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EXPLORING THE PHENOL REMOVAL STRATEGIES; BY THE MEANS OF ADSORPTION

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ABSTRACT. Phenol and phenolic compounds formed from industrial activities mix with water resources through wastewater. Phenol and its derivatives mixed into water resources are environmental pollution factors that have attracted attention recently and also threaten human health when they reach drinking water. For these reasons, removing phenols found in high amounts in wastewater has gained importance. One of the methods suggested for phenol removal from aquatic environments is adsorption. This study aims to review the current literature on the adsorption process used as a phenol removal strategy from aqueous solutions. For this purpose, the results of different adsorbents used in the adsorption process were examined and the factors affecting phenol adsorption were expressed. Additionally, the results of isotherm and kinetic studies used to explain the adsorption mechanism for phenol were compiled. It is anticipated that the data presented in this study will constitute an important resource for designing systems for phenol treatment in the future.

Keywords: *adsorption, phenol, wastewater treatment*

FENOL GİDERME STRATEJİLERİNİN ARAŞTIRILMASI; ADSORPSİYON YOLUYLA

ÖZET. Endüstriyel faaliyetler sonucu oluşan fenol ve fenolik bileşikler atıksu yoluyla su kaynaklarına karışmaktadır. Su kaynaklarına karışan fenol ve türevleri son zamanlarda dikkat çeken ve içme suyuna ulaştığında insan sağlığını da tehdit eden çevre kirliliği faktörleridir. Bu nedenlerden dolayı atık sularda yüksek miktarda bulunan fenollerin giderimi önem kazanmıştır. Sucul ortamlardan fenol giderimi için önerilen yöntemlerden biri de adsorpsiyondur. Bu çalışma, sulu çözeltilerden fenol giderme stratejisi olarak kullanılan adsorpsiyon işlemine ilişkin mevcut literatürü gözden geçirmeyi amaçlamaktadır. Bu amaçla adsorpsiyon prosesinde kullanılan farklı adsorbanların sonuçları incelenmiş ve fenol adsorpsiyonunu etkileyen faktörler ifade edilmiştir. Ayrıca fenolün adsorpsiyon mekanizmasını açıklamak için kullanılan izoterm ve kinetik çalışmaların sonuçları derlenmiştir. Bu çalışmada sunulan verilerin gelecekte fenol arıtımına yönelik sistemlerin tasarlanması için önemli bir kaynak oluşturacağı öngörülmektedir.

Anahtar Kelimeler: *adsorpsiyon, fenol, atık su arıtımı*

INTRODUCTION

Phenol is the name given to organic compounds formed by bonding the OH- molecule group to the benzene ring. In other words, phenolic compounds are aromatic organic compounds in which one or more hydroxyl groups are attached to the aromatic ring (Fig. 1, [1]). Phenol is widely used in various industries as well as in the chemical industry for the production of other phenol derivatives such as alkylphenols, cresols, aniline, and resins [2]. In this context, phenolic compounds are found in the wastewater of various industries such as oil refining, petrochemical, pharmaceutical, coking processes, resin production, plastic, paint, pulp, paper, and wood products [3]. Phenol has been reported to be present in industrial wastewater at concentrations ranging from 50 to 2000 mg/L [4]. The toxicity level of phenol for humans and aquatic creatures has been reported to be in the range of 9-25 mg/L [5]. Phenol derivatives are less toxic than pure phenol, with lethal doses reported to be between 3 and 30 g [6]. Chronic effects of phenol exposure in humans may include loss of appetite, weight loss, diarrhea, dizziness, salivation, and dark coloration of urine, and it has been determined that chronic exposure to phenols irritates the gastrointestinal and central nervous systems and liver, kidney and cardiovascular tissues in animals [7]. The toxicity level of phenol for humans is between 10-24 mg/l, with a lethal blood concentration of approximately 150 mg/100 ml [8]. For this reason, industrial wastewater containing phenol must be released into the environment after treatment processes.

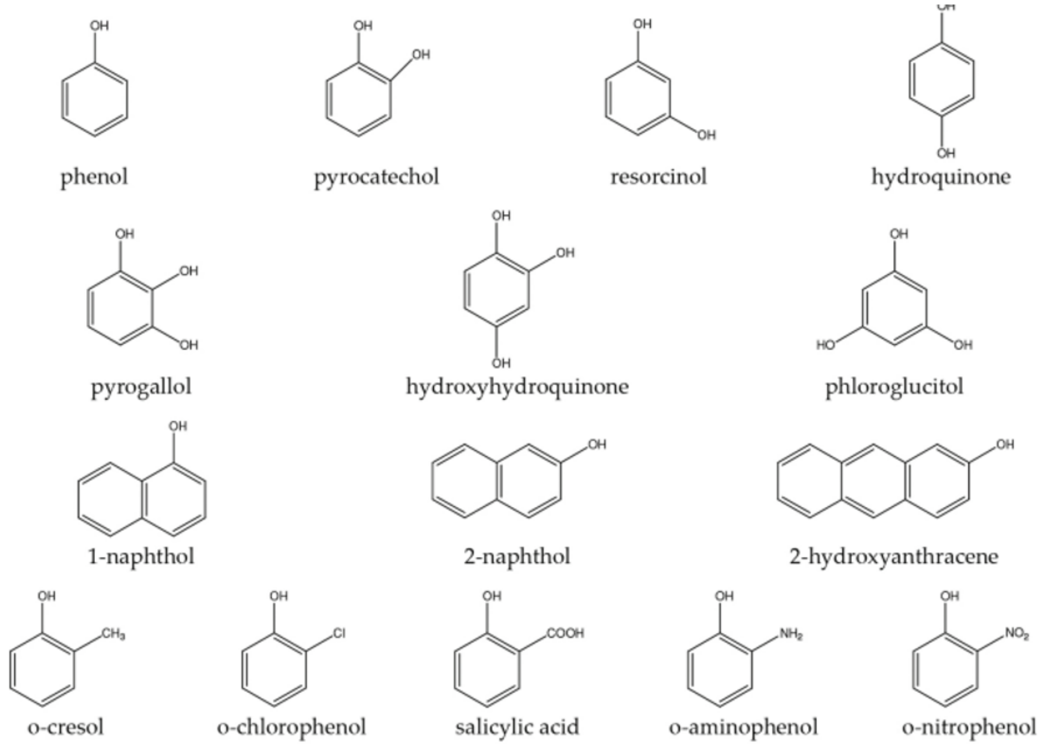

Fig.1. *The structure of phenolic compounds (modified from Sobiesiak,* [1].

Various techniques such as advanced oxidation, biological treatment, membrane separation, and adsorption are used to remove phenol from industrial wastewater [9]. Among these techniques, adsorption attracts attention because it shows the potential for effective phenol removal at low cost with the use of cheap adsorbents. The aim of this study is to compile the results of recent studies on the treatment of phenol by adsorption and to create an important data source for future studies on phenol purification.

Adsorbents Used In Phenol Removal

When the literature is examined, it is seen that a wide variety of adsorbents with different properties are used in phenol removal. Activated carbon, iron-oxide nanoparticles, zeolite, fly ash, and bentonite were used as successful adsorbents to remove phenol from solutions. Also, agricultural wastes such as rice husk, banana peels, pineapple peels, and coconut shells were used to adsorb phenol or phenolic compounds from water. In addition to this, biomass of bacteria, fungi, and algae were used as biosorbents for the removal of phenols.

The properties of the selected adsorbent are very effective in the success of the adsorption process. For example, when activated carbon is used as an adsorbent, the carbon source and activation method affect the removal efficiency of specific pollutants. These two factors are related to the formation and amount of activation sites on the activated carbon surface. The number of active sites on the surface of the activated carbon to be used as adsorbent greatly affects the adsorption efficiency. Fig. 2 is a diagram showing the active sites of activated carbon particles involved in adsorption.

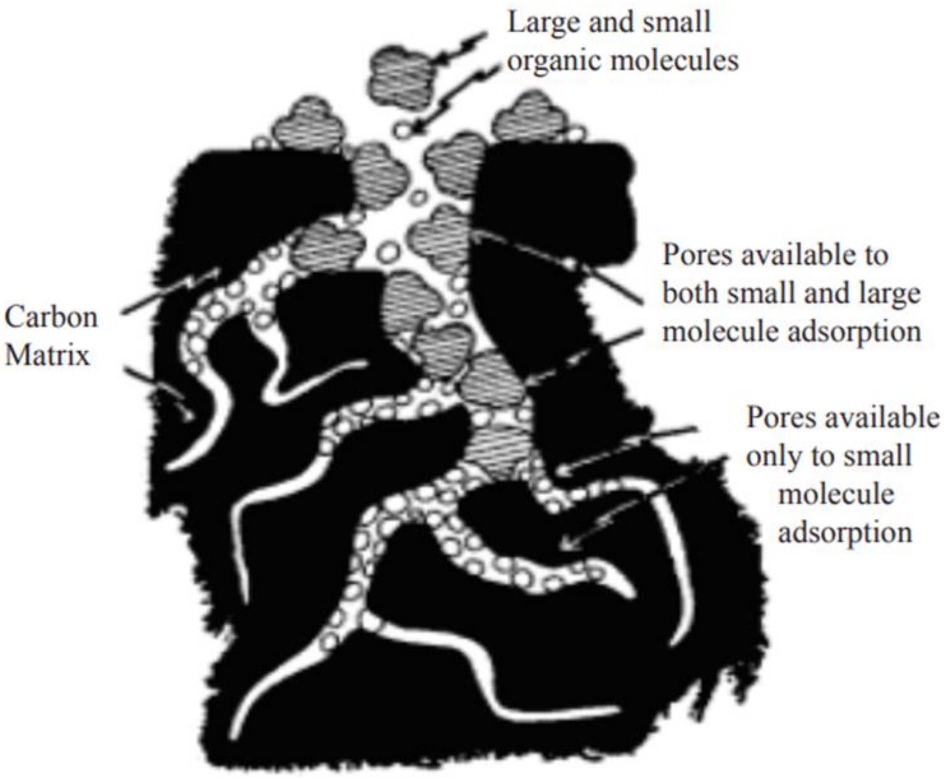

Fig.2. *Active sites of activated carbon. Adapted from Culp and Culp [10]*

There are many articles in the literature using activated carbon obtained from different carbon resources as an adsorbent for phenol removal [11; 12; 13; 14; 15; 16; 17]. Afsharnia et al. [11] carbonized pomegranate peels to prepare adsorbent and showed that the adsorbent performed a maximum adsorption capacity of 148.38 mg/g. Ignole et al. [12] used activated carbon from banana peel to remove phenol and determined 83% phenol removal (50 mg/L). Mojoudi et al. [13] pyrolyzed oily sludge to prepare activated carbon (AC) for phenol removal. Xie et al. [14] examined the phenol adsorption capacity of commercial activated carbons such as coal-derived granular activated carbons (GAC950), coal-derived powdered activated carbons (PAC800), and coconut shell-derived powdered activated carbons (PAC1000). According to the results of this study, the phenol adsorption capacity of GAC950, PAC800, and PAC1000 were found as 169.91, 176.58, and 212.96 mg/g, respectively. Also, Ho and Adnan [15] used coconut shell-based activated carbon as a successful adsorbent to remove 92% of phenol (10 g/L). Recently, Darla et al. [16] determined the phenol removal performance of activated carbon derived from *Saccharum officinarum* biomass (SBAC), and the maximum phenol removal capacity reached 166.95 mg/g at 500 mg/L phenol concentration. Recently, Dehmani et al. [17] used activated carbon (AC) originating from oak wood to remove 99% of phenol. As can be understood from these studies in the literature, activated carbon obtained from different sources can effectively remove phenol.

Senturk et al. [18] studied the phenol adsorption onto surfactant-modified Bentonite. Asnaoui et al. [19] showed the phenol removal success of Na-bentonite. Also, He et al. [20] reported that the phenol adsorption capacity of cetyltrimethylammonium bromide surfactant-modified bentonite was 10.1 mg/g at 150 mg/L phenol concentration. Recently, Alidra et al. [21] examined the phenol removal performance of tetrahepthylammonium bromide surfactant-modified bentonite. The surfactant-modified bentonite-based adsorbent removed 98.2% of phenol at 5 mg/L.

In an article on phenol removal of organic factory wastes, waste containing lemon pulp was dried and powdered and its usability as an adsorbent in phenol adsorption was examined [22].

There are studies in the literature on the usability of different microorganisms in phenol removal [23]. Ertuğrul Karatay [24] reported that the fungus *Aspergillus versicolor* removed 99.48% of phenol with the enhancement of surfactants. Abujayyab and Al-Zuhair [25] showed the effective phenol removal potential of microalgae (Chlorella sp. and Tetraselmis sp.). Campaniello et al. [26] reported the efficiency of mixed culture of *Candida boidinii* and *Bacillus pumilus* to remove phenol from wastewater. Benit et al. [27] showed that bacteria, *Halomonas halodurans* and *Bacillus halodurans*, successfully removed phenols from wastewaters. Bacteria, fungi, or microalgae can efficiently remove phenols or phenolic compounds from wastewaters [28].

Factors Affecting Phenol Removal

Many factors affect phenol removal. It seems that factors such as ambient pH, temperature, initial phenol concentration, and adsorbent amount are very effective in phenol removal.

Effect of pH

In a study, the effect of pH on phenol adsorption to organobentonite was examined and it was observed that while pH was almost constant between 1-9, adsorption decreased rapidly after pH 9 [18]. It has been reported that this situation is related to the weak acidic nature of phenol and its pKa value of 9.8. In an environment where pH is higher than 9, the ionization degree of phenol and the number of Oh-ions increase, and while the diffusion of phenol ions is prevented, adsorption decreases as a result of electrostatic repulsion with the negatively charged surface of the adsorbent. As a result, it was determined that the optimal pH was 9 when bentonite-derived adsorbents were used [28]. Similarly, Alidra et al. [21] showed that the modified bentonite (THPA-Ben) performed maximum phenol removal as 98.2% at pH 4.26 and ~ 9.

Beker et al. [29] examined the effect of pH on the phenol adsorption of activated carbon obtained from cherry stone and showed that phenol removal occurs best at basic pH values such as pH 6.5- 9 due to the electrical interactions between the phenol and activated carbon surface. Mojoudi et al. [13] reported that AC obtained by pyrolyzed oily sludge showed maximum phenol removal at pH 6.

According to the literature, phenol is an anionic molecule [17]. In the adsorption process, the interaction between the adsorbate load and the adsorbent surface is closely related to the pH of the medium. The pH of the environment affects the functional groups on the adsorbent surface and contributes to the electrostatic interaction between the adsorbate and the adsorbent surface. The results obtained from previous studies confirm this situation [13; 17; 18; 29].

Effect of Contact Time

Senturk et al. [18] showed that phenol adsorption onto organobentonite occurs rapidly at the beginning due to the high number of active sites required for adsorption on the adsorbent surface, and equilibrium is achieved at 60 minutes. Dehmani et al. [17] reported that activated carbon obtained from oak wood charcoal performed maximum phenol removal at 100 minutes. Recently, Darla et al. [16] studied the effect of contact time on phenol adsorption by activated carbon originating from *Saccharum officinarum* biomass. According to the results of this study, the highest phenol adsorption occurred in 90 minutes with 86%. Adsorption is related to the number of active sites on the adsorbent surface. Accordingly, while adsorption occurs rapidly over time, it slows down after a certain period due to the decrease in active sites on the adsorbent surface, and when there is no site interacting with the adsorbate, there will be no change in phenol removal.

Effect of Initial Phenol Concentration

The phenol concentration at the beginning of adsorption creates the driving force that can overcome the mass transfer resistance required for phenol transport between the organobentonite adsorbent surface and the solution [18]. On the other hand, adsorption decreases due to the saturation of active sites on the adsorbent surface at high concentrations in the solution. In their study, Galdino et al. [30] drew attention to the relationship between phenol concentration and micropore spacing in the adsorbent. It was stated that the lower the phenol concentration, the narrower the micropore space should be for effective adsorption [30]. Darla et al. [16] showed that initial phenol concentration affects adsorption onto SBAC and the phenol sorption efficiency decreased by augmentation of higher initial phenol concentrations. In general, adsorption decreases due to saturation of active sites on the adsorbent surface at high concentrations in the solution.

Effect of adsorbent dosage

Senturk et al. [18] stated that with the increase in the amount of organobentonic adsorbent, there was a decrease in the equilibrium adsorption capacity of the adsorbent, but phenol removal increased. The increase in phenol removal here is probably due to the increased availability of active adsorption sites as a result of the increase in organobentonite concentration. On the other hand, there may be two reasons for the decrease in the equilibrium capacity of organobentonite in phenol adsorption. According to the first of these reasons, organobentonite particles may have clustered with the increase in adsorbent concentration and as a result, the total surface area of the adsorbent may have decreased. According to a second reason, the increase in the amount of adsorbent with the constant phenol concentration and volume caused the formation of unsaturated adsorption sites and as a result, the equilibrium adsorption capacity of the adsorbent may have decreased [31].

In another study, it was shown that polyphenol adsorption efficiency increased with the increase in the adsorbent amount [32]. This situation can be associated with the increase in adsorption sites as a result of the increase in the amount of adsorbent [33]. However, the decrease in adsorption equilibrium capacity was reported with the augment of adsorbent dosage by Senturk et al. [18] and Garba et al. [31]. Similarly, Ververi and Goula [34] found that the adsorption of phenolic compounds onto wheat bran decreased with the increase in the amount of adsorbent and associated this decrease with the decrease in the surface area resulting from the clustering of adsorbent particles. Elayadi et al. [32] stated that a different result was obtained in their study compared to previous studies and that phenol adsorption increased with the increase in the amount of adsorbent in their study. Elayadi et al. [32] explained that this situation may be due to the balance between adsorbed and unadsorbed phenolic compounds in the experimental environment. Elayadi et al. [32] also reported that this result showed that the interactions between the factors were important in the process. The optimal conditions of different adsorbents for phenol removal published in the literature are given in Table 1. As it turns out optimal conditions are varied according to the adsorbent properties.

Effect of temperature

The effect of temperature on phenol removal was also investigated previously. Şentürk et al [18] showed that the phenol removal percentage by Organobentonite decreased from 59% to 49.5% as the temperature was increased from 0 to 40 ◦C and stated that phenol uptake by the adsorbent was preferred at low temperatures. Similarly, Mojoudi et al. [13] reported that phenol adsorption decreased with increasing temperature and that the best phenol removal was achieved at 25 °C, which was the lowest temperature they tried in their experiments. The decrease in adsorption with increasing temperature may be due to the weakening of the forces between the active sites of the adsorbent and adsorbate molecules. This situation may actually indicate a weak chemical reaction interaction between the adsorbate and adsorbent surface functionalities.

Table 1. *The optimal condition for phenol removal by different adsorbents in the literature (T: Temperature; CT: Contact time; PC: Initial phenol concentration; Ad. C: Adsorption capacity)*

Adsorbent	**pH**	**T**	**CT** (min.)	**PC** (mg/L)	**R%**	**Ad.C** (mg/g)	**Reference**
Organobentonite	9	0 °C	60	100	**-**	333.00	[18]
Activated carbon (Oily sludge)	6	25 °C	30	200	87.80	-	[13]
Activated Carbon (Coconut Shell)	3	25 °C	120	10	92.00	19.02	[15]
Activated carbon (Oak wood charcoal)	4	323 K	120.	200	98.77	**-**	[17]
Activated carbon (Commercially)	4	323K	120	200	91.48	**-**	[17]
Activated carbon (*Saccharum officinarum* biomass)	5.5	313 K	90	150	-	64.59	[16]
Organo-modified bentonite (Surfactant used)	4.25	55°C	90	5	98.20		[21]

ADSORPTION ISOTHERM AND KINETICS IN PHENOL REMOVAL

Adsorption isotherm and kinetic calculations provide detailed information about how the adsorption process occurs. The most common isotherm models are the Freundlich and Langmuir isotherm models. According to the Freunlich isotherm model, the amount of substance adsorbed by a certain amount of adsorbent first increases rapidly with pressure or concentration, and then gradually increases as the gas molecules on the solid surface reach saturation. The Langmuir isotherm model assumes that an adsorbate behaves as an ideal gas under isothermal conditions. According to this model, adsorbate molecules cover the entire surface of the adsorbent as a monolayer. In order to understand the efficiency of adsorption, the sorption rate must be determined. To determine this, pseudo-first order and pseudo-second order models are used in the analysis of sorption kinetics. The pseudo-first order model equation is probably the first known equation used to determine the adsorption rate in liquid phase systems. The equations used in the calculation of commonly used adsorption models are given in Table 2.

Table 2. *Commonly used adsorption isotherm and kinetic models*

Model		Equation	Explanation		Reference
İzotherm	Freundlich	$\ln(q_e)= \ln(K_F) + 1/n \ln(C_e)$	q_e (mg/g): the maximum capacity of adsorption at equilibrium C_e: solution concentration at equilibrium q_m (mg/g): the maximum capacity of adsorption q_t (mg/g): the maximum capacity of adsorption at time	K_F (L/mg): Freundlich isotherm constant	[35]
	Langmuir	$C_e/q_e = (1/q_m)C_e + 1/K_L q_m$		K_L(L/mg): Langmuir isotherm constant	[36]
Kinetic	Pseudo-first-order	$\log (q_e-q_t) = -k_1/2.303t + \log q_e$		k_1: the rate constant of the pseudo-first-order kinetic model	[37]
	Pseudo-second-order	$t/q_t =1/k_2 q_e^2 + 1/q_e.t$		k_2: rate constant of the pseudo-second-order kinetic model.	[38]

To date, there are many studies in the literature focused on adsorption isotherm and kinetic calculations regarding phenol removal [13; 14; 16; 18; 21; 39; 40; 41]. Senturk et al. [18] showed that phenol adsorption by organobentonite was suitable with Langmuir and Freundlich models. This situation is associated with both homogeneous and heterogeneous distribution of adsorption sites on the organobentonite surface. Mojoudi et al. [13] explained that the phenol adsorption of AC originating from pyrolyzed oily sludge was suited with pseudo second order and Freundlich isotherm models, respectively. Xie et al. [14] showed that the phenol removal phenomena of commercial activated carbon adsorbents (GAC950, PAC800, and PAC1000) were fitted with pseudo-second order kinetic and Langmuir models. Darla et al. [16] reported that phenol adsorption by SBAC was compatible with pseudo-second order and Radke-Prausnitz isotherm models. Alidra et al. [21] determined that phenol adsorption of THPA-Ben was fitted with pseudo-second-order model. Also, both the Langmuir and Freundlich isotherm models were best suitable to describe the phenol adsorption on THPA-Ben [21].

CONCLUSION

Phenol and phenolic compounds, which emerge as a result of both industrial and agricultural activities, are important environmental pollution factors and also have negative effects on human health. These compounds need to be treated to prevent their accumulation in high concentrations in aquatic environments. Adsorption appears as a successful treatment strategy. In this study, data from current literature focusing on the treatment of phenol by adsorption were compiled. Additionally, the factors affecting phenol adsorption were discussed in this study. The information compiled in this study will contribute to designing systems for phenol treatment in the future. In future studies, it is recommended to conduct laboratory-scale trials for phenol removal considering the optimal conditions discussed in this study.

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