

Phenol Removal by Meliaazedarach Seed Ash from Aqueous Solutions

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Abstract

Background: Pay attention to phenol is important because of its sustainability in the environment and creating health problems. Due to the high solubility of phenol in water, it must be removed in order to prevent contamination of water resources and the environment. The aim of this study is to assess the efficacy of M. azedarach seed ash for removal of phenol from aqueous solutions.

Methods: In this experimental study, M. azedarach seed ash was used for removal of phenol from aqueous solutions. In this study, the effect of the factors affecting adsorption was studied, including adsorbent dosage, initial phenol concentration, contact time, pH and temperature. Data obtained from the tests were analyzed using Excel software. Langmuir and Freundlich isotherm models were employed to verify compliance of absorption process from the isotherm models.

Results: Experimental results indicated that the efficacy of M. azedarach seed ash for removal of phenol had a direct relationship with increasing adsorbent dosage, temperature and contact time, as well as an inverse relationship with increasing pH and initial phenol concentration. According to the correlation coefficient of adsorption isotherm models, adsorption of phenol followed the Freundlich model.

Conclusions: The results of the present study showed that M. azedarach seed ash is effective for removal of phenol from aqueous solution. Given the ability to grow of M. azedarach tree in hot and dry climates, it can be used as an inexpensive and accessible adsorbent.

Keywords: Phenol; Adsorption; Meliaazedarach Seed; Isotherm

1. Introduction

Phenol is a toxic polycyclic aromatic hydrocarbon with a molecular weight of 94.11 g/mol and a benzene derivative that is colorless or white solid in its pure form with very high solubility in water, and gives very low-acid property to water after dissolving (1-4). Phenolic compounds can cause complications such as intoxications, carcinogenesis and teratogenesis in human beings and other organisms, which are the most common water pollutants (5). Drinking water contaminated with phenol even in small amounts can cause toxicity, health, taste and odor problems (6). Adverse health effects caused by exposure to phenol depending on the extent of absorption and contact time are ranging from skin irritation and burns to systemic toxicity along with decreased blood pressure, increased heart rate and coma (7). Phenol is used in industry as a precursor for

the production of various compounds (8). The United States Environmental Protection Agency (EPA) has warned about poor control of phenolic compounds in the environment (9). The maximum allowable concentration of phenol discharge into surface water is less than 1 mg/L and into drinking water is less than 0.5 µg/L (10). Hence, it is one of the priority pollutants (11, 12). Phenol contamination with concentration of about 0.5 mg/l gives water unpleasant taste and odor (13). Many different methods have been used for treatment of wastewater containing phenol, including advanced chemical and photochemical oxidation, deposition, ion exchange, electrochemical methods, steam distillation, irradiation, photocatalytic degradation, enzymatic removal and adsorption (14). Among the physicochemical processes,

adsorption technology is widely used (15). Studies have shown that activated carbon is effective in absorbing many refractory organic pollutants from aqueous solutions. The EPA has recommended absorption by activated carbon as one of the best technologies available in the removal of organic compounds, but its application because of high cost and reactivation problems is limited in developing and low-income countries. This has encouraged researchers to find inexpensive and locally available adsorbent to replace the activated carbon for removal of organic compounds such as phenol (16, 17). In this study, *M. azedarach* seed ash was used as an affordable and accessible alternative to adsorbents. *M. azedarach* tree has ability to grow in most parts of country and numerous seeds are produced as *M. azedarach* tree fruits that are unused and are regarded as the residue of the tree. The genus *Melia* L. belongs to family *Meliaceae*, which has several tree species. More than 15 species of this genus have been reported in the world and two species in Iran; *M. azedarach* species grows mostly in northern Iran. Several applications such as the use of *M. azedarach* fruit extraction as pesticides have been proven in recent studies (18, 19). The present study was conducted to evaluate the efficacy of *M. azedarach* seed ash as a natural adsorbent for removal of phenol from aqueous solutions, to assess the effect of various environmental factors such as adsorbent dosage, initial phenol concentration, contact time, pH and temperature on the absorption capacity and eventually to check the adsorption isotherm models.

2. Methods

In this lab scale study, at first in order to prepare adsorbent, after providing *M. azedarach* that is of agricultural waste and the seeds were separated and washed several times with distilled water. Then the washed seeds were placed in an oven at 70°C for 24 hours and after that were placed in an electric furnace at 600°C for one hour. The obtained ash was ground and sieved using 20-mm mesh sieve. In order to produce synthetic aqueous solution, phenol with the molecular weight of 94.11 g/mol and purity of 99.9% was prepared from the Merck Company and double distilled water was used. All tests were performed according to standard methods base on 5530 reference method of standard methods for the examination of water and wastewater (20). The pH of solutions was adjusted

using 0.01M NaOH and H₂SO₄. The remaining phenol concentration in the samples was measured in accordance with standard methods using UV/VIS (UNICO Model 2100) at a wavelength of 500 nm. This study examined the effects of adsorbate (phenol) at the concentrations of 20, 50, 100, 150 and 200 mg/l, adsorbent dosage (*M. azedarach* seed ash) at the doses of 0.5, 1, 3 and 5 g/l, pH values of 2, 4, 6, 8 and 10, temperature at 20, 30, 40 and 50°C, and contact time (0-60 min) on removal of phenol by *M. azedarach* seed ash. Shaking incubator at 180 rpm was used to mix and adjust the temperature of the samples. At each step, after the end of the contact time, adsorbent was completely separated from the aqueous solution containing phenol using 0.45-μm Whatman filter paper. Then, remaining phenol concentration in the filtered solution was read using the spectrophotometer. Data from the experiments were analyzed using Excel software. Langmuir and Freundlich isotherm models were employed to verify compliance of absorption process from the isotherm models.

3. Results

The phenol solution with a concentration of 100 mg/l was made to determine the effect of adsorbent dosage on the phenol removal efficiency. Then the effect of adsorbent (*M. azedarach* seed ash) at the doses of 0.5, 1, 3 and 5 g/l was investigated. Figure 1 shows that the phenol removal efficiency was enhanced with increasing adsorbent dosage whose reason can be attributed to an increase in the number of adsorption sites.

The effect of initial phenol concentration on the absorption process was evaluated at concentrations of 20, 50, 100, 150 and 200 mg/l and adsorbent dosage of 3g/l at the contact times of 5, 15, 30 and 60 minutes. Figure 2a indicates that phenol removal efficiency by *M. azedarach* seed ash is reduced by increasing the initial phenol concentration; the reason can be attributed to the limited free sites of the adsorbent in high concentrations of phenol. Figure 2b shows that the absorption capacity of adsorbent is elevated by increasing concentrations of phenol, probably due to greater access of adsorbent to adsorbate. The figures also demonstrated that the highest phenol removal occurred within the first 15 minutes, and after this phase, the phenol removal efficiency was not significant.

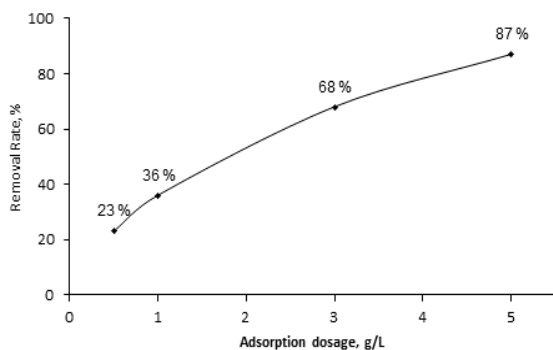


Figure 1: Effect of adsorbent dosage on phenol removal efficiency (phenol concentration of 100 mg/l and contact time of 60 minutes)

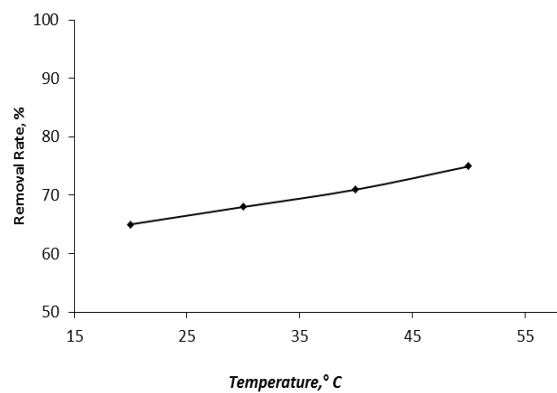


Figure 3: Effect of temperature on phenol removal efficiency (phenol concentration of 100 mg/l, adsorbent dosage of 3 g/l, contact time of 60 minutes)

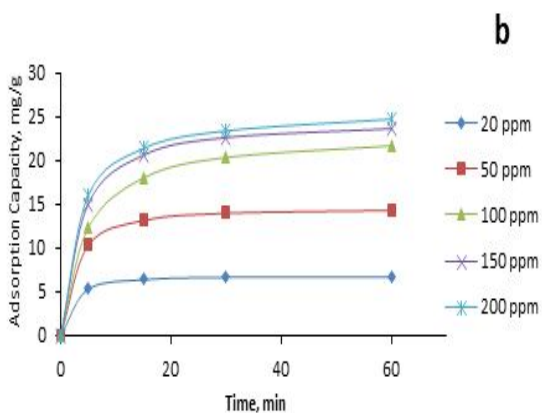
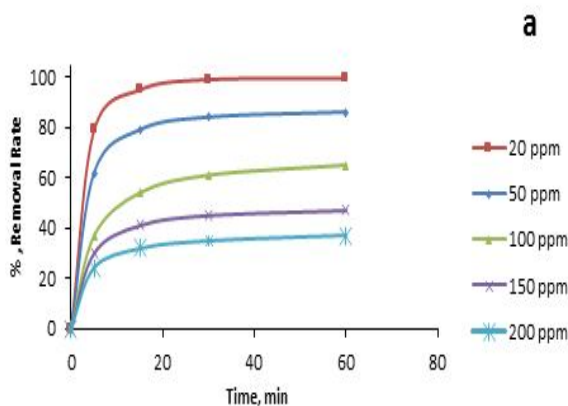


Figure 2:a: Effect of initial concentration of adsorbate on phenol removal efficiency, b: Adsorption capacity of adsorbent in various concentrations of phenol (adsorbent dosage of 3 g/l)

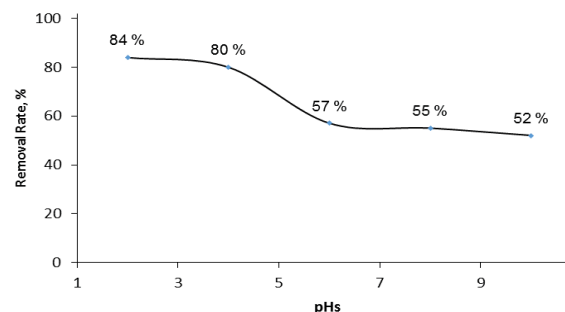


Figure 4: Effect of pH on phenol removal efficiency (Phenol concentration of 100 mg/l, adsorbent dosage of 3 g/l, contact time of 60 minutes)

The phenol removal efficiency with initial concentration of 100 mg/l and adsorbent dosage of 3 g/l was investigated at temperatures of 20, 30, 40 and 50°C. Figure 3 reveals that the phenol removal efficiency by *M. azedarach* seed ash increases with increasing temperature because the increase in temperature enhances the number of collisions between particles and the surface of adsorbent, increasing the absorption rate.

The effect of pH on phenol removal efficiency by *M. azedarach* seed ash was investigated in the range of 2 to 10. As shown in Figure 4, removal efficiency decreases with increasing pH, probably due to increased OH⁻ ions and competitive effects of phenol anions. The greatest level of phenol removal can be seen at pH=2.

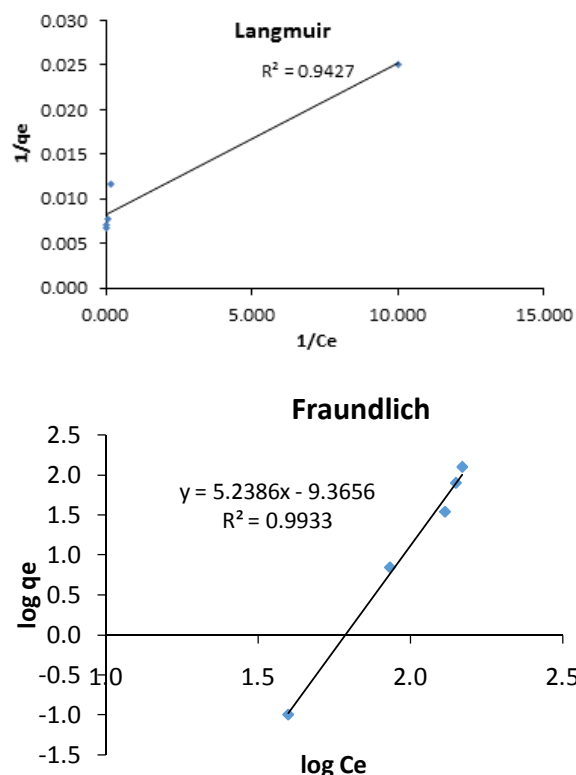


Figure 5: Langmuir and Freundlich isotherm models for phenol adsorption process (adsorbent dosage of 3g/l, phenol concentration of 100 mg/l)

The isotherms of Langmuir and Freundlich of Phenol adsorption illustrate in figure 5 at the condition of ash made at 600 °C, time 60 min, initial concentration of phenol was 100 mg/L, adsorbate dosage 3 g/L and pH 2. The result showed that phenol adsorption on the Meliaazedarach seed ash seeds is following by Freundlich model ($R=0.99$) rather than the Langmuir model ($R=0.94$).

4. Discussion

The effect of adsorbent dosage:

The results showed that removal efficiency has improved from 23 percent to 87 percent by increasing the adsorbent dosage from 0.5 mg/l to 5 mg/l. The reason for the increase in phenol removal is increased adsorbent dosage due to the increase in the number of sites available for phenol adsorption (21). The phenol removal efficiency in adsorbent dosage of 3 g/l has reached to 68%. Since phenol removal efficiency by increasing adsorbent dosage from 3 g/l to 5 g/l, had dramatic change, so adsorbent

dosage of 3 g/l was selected as the optimal dosage. Overlapping the adsorption sites on the surface of adsorbent is one of the reasons stated for the absence of phenol adsorption on activated carbon produced from *M. azedarach* seed for adsorbent doses more than 3g/l, resulting in decreased absorption sites and thus reduced absorption efficiency (22). Kilic et al. in a study for the removal of phenol from aqueous solutions on activated carbon produced by tobacco residues reported that the lack of elevation in absorption by increasing adsorbent dosage is for this reason that higher concentration of adsorbent causes increased competition of phenol anion to achieve surface active sites and therefore enhanced absorption density. Furthermore, higher doses of adsorbent per unit volume of water could lead to overlapping and accumulation of adsorbent surface. Hence, their outcome reduces the total available surface and absorption rate of pollutants because the accumulation of adsorbents increases the diffusion path during the emission of pollutants on absorbable surface of adsorbent, thereby reducing absorption rate (17). JafariMansourian et al. in a study on removal of phenol by *Acacia tortilis* bark reported the optimal adsorbent dosage of 0.2 g/l at the concentrations of 0.5, 1, 2, 4, 8, 16, 32, 64 mg/l (14). In a study by Fazelian et al., adsorbent dosage of 3g/l has been selected as the optimal dosage in the initial concentration of 40 mg/l, which is in compliance with the optimal dosage in the current study (23).

The effect of initial concentration on removal efficiency:

The results of assessing the effect of initial phenol concentration on the absorption process showed that the amount of phenol removal is a function of the initial concentration. With the increase in the initial phenol concentration, the removal efficiency is reduced because the number of active sites of absorption on the surface of the adsorbent for the phenol ion is reduced; this is due to completing the absorption capacity and lacking of its efficiency at higher concentrations (24). As Figure 2 illustrates, the absorption capacity of adsorbent was improved with increasing concentrations of phenol, while the phenol removal efficiency is decreased with increasing initial phenol concentration. The reason for promoted absorption capacity of adsorbent by increasing the initial phenol concentration can be attributed to increased likelihood of collision between adsorbent and adsorbate (25). The maximum removal of phenol was obtained in the

concentration of 20 mg/l, but the absorption capacity of adsorbent in this concentration of phenol is about 6.6 mg/g. By concurrent examining the phenol removal efficiency and the absorption capacity, it can be said that there are favorable conditions in the concentration of 100mg/l; because in this concentration, phenol removal efficiency is about 66% and the phenol absorption capacity is 21.6mg/g. Ahmed et al. used date palm kernel to remove phenol; the removal efficiency was decreased with increasing the initial phenol concentration (26). In a study by Bazrafshan et al. who utilized pistachio husk ash as adsorbent to remove phenol, the removal efficiency was decreased with increasing the initial phenol concentration (27). In a study by Hashemi et al. who employed walnut hulls for the removal of phenol from aqueous solutions, similar results were obtained that are consistent with the results of this study (7).

The effect of pH on removal efficiency:

The pH value of adsorption solutions is one of the most important parameters affecting the adsorption efficiency (28). In this study, the adsorption efficiency was decreased with increasing pH value. As can be seen in Figure 4, the phenol removal efficiency by adsorbent *M. azedarach* seed ash at pH 2 is higher than in other values of pH. Phenol is a weak acid that is partially ionized (29). Therefore, degradation of phenol in solution is highly dependent on pH; the adsorbent surface is surrounded by carboxylic ions in the acid pH values. According to the anionic conditions of phenol (pKa 10), electrostatic attraction between adsorbent and pollutant is increased (23, 30). Fazelian et al. in a study on removal of phenol from aqueous solutions by activated carbon produced from poplar tree have attributed the effects of low pH on increasing the absorption of pollutants on the surface of the adsorbent to the relationship between pH and adsorbent charge (23). Gholizdeh et al. reported that the phenol removal efficiency with unmodified rice husk ash was decreased with increasing pH (31).

The effect of temperature:

Figure 3 shows the effect of temperature on the absorption process of phenol by activated carbon of *M. azedarach* seed. The results of assessing the effect of temperature in the present study indicate that the absorption rate is elevated with increasing temperature because the increase in temperature enhances the number of collisions between particles and the surface of adsorbent, increasing

the absorption rate. The results of assessing the effect of chemicals on the absorption capacity of rice husk ash for removal of phenol from aqueous solutions carried out by Allah Abadi et al. indicated that the absorption efficiency could increase with increasing temperature (32). In a study conducted by Chen et al. on wood rich in potassium for removal of phenol, the results showed that efficiency would enhance with rising temperature due to the endothermic feature of activated carbon, which is in line with the results of the current study (33).

The contact time:

Figure 2 shows the absorption occurs rapidly at first and then slows down. The rapid absorption in the first phase is due to the large motive force that is created because of the availability of extensive surface of adsorbent. By prolonging the contact time from 15 to 60 minutes, the increase in phenol removal efficiency was low. In other words, the greatest amount of phenol removal by studied adsorbent is in the first 15 minutes. In addition, the absorption process reached equilibrium after 240 minutes. The adsorbent efficiency in the first contact time of 15 minutes has increased due to the large number of absorption sites and a large difference in concentrations of adsorbate in the solution and on the surface of adsorbent; but over time, the adsorbent efficiency had very gentle slope because of a phenol layer on the surface of adsorbent. With time, it is difficult to occupy the remaining free surface sites because repulsion occurs between phenol molecules adsorbed on the adsorbent and the molecules in solution phase (17). Ghader et al. found that phenol adsorption on studied adsorbent reached equilibrium in 15 minutes (34). Kilic et al. by studying the adsorption of phenol by activated carbon reported that the equilibrium appears in the first 30 minutes when adsorption of phenol on the studied adsorbent (17). Daraee et al. found five hours for the equilibrium of phenol adsorption using the studied adsorbent (35). Fazelian et al. in the study on removal of phenol by activated carbon of poplar tree reported 35 minutes as the equilibrium time (23). Allah Abadi et al., in a study that examined the effect of chemicals on the absorption capacity of rice husk ash for removal of phenol from aqueous solutions, obtained the equilibrium time of 60 minutes (32). In a study conducted by Gholizdeh et al., the equilibrium time was 240 minutes, which corresponds with the results of this study (31).

Isotherm of phenol adsorption on activated carbon prepared from *M. azedarach* seed ash:

In previous studies related to the absorption of pollutants on different adsorbents, determining the isotherms of adsorption should be considered among the most important characteristics. The results of this study showed that the obtained findings follow the Freundlich isotherm ($R^2=0.993$), So the absorption occurred on the surface layer of ashes.

and the maximum absorption capacity of adsorbent for *M. azedarach* ash was achieved Based on the Langmuir isotherm calculations 40.65 mg/g. in table 1 achieved isotherm data are shown. The results of the current investigation are consistent with the studies of Bazrafshan et al. (27) and Rahmani et al. (36). However, some studies have followed the Langmuir model, but Langmuir equation is based on the assumption that the surface of adsorbent is homogeneous, for example the studies of Ahmed et al. (26), Hamed et al. (37) and Hashemi et al. (7). They know the reason for this issue owing to the homogenous distribution of active sites on the surface of activated carbon. The difference in reports could be due to differences in the type of used adsorbents or in the conditions of absorption tests.

Table1: Isotherm parameters in different temperatures

Temperature (C°)	Langmuir isotherm			Freundlich isotherm		
	qm	KL	R ²	kf	n	R ²
25	40.65	4.83	0.9427	20.51	5.27	0.9933

5. Conclusion

The results obtained from the present study suggest that activated carbon of *M. azedarach* seed ash has a good capability for removal of phenol. The pH plays an important role in the phenol absorption by *M. azedarach* seed ash. The adsorption of phenol is higher in the low pH values and this rate is reduced by increasing the pH values so that the removal efficiency is diminished from 83% at pH 2 to 50% at pH 10. Moreover, the results indicated that phenol removal efficiency is increased by rising adsorbent dosage, contact time and temperature. Therefore, the activated carbon of *M. azedarach* ash because of easy access and low cost has appropriate potential to absorb phenol in operational scale.

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