_3-N removed from the aqueous solution (qe, mg/g) and the equilibrium concentration remaining in this solution (Ce, mg/L) at constant temperature [27]. It can be concluded that the amount of NH3-N removed per unit mass of WFS at equilibrium increased rapidly at the low initial concentration of contaminant and it begins slightly increasing with increase of concentration. This figure certifies that there is a satisfactory matching between the predicted and measured values with correlation coefficient not less than 0.98. This means that the ANN model is a dependable tool to describe the amount of pollutant sorbed onto reactive material through its transport. In addition, the calculated maximum sorption capacity of the NH_3-N onto WFS was equal to 0.9.

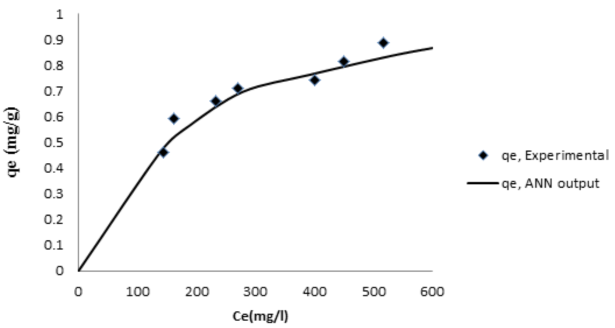

Figure 9: Predicted and measured values of q_e for sorption of NH_3-N onto WFS

5. Conclusions

Batch experiments proved that the suitable parameters for sorption of NH_3-N from contaminated water onto WFS are initial pH of 10, WFS dosage of 90g/100mL, contact time of 120 min and agitation speed of 200 rpm when the initial concentration of 400 mg/L. Experimental results signified that the removal efficiency of NH_3-N could be effectively improved up to 95 % using WFS byproduct and the calculated maximum sorption capacity of 0.9 mg/g.

Results elucidated that the linear and tangent sigmoid transfer functions at the output and hidden layers respectively are able to predict the removal efficiency of NH_3-N where the best hidden number of neurons for the LMA was 8 with MSE of $3.63x10^{-4}$. These results proved that the ANN model is able to describe the measured results with correlation coefficient not less than 0.98 for five operating parameters. The contact time has the greater value of relative importance (= 36.9%) and this means it is the most influential variable in the removal of NH_3-N by WFS.

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الخلاصة – الدراسة الحالية تتحرى كفاءة إزالة الأمونيا نتروجين من مياه الصرف الصحي المحاكاة بواسطة مخلفات رمل المسبك اعتمادا على 120 تجربة من نوع الدفعة تم نمذجتها بواسطة تقنية الشبكات العصبية الاصطناعية ذو الثلاث طبقات. ان العوامل التي تم دراستها هي زمن التماس (5-120 دقيقة)، الدالة الحامضية الابتدائية للمحلول المائي (3-10)،التركيز الابتدائي (400-600 ملغم/ لتر)، المادة المازه (20-120 غم/ 100 مل) واخيرا سرعة الاهتزاز (50-250 دوره/دقيقه). أثبتت النتائج أن أفضل القيم لهذه العوامل كانت 90 دقيقة و 10 و 400 ملغم / لتر و 90 غم / 100 مل و 200 دورة في الدقيقة على التوالي حيث بلغت كفاءة الإزالة المتحققة 95٪. ان النموذج المعتمد اثبت قدرته على تمثيل عملية الامتزازاعتماد على استخدام دالة النقل المماسية في الطبقة المخفية ودالة النقل الخطية في الطبقة الناتجة وبواقع ثمانية خلايا عصبية حيث ان قدرة الامتزاز العظمى هي 0,9 ملغم/غم. وأظهر تحليل الحساسية أن زمن التماس هو الأكثر تأثيرا في إزالة النتروجين الأمونيا وبنسبة 36.9٪، تليها سرعة الاهتزاز، كمية المادة الممتزة التركيز و ثم الدالة الحامضية.

الكلمات الرئيسية –.