

Original Research

Survey on Treatment Ability of Stabilization Pond as a Natural Treatment System in Oil Refinery Wastewaters

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Abstract

An oil refinery is one industry that introduces large amounts of toxic phenol in receiving waters annually. Hence, the aim of this study was phenol removal from oil refinery wastewater by a natural treatment system of a stabilization pond. In this study, a pilot-scale pond volume, hydraulic load, and hydraulic retention time were 200 L, 40 L/day and five days, respectively. Influent concentration of phenol was 100-400 mg/L. Responses of the process were NH₃, PO₄, phenol, TCOD, SCOD, TBOD, SBOD, and pH. The results showed that increasing phenol concentration and decreasing temperature have negative effects on system efficiency. Maximum removal of NH₃, PO₄, phenol, TCOD, SCOD, TBOD, and SBOD were 61.08, 70.09, 93.58, 80.18, 78.89, 78.7, and 76.84% in high temperature, respectively. Maximum efficiency of wastewater treatment system was obtained in phenol concentration at 100 mg/L. Also, anaerobic stabilization ponds are cost-effective options with simple operation that can be employed for the treatment of phenol content of oil refinery wastewaters.

Keywords: anaerobic, stabilization pond, phenol, oil refinery wastewater

Introduction

Phenol (C₆H₅OH) is one of the toxic aromatic hydrocarbons with molecular weight 94.11g/mol. The pure solid form of phenol is white, but it is mostly colored due to the presence of impurities [1]. Phenol

has a sweet taste, tar-like odour, and is soluble in organic solvents such as alcohol, glycerol, petroleum, etc. However, it is partially soluble in water [2]. Phenol is one of the most common organic pollutants due to its toxicity even at low concentrations [3-4]. Phenol and its derivatives are reported to be present in the effluent of many industries such as paper and pulp, pesticides, dyes, and chemical manufacturing [4]. Additionally, other industrial wastewaters such as resin manufacturing, gas and coke manufacturing, tanning, textiles, plastics,

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rubber, pharmaceuticals, oil refineries, ceramics, steel, coal conversion processes, phenolic resin industries, and petroleum contain different forms of phenols [5]. Phenols also are present in domestic effluents [6]. Therefore, wastewaters containing phenolic compounds can cause serious water pollution due to their poor biodegradability, high toxicity, and ecological aspects [4,7].

According to the World Health Organization (WHO), the maximum permissible concentration of phenol in potable water is 0.002 mg/L. The U.S. Environmental Protection Agency (EPA) has stated that maximum permissible phenol content in wastewaters must be less than 1 mg/L [8]. Several technologies have widely been applied for phenol and phenolic compound removal from wastewaters such as physicochemical, biological, and advanced oxidation processes (AOPs) [9-10]. But high cost, low efficiency, the formation of toxic by-products, and applicability to a limited concentration range are problems associated with the above methods. On the other hand, biological processes have an advantage over these methods which have little or no harmful effects on the environment, because these techniques do not involve the use of harmful reagents [11]. In recent years, research on various methods of biological treatment, including biodegradation of oil refinery effluents in a pond of activated sludge, was performed. Results indicated that the total chemical oxygen demand (TCOD) removal efficiency by this system was desirable [12]. Among these biological treatment systems, wastewater stabilization ponds (WSPs) are useful for both industrial as well as municipal wastewater treatment. WSPs have been widely utilized in developing countries, especially in rural areas. WSPs offer a cheap and attractive alternative to conventional processes, in case adequate land is available. Nowadays, WSPs have been used in many parts of the world as a series of anaerobic, facultative, maturation ponds [13].

Anaerobic ponds are the smallest unit in the series. They are sized according to their volumetric organic loading, which may be in the range of 100 to 350 g BOD₅/m³ d, depending on the design temperature. The depth of anaerobic ponds is in the range of 2 to 5 m and hydraulic retention time (HRT) is usually between two and five days [14]. Anaerobic ponds work extremely well in warm climates: for example, a properly designed pond will achieve around 60% BOD₅ removal at 20°C and over 70% at 25°C and above [15]. Results of the study by Moussavi et al. showed that phenol removal efficiency using the aerobic granular SBR was more than 99% [16]. On the other hand, Zhai's et al. study showed that using the chitosan-halloysite hybrid-nanotubes process had high phenol removal efficiency [17]. It is worth mentioning that the technology used in the above study is expensive and requires specialized experts, while the applied method in this study is the simplest and most flexible environmental technology. A literature survey revealed that there are no comprehensive reports on phenol removal from oil refinery wastewater by anaerobic WSP. Therefore, the main objective of the present study was to investigate and

assess an anaerobic pond at pilot scale for treatment of Kermanshah oil refinery wastewater.

Materials and Methods

This experimental study was done as a pilot scale. The WSP with dimensions of 0.2×1×1 m were made of fiberglass plate. Experiments were carried out at ambient air temperatures ranging 25-42°C. Average temperature of the pond was kept at 21±2°C. In this study, HRT of the anaerobic pond was five days, and hydraulic load of this system was 40 L/day. The inlet of the anaerobic pond was positioned at 30 cm below the pond surface. The pond was loaded daily by the wastewater output of oil and grease separator unit of Kermanshah oil refinery. Before launch of the system, it underwent seeding and inoculation measures. A seeded sludge was prepared by adding 1.5 liters of sewage sludge and a liter of previously prepared sludge from the oil refinery plant to the system input, before loading the system with wastewater. WSP was ready for launch after three months of seeding. The molasses was used to adjust the anaerobic pond loading besides increasing phenol concentration. In this study, phenol was added to the input of the pilot in various concentrations (100, 200, 300, and 400 mg/L) for both warm (higher than 20°C) and cold (lower than 10°C) temperatures. Then the parameters of NH₃, PO₄, and phenol were measured using Varian spectrophotometer (model UV-120-20) at wavelengths of 425, 690, and 500 nm, respectively, and also total chemical oxygen demand (TCOD), soluble chemical oxygen demand (SCOD), total biochemical oxygen demand (TBOD), soluble biochemical oxygen demand (SBOD), and pH were measured.

Oxidation and reduction potential (ORP) of the pool were measured to maintain and provide anaerobic conditions. This parameter was determined using a Kent ORP meter (7020 model and with the Eil sensors).

Phenol (purity > 99.5%) was obtained from Merck, Germany. To clarify the phenol volatility theory, the

Table 1. Characteristics of Kermanshah Oil Refinery raw wastewater.

Parameter	Concentration (mg/L)
TCOD	622±48
SCOD	495±61
TBOD	204±16
SBOD	126±8.3
TSS	56±5.2
VSS	44±4.7
N-NH3	13.1±2.2
Phenol	69.6±4.9
pH	7.9±0.31

pond's surface was isolated with a layer of paraffin and plastic cover and system performance was evaluated. Five consecutive samples showed that the performance rate of the anaerobic pond is almost equal in both open and closed conditions. In this study, a total of 2,400 samples were measured at two temperatures and four phenol concentrations. Descriptive statistics used for presenting data and analytical statistics by SPSS ver 21. T-test, and ANOVA were applied for comparing WSP efficiency in removal of different phenol concentrations. All sampling procedures and parameter analysis were carried out according to standard methods [18]. Table 1 shows the quality characteristics of Kermanshah oil refinery raw wastewater.

Results and Discussion

Tables 2 and 3 show the wastewater characteristics of influent and effluent of the anaerobic WSPs at high and low temperatures, and also the mean removal efficiency of measured parameters in different phenol concentrations for wastewater treatment of Kermanshah oil refinery. The operation of pilot anaerobic WSPs was provided under completely anaerobic conditions. Meeting the anaerobic conditions was confirmed by oxidation-reduction potential (ORP: -246). In the system, loading volume in warm temperatures was 104.2, 121.54, 136.01, and 148.12 g BOD₅/m³ d in phenol concentrations of 100, 200, 300, and 400 mg/L, respectively. Also, in cold temperatures the mentioned concentration was 100, 118.55, 131.74, and 143.48 g BOD₅/m³ d, respectively, by considering that the standard loading volume in anaerobic ponds was in the range of 100-400 g BOD₅/m³ d.

Results indicated that the studied parameters (including phenol concentration and temperature) have dramatically affected the WSP efficiency in oil refinery wastewater treatment. Therefore, the removal efficiency was significantly increased with decreasing phenol concentration and increasing temperature ($P < 0.001$). According to the obtained results, the performance of a laboratory-scale anaerobic stabilization pond in removing the COD and BOD₅ is better than that of Papadopoulos et al. [19]. Study results were recorded even in the worst possible conditions (high phenol concentration and low temperature). The results of the current study showed that the removal efficiencies of BOD₅ and COD from the urban sewage by anaerobic WSP were 45% and 50%, respectively. COD/BOD₅ ratio in the input and output of the system were 2.07 and 2.05, respectively. Moreover, WSP performance in the treatment of the oil refinery wastewater in different input phenol concentrations was better than that of Ghazy et al. [20] study results. In the current study, the removal efficiencies of COD, BOD₅, and PO₄ by WSP were 29.63%, 28.38%, and 16.74%, respectively. In our study, the increase of COD removal compared with BOD removal can be attributed to the multi-phase state of oil wastewater in which its layers have been settled on the surface and there is the volatility

potential for them. Moreover, some of the layers are separated from the liquid due to hydrophobic property and precipitate in the water column of the reactor or remain suspended in the liquid column. Another advantage of this system is biodegradation of resistant materials. Due to bacterial hydrolysis, these compounds were converted into catechol, aldehydes, and acids, and can be degraded by WSP. For this reason, besides the higher removal of COD in the reactor effluent compared with the BOD₅, the COD/BOD₅ ratio was identical in both input and output of the anaerobic bioreactor. These results are consistent with the Abdel-Raouf et al. study [21].

The closeness of results in our study in BOD₅ and COD removal with the results of other authors like Park [15], Leven [22], and Ebrahimi [23] is evident. Their study showed that the removal efficiency of BOD₅ from wastewater in anaerobic ponds under warm conditions (summer) is 60-70% and in cold conditions (winter) is 40-50%. Also, it is consistent with the study undertaken by Almasi, who used the anoxic pond for wastewater treatment in both warm and cold conditions with different volumetric organic loads [24]. Almasi and Pescod [24] have shown that the rate of BOD₅ and COD removal under warm conditions by the anoxic pond system were 77% and 68.28%, and under the cold conditions were 62.3% and 48.95%, respectively. The temperature effect on the speed of the biological reactions is an evident and proven fact. In fact at temperatures below 15°C an anaerobic pond acts as a sedimentation tank, which would collect plenty of the sludge [25]. The results of the Almasi et al. study showed that in domestic wastewater the COD removal efficiency decreased with decreasing temperature [26]. Moreover, the result of the Phan et al. study on phenol removal at different temperatures showed that the efficiency of the system increased with increasing temperature [27].

In a study undertaken by Ramos et al using the laboratory scale facultative stabilization pond in which wastewaters with high phenol content was used for removal of phenol with different concentrations, the results showed that the highest and lowest rates of phenol removal relates to 1,000 mg/L (92%) and 4,000 mg/L (22%) concentrations, respectively [28]; but for the anaerobic pond for phenol removal no independent study was found. Results of the study by Nahid et al. showed that by the increase in concentrating phenol within the range 0-200 mg/L, the COD removal rate is reduced due to the phenol toxicity effect on the microbial mass activity [29]. That was in agreement with this study. The study results carried out by Li et al. showed that phenol removal efficiency from coal gasification wastewater by the moving bed biofilm reactor has been 89% [30]. Yousef et al. showed that by increasing the concentration of phenol in influent wastewater, the efficiency of removal by the pond is reduced [31]. The result of Yousef et al. is according to this study.

Optimal conditions resulting from this study were evaluated considering the performance of anaerobic pond in oil wastewater treatment and the decrease in the phenol concentration in the output. The highest efficiency of phenol removal in this study was obtained in a phenol

Table 2. Wastewater characteristics of influent and effluent of anaerobic stabilization ponds in different phenol concentrations at high temperature.

Parameter	Phenol Concentrations (mg/L)												Removal Efficiency (%) for phenol Concentrations (mg/L)			
	100			200			300			400			100	200	300	400
	Influent	Effluent		Influent	Effluent		Influent	Effluent		Influent	Effluent		Influent	Effluent		
pH	7.93±0.41	7.09±0.3		7.91±0.35	7.09±0.2		7.85±0.33	7.13±0.3		7.9±0.38	7.2±0.33		-	-		-
BOD ₅ (mg/L)	500.07±34.81	106.48±16.9		418.67±34.8	157.84±15.67		658.74±20.21	194.3±15.43		717.36±65.61	293.5±33.72		78.7	73.32	70.52	59.13
SBOD	418.67±34.8	96.92±10.6		494.88±20.21	138.94±15.67		513.74±20.21	170.4±15.42		630.6±65.61	271.6±33.72		76.84	71.84	66.85	56.96
TCOD	1,586.06±199.6	316.32±69.4		1,873.49±69.18	440.38±43.74		2,061.85±63.26	567.3±15.41		2,378.56±255	900.67±124.8		80.18	76.44	72.47	62.12
SCOD	1,435.46±199.6	303.62±56.8		1,608.49±69.18	424.88±43.74		1,780.35±63.26	554.3±15.41		2,171.2±245	897.14±119.97		78.89	73.51	68.84	58.67
NH ₃ (mg/L)	16.38±2.18	6.39±1.1		20.7±3.01	9.04±2.01		20.19±4.4	10.08±2.24		19.33±3.83	11.45±2.14		61.08	56.22	49.1	40.12
PO ₄ (mg/L)	2.46±0.47	0.73±0.2		2.64±0.46	0.9±0.25		2.74±0.4	1.27±0.21		2.78±0.41	1.38±0.2		70.09	65.19	53.16	49.73
Phenol (mg/L)	1,71.71±12.39	11.01±1.8		266.44±9.65	48.9±13.37		369.38±14.3	113.05±11.17		464.42±15.4	180.5±10.5		93.58	81.63	69.38	61.12
Volumetric Loading (g BOD ₅ /m ³ d)	100	-		118.55	-		131.74	-		143.48	-		-	-		-
TCOD/TBOD	3.172	2.98		4.48	2.8		3.13	2.92		3.31	3.07		-	-		-
ORP																

Table 3. Wastewater characteristics of influent and effluent of anaerobic stabilization ponds in different phenol concentrations at low temperature.

Parameter	Phenol Concentrations (mg/L)												Removal Efficiency (%) for phenol Concentrations (mg/L)			
	100			200			300			400			100	200	300	400
	Influent	Effluent	Influent	Effluent	Influent	Effluent	Influent	Effluent	Influent	Effluent	Influent	Effluent	Influent	Effluent	Influent	Effluent
pH	7.94±0.44	7.07±0.31	7.95±0.34	7.12±0.27	7.89±0.35	7.11±0.25	7.95±0.36	7.18±0.31	-	-	-	-	-	-	-	-
BOD5 (mg/L)	521.03±25.61	291.83±26.32	607.8±29.15	365.49±29.11	680.07±23.34	427.77±23.6	740.6±60.14	529.7±38.54	43.95	39.86	35.61	28.38	41.42	38.54	33.87	26.61
SBOD (mg/L)	379.23±33.55	221.83±25.46	468.73±27.17	288.06±28.82	508.74±20.18	339.87±22.6	603.11±59.99	441.56±38.3	41.42	38.54	33.87	26.61	47.3	42.65	36.87	29.63
TCOD (mg/L)	1,613.21±203.6	850.61±121.13	1,902.85±71.3	1,091.47±105.79	2,085.87±64.37	1,316.66±57.5	2,406.06±254	1,691.72±194	44.52	40.22	34.94	27.46	44.52	40.22	34.94	27.46
SCOD (mg/L)	1,337.01±197.8	741.5±127.31	1,639.37±69.2	980.3±104.93	1,759.85±62.87	1,152.66±56.5	2,107.24±254	1,526.25±193	25.19	21.64	16.89	12.66	25.19	21.64	16.89	12.66
NH3 (mg/L)	11.82±2.16	8.81±1.51	15.49±2.88	2.09±2.46	17.44±3.75	14.52±3.3	15.64±3.87	13.68±3.49	29.15	25.57	21.65	16.74	29.15	25.57	21.65	16.74
PO4 (mg/L)	2.27±0.46	1.58±0.25	2.34±0.49	1.74±0.42	2.09±0.37	1.63±0.3	2.38±0.4	1.98±0.36	46.13	35.92	29.82	21.24	46.13	35.92	29.82	21.24
Phenol (mg/L)	176.52±12.81	95.13±12.73	272.84±8.92	174.72±10.57	373.71±14.32	262.28±15.6	466.63±15.46	367.29±10.9	-	-	-	-	-	-	-	-
Volumetric Loading (g BOD ₅ /m ³ d)	104.2	-	121.54	-	136.01	-	148.12	-	-	-	-	-	-	-	-	-
TCOD/TBOD	3.09	2.92	3.13	2.98	3.06	3.08	3.25	3.19	-	-	-	-	-	-	-	-
ORP																

concentration of 100 mg/L (93.58%) after five days and warm weather, which is more than phenol removal by the UV/TiO₂ process and less than the RBC biological system. Considering the fact that the anaerobic ponds in all different phenol concentrations (except 100 mg/L) is not by itself capable of removing the organic pollutants up to permissible standards of discharge into the environment, it must be employed as pre-treatment and subsequently the anoxic and facultative stabilization pond must be used.

The results showed that in cold conditions the efficiency of anaerobic pond in oil wastewater treatment with different concentrations of phenol is relatively low. This can be related to the low growth activity of microorganisms and slow reaction rate of decomposition of dissolved materials by them. Besides, phenol as part of organic compounds forming BOD₅ and COD is dissolved into solution and lacks the potential of sedimentation in an anaerobic pond; this finding is consistent with Almasi and Pescod [24]. On the whole it can be concluded that anaerobic stabilization ponds, if properly operated, show favorable performance in removing the organic compounds at different concentrations of phenol in warm temperatures.

Conclusions

Just as seen in the results, maximum removal of NH₃, PO₄, phenol, TCOD, SCOD, TBOD, and SBOD were 61.08, 70.09, 93.58, 80.18, 78.89, 78.7, and 76.84%, respectively. That was obtained in phenol concentration of 100 mg/L and high temperature. Climate conditions affected pond treatment performance so that system efficiency was minimum at low temperature. Considering the advantages of a WSP system such as flexibility, simplicity of operation, and relatively high efficiency, it can be used as a better alternative in comparison of other expensive and complex systems. Since the WSP removal efficiency for phenol and phenolic compounds was better in comparison with conventional biological treatment methods, it can be concluded that anaerobic pond systems as an option with proper cost-effectiveness can be employed for treatment of petrochemicals and oil refinery phenolic wastewaters.

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