

# Modeling the Organic Pollutants Removal from Refinery Wastewaters Using the Activated Carbon Adsorbent in the Neural Network through Experimental Data

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Received: 18 November 2017 / Received in revised form: 30 April 2018, Accepted: 03 May 2018, Published online: 05 September 2018  
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## Abstract

**Background:** Oil pollution is one of the serious environmental problems that can cause irreparable environmental damages. Petrochemicals are almost unavoidable. In this study, the results of phenol removal by activated carbon adsorbent using perceptron neural networks with the variable number of neurons -1 to 20 neurons- were estimated and predicted. In this network, the data of initial concentrations like time, adsorbent weight, pH and temperature being reported in the background section of the study were selected as input variables, while the phenol removal efficiency values were considered as the network's outputs. **Methodology:** The laboratory collected 79 data from different sources. In order to develop and validate this extended model, the entire database was randomly divided into three types: 70% (55 data points), 15% (12 data points) and 15% (12 data points) were respectively used as training, validation, and test. In order to improve the performance of the artificial neural network, input values and target values were normalized in the range of -1 to 1. **Results:** The results of the prediction showed that the developed artificial neural network model provides an accurate prediction of the removal efficiency values with a total correlation coefficient of 0.8986 and a total squared error of 0.0002788. **Conclusion:** The study of the ability of the artificially trained neural network to predict the effect of input variables in the artificial neural network on the removal efficiency values confirmed the good performance of the developed artificial neural network model.

**Keywords:** Removal Efficiency, Neural Network, Activated Carbon Adsorbent

## Introduction

To remove oil from oily wastewaters, existing oil droplets should be isolated. Among the existing techniques, physical separation technique is the easiest way. This technique relies on the difference in the physical properties of oil and water (including density difference).

Manual collecting of oil pollution from the surface of the water, encircling the oil pollution by physical means, using natural and artificial absorbers, firing, using bipolar solvents, biodegradation all have advantages and limitations. Among the processes used to remove oil pollutants, the use of absorbent materials is considered to be the most cost-effective and economical way to eliminate oil pollution.

One of the most widely used adsorbents in this field is activated carbon. One of the major and important technologies for removal of low concentrations of chemical pollutants in industries is the use of adsorption phenomena in activated carbon substrates. Today, this technology replaces many of the isolation methods.

Activated carbon is a form of carbon that has mostly microspores on its surface and has not reacted to any substance. This material is produced by pyrolysis of carbon-based plant materials and is used for activation. In this study, active carbon derived from coal ash in the laboratory, as a natural absorbent, was used to remove oil pollution from aqueous solution.

One of the most polluting materials is copper. From copper metals to Bismuth metals are listed as heavy metals in a periodic table with densities greater than 5 gr/cm. In the periodic table, metals from group 3 to 16 in the 4th periodicity are heavy metals. (Choi and et al.,

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2004) The materials used in agricultural activities are another major decentralized source of metal contaminants-heavy metals and organic pollutants in agricultural pesticides such as arsenic, manganese, and copper (Choi and et al, 2004).

Copper does not react with water, but it can react slowly with air oxygen and form a layer of brownish-black copper oxide. In addition to being in various mineral gems, copper is also found in phase (Olanrewaju and et al., 2014; Principe, 2014; Demuth and et al., 1994)

Acute symptoms of copper poisoning associated with psychology include autism symptoms (such as depression, hallucinations, insomnia, paranoia, personality changes, and insanity) and schizophrenia symptoms (such as high irritability, lack of awareness and understanding of the senses, and time).

When the concentration of manganese exceeds its usual limit, it is very poisonous for the human body. If anybody does not consume an adequate amount of manganese of the body, his health will be at risk. But if manganese consumption is high, problems occur in the human body. Manganese compounds may cause a tumor in the human body, but laboratory research has led to a vague result, and yet this effect of manganese has not been properly verified.

Another pollutant is the fly ash, which is melted during the coal combustion. These materials become solid while they are suspended in the exhaust gas from the furnace (very fine solid particles) and are collected from exhaust gases by electrostatic precipitators. In general, it can be said that about 5-20% of the total mass of coal remains in the form of ash after burning. Depending on the type of coal and the way in which it is burned, the fly ash compositions are different, but in general, it can be said that the fly ash contains a significant amount of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>. Mineral resources in coal indicate the elemental composition of the fly ash, but the crystalline and mineralogy structure of the fly ash is determined by designing boiler and the mode of operation.

Another pollutant is zeolites. Natural zeolites usually have a negative charge on their surface and therefore they are mostly cation exchangers. Zeolites have catalytic properties, and their most usage is in refinery processes such as in hydro cracking processes, enhancement of gasoline, octane number and aromatization. Natural and synthetic zeolites applications are a function of their physical and chemical properties, which depend on the crystal structure and chemical composition of zeolite. In terms of the phenomena such as penetration, molecular absorption, molecular sieve, the behavior of zeolites is influenced by the anionic network topology, as well as the size of the load and the location of interchangeable cations within their framework.

One of the methods useful for removing heavy metals is using physical methods. At first, filtration and sedimentation processes are performed to remove the precipitated metals. Therefore, the active carbon and the adsorption process are further used to complete the purification process. Chemical methods for removing heavy metals and organic pollutants contain chemical deposition, oxidation-reduction, and ion exchanges.

The adsorption method can remove pollutants that cannot be eliminated by some other methods. Adsorbents can be organic or mineral; mineral adsorbents are superior to organic adsorbents in terms of electrical resistance and chemical stability, high specific surface area and stability against microbial decomposition. Compounds such as activated carbon, natural zeolites, bentonite, chitosan, corn, rice, barley, and dried activated sludge were used as adsorbents.

Juranova (2007) showed that by fixing nanoparticles onto a substrate, the dimensions of the nanoparticles did not change, and the aldehyde degradation increased with increasing irradiation time and the thickness of the stabilized film TiO<sub>2</sub>. Yong Joon (2006) stated an increase in photo-catalytic activity of reactors with fixed layers onto the substrate was influenced by the increasing number of layers and the number of nanoparticles fixed onto the substrate per unit area, also it was affected by increasing the radiation intensity and the duration of exposure to ultraviolet radiation.

Venkata (2003) has identified a suitable substrate in stabilized bed reactors as an effective factor in improving stabilization, activity, and adhesion of catalyst onto the substrate and homogenization of catalyst.

Since industrial tests are costly and not readily feasible to perform, in this research, neural network modeling will be used to predict the system output in circumstances which would not be considered in the test.

In this research, we try to model the removal efficiency of phenol with a maximum correlation coefficient (close to 1) and the minimum sum of squared errors (close to zero) in the neural network using the data extracted from phenol removal through the activated carbon adsorbent. As a result, a suitable model can be provided to predict the removal efficiency with the variables (initial gravity, time, adsorbent weight, pH, temperature).

## Materials and Methods

In this research, an artificial neural network is used to achieve the results. In this network, the data of initial concentrations, time, adsorbent weight, pH and temperature reported in the research background were selected as input variables, while the phenol removal efficiency values were considered as network output. The laboratory collected data consists of 79 data collected from different sources. In order to develop and validate this extended model, the entire database was randomly divided into three sections: 70% (55 data points), 15% (12 data points) and 15% (12 data points) data points were used as training, validation, and test, respectively. In order to improve the performance of the artificial neural network, the values of input and target data were normalized in the range of -1 to 1. The removal efficiency of organic pollutants was selected as the target of the network. The characteristics of activated carbon used in the work of others are generally shown in Table 1.

**Table 1-** Databanks used for this modeling

Year	Pollutant	Absorbent	Researchers
1387	Phenol	Activated carbon	Pajoooheshfar & Saeedi (2009)
2016	Phenol	Activated carbon	Ingole and et al., (2017)
2007	Phenol	Activated carbon	Mukherjee and et al., (2007)

**Table 2-** Characteristics of activated carbon

Activated carbon	Parameter
891.1745	Special surface (m <sup>2</sup> /g)
942.8134	Single pointspecial surface (m <sup>2</sup> /g)
2.1060	Density (g/cm <sup>3</sup> )
Insignificant	Humidity (%)
0.15-0.85	Particle size (mm)
0.487	Volume of closed pores (cm <sup>3</sup> /g)

In Table 3, the values and conditions for conducting the experiments are shown in different pH, contact time, initial concentration, adsorbent weight and different temperatures for each study.

**Table 3-** Values and conditions for carrying out tests in different conditions

Researchers	pH	Initial concentration (mg/L)	Temperature (° C)	Time (min)	Absorbent amount (g/liter)
Pajoooheshfar & Saeedi (2009)	9-2	100	environment	180-5	20-5
Ingole and et al., (2017)	12-2	200-50	50-10	120-0	50-10
Mukherjee and et al., (2007)	9-3	100-20	environment	360-60	60-10

The following relation can be used to calculate the pollutants removal efficiency conducted by the researchers:

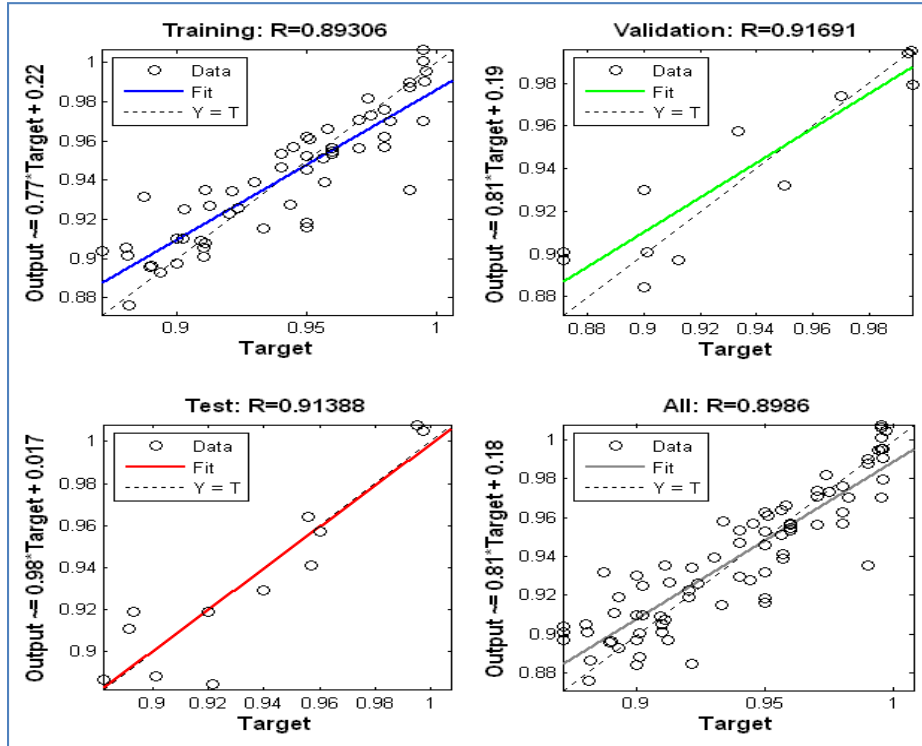
$$\text{removal efficiency in percentage} = \frac{C_0 - C_t}{C_0} \times 100$$

In which, C<sub>0</sub> and C<sub>t</sub> are the initial concentration of phenol in the solution and its concentration after testing, respectively.

In order to train the network, MATLAB and Multilayer Perceptron Network (MLP) were used. Back Propagation (BP) as a training method and the mentioned activation function was used for the training process. Also, for the number of hidden neurons, values of 1 to 2n +1 (n is the number of input neurons) were applied. Finally, the best structure of the network with the minimum mean squared error (MSE) and the maximum correlation coefficient (R) was selected.

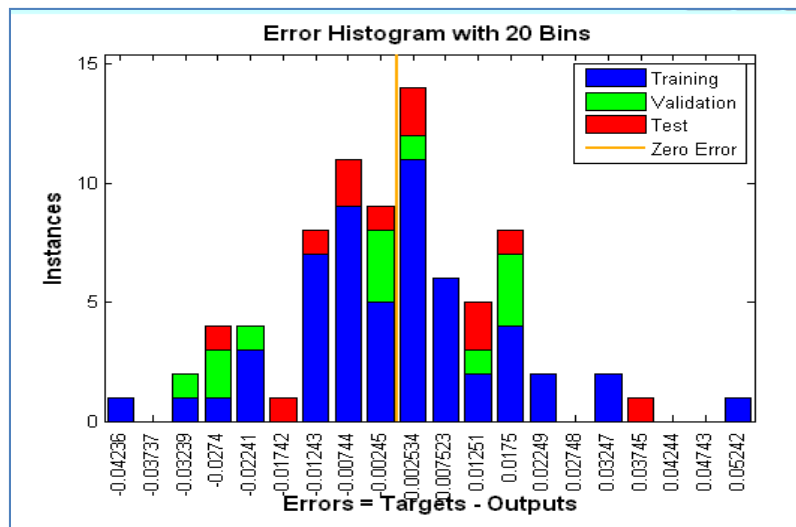
**Findings**

As shown in Fig. 1, this factor has been used for all three parts- training, validation and test, and its value was calculated by the network. Correlation coefficients for the total data were also obtained. These values were equal to 0.89306, 0.91691, and 0.91388 for three parts of the training, validation, and test, respectively which indicates the successful prediction of the network with the 20 neurons, but this value is 0.886 for the total data, which indicates a successful prediction.



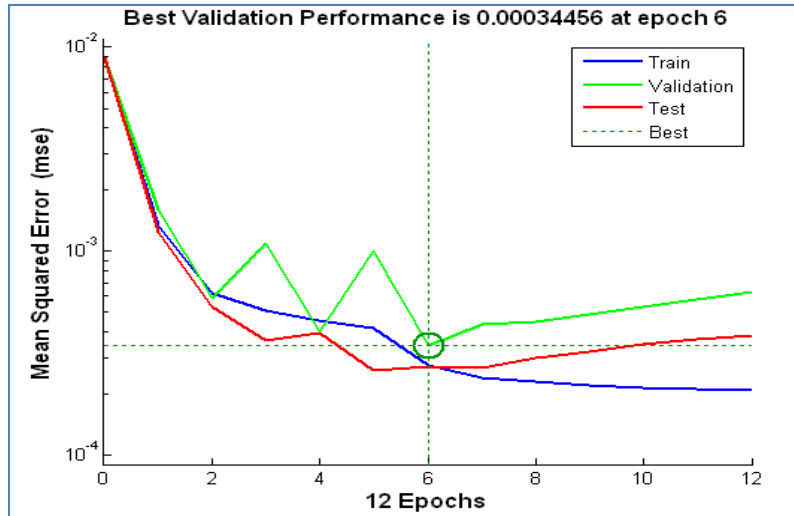
**Figure 1:** Correlation coefficients for the implemented neural network

As shown in Fig. 2, all data is close to the Zero Error line and the error distribution is very low. The maximum and minimum error values are 0.0423 and 0.0524.



**Fig 2:** The difference between the prediction results of the neural network and the experimental results of the experiments.

In Figure 4-5, the difference between the results of the prediction of the neural network and the experimental results of the experiments showed that the low value of this difference reflects the proper design of the neural network parameters.



**Figure 3 :**Changes in mean square error during training steps

As shown in Fig. 3, with the increase in the number of replications (12 replications), the best result of 0.00034456 was obtained by replication 6. In replication 6, the best results were achieved for all three parts of training, validation and test. With the increasing number of replications from 6 onwards, the results were not favorable and no successful prediction was achieved.

**Table 4- Results obtained with the different number of neurons**

Number of hidden layers	Neuron	the correlation coefficient			
		training	validation	test	All data
1	3	0.9025	0.8342	0.7449	0.8112
1	4	0.8215	0.7862	0.8536	0.7932
1	5	0.7845	0.8040	0.7541	0.7932
1	6	0.931	0.8042	0.7548	0.8317
1	7	0.8132	0.8495	0.8211	0.8327
1	8	0.8035	0.8217	0.8327	0.8135
1	9	0.9010	0.8576	0.7361	0.8845
1	10	0.7842	0.8094	0.8137	0.7985
1	11	0.7328	0.7964	0.8394	0.7541
1	12	0.8120	0.6472	0.8046	0.7726
1	13	0.8276	0.8072	0.8175	0.8112
1	14	0.8534	0.7812	0.8264	0.8195
1	15	0.8674	0.8267	0.8095	0.8264
1	16	0.8456	0.8471	0.8875	0.8328
1	17	0.8097	0.8165	0.8964	0.8367
1	18	0.7763	0.8533	0.8665	0.8298
1	19	0.7982	0.8726	0.8315	0.8475
1	20	0.89306	0.91691	0.91388	0.8986

Table 4 presents the number of neurons used in network with different results. As stated, 20 neurons are more favorable than other modes.

## Conclusion

This study uses an artificial neural network to estimate the phenol removal efficiency with activated carbon as the adsorbent. An artificial neural network model was developed using a database with 79 points reported in the review of the literature. Initial concentrations, time, adsorbent weight, pH and temperature reported in the review of literature were selected as the inputs of the network, while the removal efficiency values were considered as the output of the network. To obtain the best topology, various analyses were performed on different MPL network topologies. Finally, an artificial neural network model with neuron structures 1-20-5 was obtained as the best topology for developing the model. It also indicated that the developed artificial neural network model provides an accurate prediction of the removal efficiency values with a total correlation coefficient of 0.8986 and a total square error of 0.0002788. Considering the ability of the trained artificial neural network to predict the effect of neural network input variables on the removal efficiency values, the developed artificial neural network model was confirmed. According to the results of this study, the proposed artificial neural network model showed excellent harmony with experimental data and could predict the removal efficiency with high precision.

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