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Evaluation of the Efficiency of a Biofilter System's Phenol Removal From Wastewater

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Abstract

Phenol is a toxic hydrocarbon that has been found in the wastewater of several industries, including the petroleum and petrochemical industries. The discharge of untreated wastewater from these industries causes environmental pollution, especially in water. The aim of this study was to evaluate the efficiency of phenol removal from wastewater using a biofiltration system. In this experimental study, a cylindrical plexiglass biofilter reactor with an effective volume of 12 liters was used. A total of 30 pcs of plastic grid discs were placed inside the reactor by plastic pipes to maintain the biofilm media in the reactor. The microorganisms used in this study were obtained from the biological sludge of a municipal wastewater treatment plant. The reproduction and adaptation of the microorganisms to 500 mg/L of phenol lasted three months. The effects of pH, phenol, nitrogen, phosphorus, glucose concentration, and hydraulic retention time on the biofilter system's performance was evaluated. The results of this study showed that in optimal conditions, this system can reduce the phenol concentration from 500 mg/L to zero within about 4 hr. Maximum efficiency occurred in pH = 7, and the proper COD/N/P ratio was 100/10/2, respectively. In general, this biofilter system is capable of removing 500 mg/L of phenol concentrations and an organic load of 4 - 4.5 kg COD/m^3.d within 4 - 5 hr. with high efficiency.

Keywords: Phenol, Glucose, Wastewater, Auxiliary Substrate, Biofilter

1. Introduction

Phenol (C_6H_5OH) is a toxic aromatic hydrocarbon with molecular a weight of 94.11 g/mol that is colorless or white solid in its pure form (1). This material and its derivatives have been used in several industries, such as in oil refineries; in the manufacture of resins, colors, pesticides, and pharmaceuticals; and in the petrochemical, coal mining, and steel and aluminum industries (2, 3). Phenolic compounds are obtained from raw oil degradation and thermal cracking or catalytic in oil refinery industries (4). Due to the extensive use of phenol in industrial processes, this pollutant enters the environment in different ways. Phenolic compounds have high solubility in water, and as a result, their existence possibility in water resources is high. Due to phenol's properties, such as its solubility in water and stability in the environment, this compound remains in the environment for a long time and can be transferred over long distances through water.

Due to their specific properties, such as toxicity, changes in the taste and odor of water, and adverse effects on human health and living organisms, phenolic compounds are classified as priority pollutants by the US environmental protection agency (5). Priority pollutants are organic or inorganic compounds with known effects that are suspected of having carcinogenic, mutagenic, teratogenic effects or that have high toxicity (6). Therefore, detecting and determining the level of phenolic compounds in the environment, particularly in water resources, and subsequently monitoring these levels are highly important for controlling the emission of this material and reducing its effect on the environment.

Various methods have been considered for treating wastewater that contains phenol, such as chemical oxidation, adsorption, and biological treatment (7, 8). Of these methods, biological systems have been used most often due to their advantages over other methods. One of these advantages is compatibility with the environment (9). Additionally, this method does not employ harmful chemical compounds, so the effluent and sludge disposal produced by this process has lower adverse effects on acceptor resources in comparison with chemical processes (10).

One biological method for removing pollutants from wastewater is biofiltration. Using a biofilter for the emission control of volatile organic compounds is considered to be a new technique. Wastewater that has low biodegrad-

able compounds is stable enough for biofiltration. Biofilters are reactors that have media with microorganisms growing on them that enable the treatment process (11). A biofilter has a chamber with media inside it that promotes microorganism stabilization. These microorganisms produce a thin layer called biocoating (12). Generally, pollutants passing through these porous biological media are separated from water and are treated.

The ability of biofiltration to remove hydrocarbons has been investigated in a few studies (13-15). Neves et al. (2006) (14) showed that biofiltration can be used to remove phenol present in liquids or gaseous effluents through the use of aerobic microorganisms that are immobilized on solid or porous supports. Khalil and Singh (2012) (16) demonstrated that the parameters of surface area, rate constant, biofilm thickness, porosity, effective diffusivity, gas flow rate, and initial phenol concentration are important for phenol removal through biofiltration. In a mixed-culture environment with both biodegradable materials and materials that are resistant to biodegradation, the microorganisms in a biofilter could overcome the biodegradation-resistant molecules and degrade the toxic materials.

These components have been used as an auxiliary substrate in various reactors, such as an up flow anaerobic blanket for the removal of phenol and chlorophenol (17, 18). According to some studies, volatile fatty acids, sucrose (19), glucose (20-22), acetate (23), and other similar compounds can be biologically degraded. These compounds have been used by microorganisms as stable substrates of carbon and can be introduced to accelerate the act and to reduce the set-up time and preparation of reactors to degrade persistent material effectively. These methods could reduce the hydraulic retention time (HRT) of reactors by a significant degree.

The present study evaluated phenol removal in an aerobic condition and in the presence of various concentrations of glucose, which has high biological degradation efficiency. According to these resources, the present study has evaluated the effects of phenol and glucose concentration on a biofiltration system's efficiency when removing phenol from wastewater.

2. Materials and Methods

This study employed a cylindrical biofilter reactor made from Plexiglas with an effective volume of 12 liter. A total of 30 plastic reticulated discs were placed inside the cylinder by the plastic tubes as biofilm media. The biofilter's schematic is shown in Figure 1.

The air required for biological reactors was provided by a refrigerator compressor with a power level of 1 hp. The hydraulic flow in the studied system was continuous, and the type of distribution in the water was upward. For this reason, the entrance of the initial solution into the system was at the bottom, and the exit of the effluent treatment was at the end of each of the reactor's entrances.

Two double diaphragm pumps were used to inject the considered solution into the system. The irrigation capacity of the pumps was regulated between 0.1 - 2 mL/s.

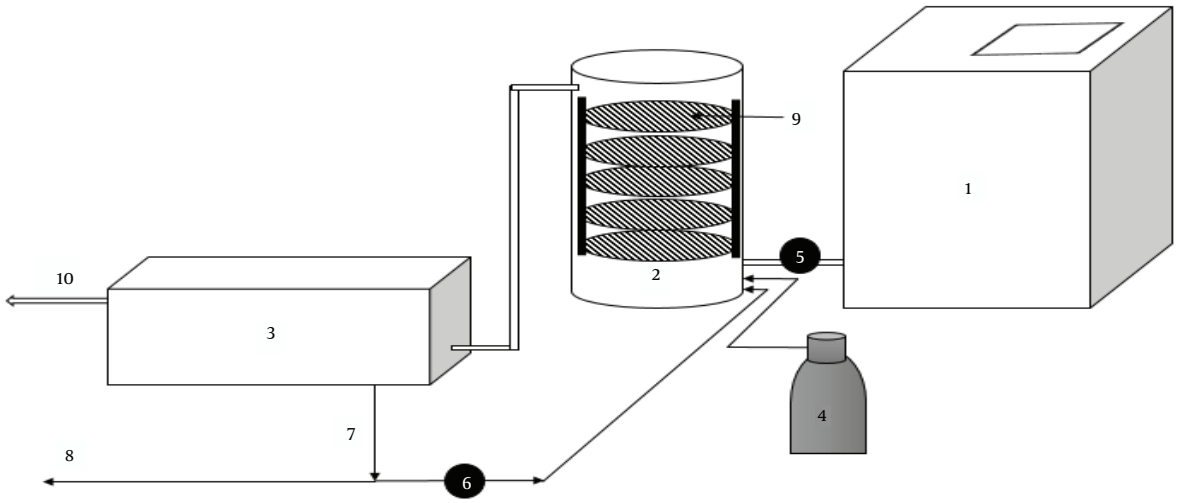
The sedimentation basin and clarifier consisted of a basin with a dimension of 20 $\times$ 30 cm and height of 90 cm.

To evaluate any phenol that may have been emitted because of aeration from the top of the biological reactor, a vacuum pump passed all the output vapors and gasses of the reactors through an impinger, which contained 0.1 normal sodium hydroxide solution.

This act continued for 14 hr. in accordance with the vacuum pump batteries' capacity. In order to ensure the accuracy of the results, the sampling was repeated three times. To adapt the microorganisms in the filter to phenol, phenol with a concentration of 0.1 mg/L and powdered milk were initially injected into the system, and the phenol concentration was gradually increased while the powdered milk concentration was decreased. Over the course of one month, the phenol concentration was increased to 500 mg/L. Phenol, as a source of carbon and energy for the microorganisms, and nitrogen and phosphorus, as nutrients, were injected into the system in the form of the primary product. To evaluate the effects of the nitrogen and phosphorus concentrations at the beginning of the study, the COD/N/P ratio was initially 100/5/1, respectively, and the concentrations of these substances changed over time. To determine the effects of N and P on phenol removal efficiency in the studied system, N was added to the phenol solution at 30, 40, 50, 60 and 80 mg/L, and P was added to the phenol solution at 6, 8, 10, 12 and 16 mg/L, and the efficiency of each was evaluated.

All chemicals used were analytical reagent grade and were used without any further purification. The chemicals were purchased from Merck (Germany). In addition, the measurement of the efficiency parameters of the bioreactor was carried out according to the standard methods of water and wastewater analysis (24). For the COD and phenol, a colorimetric technique with a closed reflux and photometric methods were developed, respectively. A spectrophotometer (DR 5000, Hach, Jenway, USA) using wavelengths of 600 nm and 500 nm was used to measure the absorbance of the COD and phenol samples.

The analysis of the results was done using SPSS version 21. The correlation coefficient level was analyzed between the variables using Pearson's correlation, Kendall's tau, and Spearman's rank correlation coefficient.



1) Storage Tank
2) Biofilter
3) Sedimentation Tank
4) Air Compressor
5) Pump
6) Return Sludge Pump
7) Sludge Discharge
8) Excess Sludge
9) Media
10) Effluent

Figure 1. Schematic of the Studied Biofilter System

3. Results and Discussion

To evaluate the effect of glucose as a competitive substance on phenol removal efficiency in the studied system, while the phenol concentration in the input solution into the system was held at 500 mg/L, glucose was added to the phenol solution in concentrations of 50, 250, and 500 mg/L. After fixing the situation's and system's performance, the phenol removal efficiency in the biofilter system was evaluated.

The results showed that the 50 mg/L concentration of glucose caused the phenol removal efficiency to increase, but with higher concentrations of glucose, there was an efficiency reduction. This result is consistent with the results of Dargahi et al.'s (2014) (25, 26) and Shokoohi et al.'s (2005) (27) studies. The evaluation of the correlation between these two variables showed that the Pearson's correlation coefficient for these two variables was -0.419 ($P < 0.042$). Table 1 shows the system's phenol removal efficiency with various glucose concentrations. In Pishgar et al.'s study (2012) (20), glucose concentrations of 500 - 3000 mg/L had a positive effect on the biological degradation of phenol. The results of that study aligned with those of the present study (with HRT < 6.5 hr.). With an increase in time, glucose had a negative effect on bioreactor efficiency. With an increase in glucose concentration, phenol removal decreased (20).

Changes in glucose concentration produced different microbial species in the system. In the absence of glucose, *Pseudomonas aeruginosa*, *Moraxella*, *Brevundimonas*, *Pseudomonas alcaligenes*, and *Acetinobacter* were recognized in the presence of glucose. With increases in glucose concentration, *Escherichia coli* was identified and gradually dominated at 250 and 500 mg/L. Additionally, as the glucose concentration increased, no *Acetinobacter* was recognized. At 250 and 500 mg/L glucose, *Brevundimonas* and *P. alcaligenes* were also removed from the system. Finally, at 500 mg/L of glucose, *Neisseria weaveri* was not found.

Table 1. Phenol Removal in 500 mg/L Phenol, 50 mg/L Nitrogen, and 10 mg/L Phosphorous With Various Glucose Concentrations

Glucose, mg/L	Phenol Removal (%)
0	23.9 ± 2.59
50	54.92 ± 19.6
250	28.75 ± 10.1
500	22.76 ± 1.8

In order to detect the correlation between the MLSS concentration of biofilter reactors and phenol removal efficiency, the samples were evaluated with various MLSS concentrations and a constant phenol condition of 100 mg/L (Figure 2 shows the results of this evaluation). The

correlation test results of these two variables showed that Pearson's correlation coefficient for these two variables was 0.68 ($P < 0.001$).

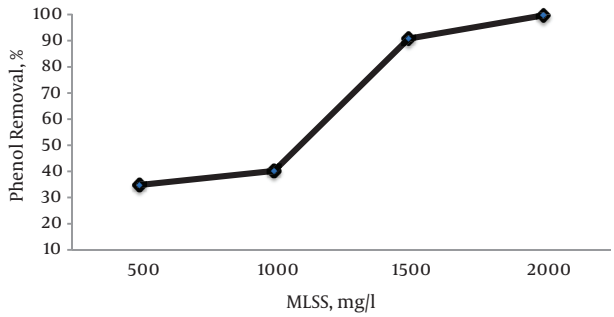


Figure 2. Phenol Removal Efficiency at Different MLSS Concentrations

In order to determine the relationship between phenol concentration and phenol removal efficiency in biofilter reactors, phenol concentrations of 100, 250, 400, and 500 mg/L were entered into the system and the biofilter efficiency was evaluated. In this stage, the effective condition of the system was stable. The results showed that when the input phenol concentration in the system increased, the phenol removal efficiency declined (Figure 3).

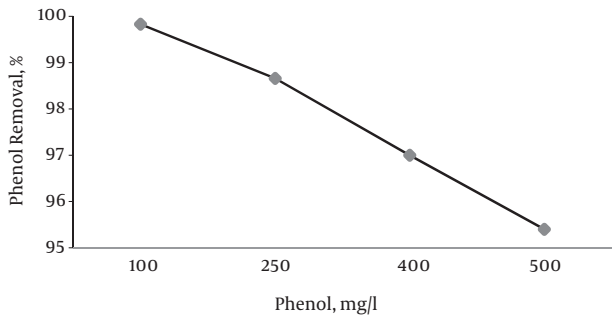


Figure 3. The Studied System's Phenol Removal Efficiency at Various Phenol Concentrations

Based on these results, the studied system could remove almost 100% of the input phenol of system with an organic load of 4 - 4.5 kg COD/m^3.d. Notably, biofilter reactors could remove phenol with 90.8% efficiency with an organic load of 8 - 9 kg COD/m^3.d.

This high efficiency could have various causes. One of these potential causes is the high microbial variety in the system. Different microorganisms have different phenol degradation capabilities. Therefore, by increasing the number and variety of microorganisms in biological treatment systems, treatment efficiency increases.

Another potential cause of the high efficiency of this system is that the high organic load, the physical situation of the system, and the type of hydraulic flow may have all contributed to the high efficiency of the studied system. This system was designed to provide a specific surface for the attachment of microorganisms, which resulted in low probabilities of drainage clogging and air in the system. For this reason, there was a high probability for the microorganisms in the system to access and attach to foodstuffs, nutrients, and oxygen in various parts of the system.

Yoong et al. (2001) (28) evaluated the SBR biological system's capability for phenolic wastewater treatment with a concentration of 1300 mg/L. The highest phenol removal efficiency in their study was 97% with an organic load of 3.12 kg BOD/m^3.d, which was consistent with results of the present study.

Determining the relation between the pH of the input solution and phenol removal efficiency in the evaluated system was one of the primary aims of this study. Usually, the most suitable pH for bacteria growth is 7. However, some bacteria, such as Thiobacillus and solphpluboos, grow in $pH < 2$. Fungus prefers a pH of 5 or lower. In contrast, Cyanobacteria grow in $pH > 7$. Usually, bacteria's growth is reduced because of the production of metabolic acid products, such as organic acids and sulfuric acid. However, in some cases, like denitrifies and algae, growth increases with higher pH levels. Microbial enzyme activity and chemical ionization reactions are affected by pH. As a result, it is effective for transferring nutrients and toxic materials into cells (29).

Liu et al. (2016) (30) evaluated the phenol removal capability of *Acinetobacter calcoaceticus* in various pH levels. The results of their study showed that the maximum phenol removal occurred with a pH of 8 and a temperature of 30°C, which is consistent with the results of the present study. The present study evaluated the relation between pH and phenol removal efficiency and determined the suitable pH for accessing maximum phenol removal efficiency in the studied system. The system's efficiency was evaluated in pH ranging from 6.5 to 8 (intervals of 0.25). The effect of pH on phenol removal efficiency was not constant. Therefore, a Pearson's correlation coefficient of 6.5 - 7 was used for the evaluation. Based on these results, the Pearson's correlation coefficients between the pH variables of 6.5 - 7 and 7 - 8 were 0.936 and -0.936, respectively ($P < 0.05$). These values showed that there was a direct relationship between the pH of the input solution and phenol removal efficiency in biofilter system with a pH of 6.5 - 7, and with a pH of 7 - 8, there was an inverse relationship between these two variables.

Therefore, the best pH for accessing the maximum phenol removal efficiency in the studied system was 7. By

changing the pH, the efficiency rate was significantly reduced. Notably, the level of pH changes that affected the system's efficiency are related to the ranges that are tolerable for most microorganisms. Out of this range, the effect could be increased, which would eventually threaten the microorganisms' lives and the system's overall efficiency. Figure 4 shows how the system's phenol removal rate was affected by changes in pH.

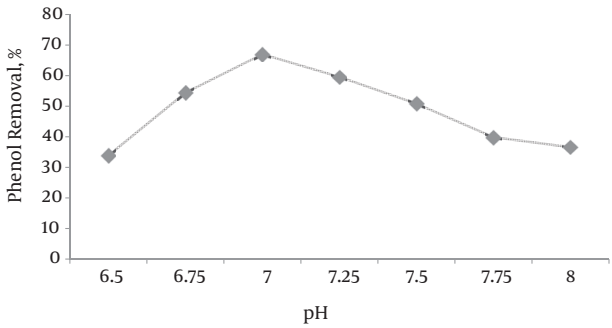


Figure 4. Phenol Removal Efficiency at various pH Values With a Constant Phenol Concentration of 500 mg/L

The concentration of treated substances is one of the most important factors in the performance and efficiency of various treatment systems, including biological systems. In this study, in order to evaluate the relationship between the input phenol concentration and the system's phenol removal efficiency, phenol was added into the system in concentrations of 100, 250, 400, and 500 mg/L. The results showed that there was an inverse relationship between the input phenol concentration and the removal efficiency of system, which was consistent with the results of Almasi et al.'s (2012) (31, 32) and Nakhli et al.'s (2014) (33) studies. In Nakhli et al.'s (2014) study, phenol concentrations of 200 - 1200mg/L were evaluated, and the maximum level of phenol removal was related to the 200 mg/L concentration of phenol. The Pearson's correlation coefficient between the removal efficiency and input phenol concentration variables was -0.446 ($P < 0.001$). Raikar et al. (2015) (34) studied the degradation of phenol in a biological system in India. Their study showed that the maximum phenol removal efficiency was 97% for 200 ppm of initial phenol concentration under 25 hr. of HRT. The HRT in Raikar et al.'s study was higher than the HRT in this study, and the phenol concentration was lower. In the present study, the HRT required to access a suitable efficiency for treatment or phenol removal in 100 mg/L concentration was a 4.4-hr. retention time with more than 99% efficiency. Figure 5 shows the changes in phenol removal efficiency with various HRT values and a constant phenol concentration of 100 mg/L.

The results of this study showed that the Pearson's correlation coefficient between HRT and phenol removal efficiency for the biofilter system was 0.643. These results showed that there was a correlation or direct relationship between HRT and phenol removal efficiency. The results of this study showed that under some circumstances, the phenol concentration of biofilter reactors could be reduced from 500 to 0 mg/L within 4 hr.

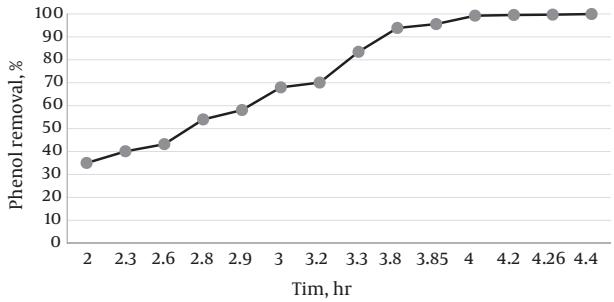


Figure 5. Phenol Removal Efficiency at Various Hydraulic Retention Times With a Constant Phenol Concentration of 100 mg/L

In order to evaluate the effects of nitrogen and phosphorous on phenol removal efficiency in the studied system, nitrogen with a concentration of 30, 40, 50, 60, 80 mg/L and phosphorous with a concentration of 6, 8, 10, 12, 16 mg/L were added to the phenol solution with a constant phenol concentration of 500 mg/L. An increase in the nitrogen concentration in the system to 50 mg/L caused an increase in efficiency, but any further change in nitrogen concentration (increase or decrease) reduced phenol removal efficiency.

The Pearson's correlation coefficient for nitrogen and phenol removal efficiency with a nitrogen concentration of 30 - 50 mg/L was 0.883 ($P < 0.002$), and with a nitrogen concentration of 60 - 80 mg/L, the Pearson's correlation coefficient was -0.894 ($P < 0.003$). Since the phosphorous concentration changes occurred at the same time as the nitrogen concentration changes, the correlation coefficients for phosphorous concentrations and phenol removal efficiency are the same.

According to the obtained results, the phenol degradation in the studied system was based on second-grade kinetic reactions. By increasing phenol concentration, phenol degradation speed was progressively reduced. The results of this stage in the research showed there was a serious correlation that between the phenol concentration and degradation speed and removal efficiency, and because the P value in all these tests was zero, the correlation between these two variables was 100% significant.

The reduction in the speed of phenol degradation due to increased phenol concentration could have several causes. One of the most important potential causes is that increases in phenol concentration caused an increase in the mortality of bacteria, resulting in the death of microorganisms that had a lower tolerance and resistance against phenol toxicity. In addition, the bacteria showed greater sensitivity to the increased phenol concentration. By slightly increasing the phenol concentration, the bacteria's mortality rate increased by a disproportionate amount. Of course, different bacteria had a different reaction to increases in phenol concentration. For example, *Pseudomonas* was placed in a higher level for the mentioned index in comparison with other microorganisms and showed greater resistance against increased phenol concentrations. Therefore, they were able to tolerate and degrade high concentrations of these substances (32).

In a study by Bandyopadhyay et al. (1998) (35), the phenol degradation capability of *Pseudomonas putida* (MTCC 1194) was in phenol concentrations of 100 - 1000mg/L. In phenol concentrations higher than 500 mg/L, *P. putida* dominated the phenol degradation process, which is consistent with this study's results.

4. Conclusion

In the present study, a biofilter system was developed for the removal of phenol from wastewater. The results of this study revealed that the biofilter provided improved phenol efficiency and complete phenol removal. The reactor can demonstrably handle a high capacity of phenol. The optimum HRT, MLSS, and phenol concentration for the biofilter are 4.5 hr., 2000 mg/L, and 100 mg/L, respectively, which produced 100% phenol removal. Additionally, inlet phenol concentrations up to 500 mg/L did not significantly affect the performance of the biofilter, with phenol removal efficiency remaining above 95% at a phenol concentration of 500 mg/L. This biofilter system could remove phenol with a concentration of 500 mg/L and an organic load of 4 - 4.5 kg COD/m^3.d in 4 - 5 hr. after three months with high-efficiency microorganism adaption. The reactor demonstrated a high capacity for phenol removal compared with the reactors reported in the literature.

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