Removal of petroleum hydrocarbons from wastewater using photolysis- moving bed biofilm reactor hybrid system

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Abstract

In this study, we investigated the effectiveness of both single and hybrid systems, incorporating UV photolysis and a Moving Bed Biofilm Reactor (MBBR), for treating synthetic wastewater contaminated with petroleum hydrocarbons. Petroleum hydrocarbons pose significant environmental threats due to their high toxicity, stability, accumulation potential, and resistance to biodegradation. In the hybrid system, the wastewater underwent chemical treatment first and then was introduced into the biological reactor. For the photolysis system, we explored the impacts of different concentrations and various radiation powers of UV-C lamps. Optimal conditions were determined to be a Chemical Oxygen Demand (COD) of 350 mg/L and a radiation power of 80 W. In the MBBR system, various concentrations were introduced into the reactor, achieving a maximum removal efficiency of 85% for an initial COD of 1000 mg/L over 72 hours with a 50% filling capacity. In the hybrid system, we achieved a remarkable hydrocarbon removal efficiency of 99% after 123 hours. Although the operational time of the hybrid system was relatively long, it demonstrated itself as a suitable treatment process compared to other conventional methods for removing these challenging, hard-to-biodegrade compounds.

key words

UV-Photolysis- Moving Bed Biofilm Reactor

Petroleum hydrocarbons

TPH-COD model

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1 Introduction

High levels of toxic multi-cyclic aromatics and petroleum-based wastewater are generally harmful [32] and may cause significant damage to the water resources and human health [2, 17], have to be treated before discharging to the environ-ment [25, 37]. In this case, miscellaneous processes including bio-logical systems have been applied [13, 30]. The most convention-al system is activated sludge [24] which is being employed in most of the Iranian refineries [36]. Moreover, dif-ferent problems such as sludge bulking and foam-ing in the suspended systems [21] the attached-growth processes have been considered for treating various types of wastewaters in recent years [7, 15, 20, 26, 28]. Due to the least deficiencies and restrictions, uses of these kinds of processes have been proven as a re-liable method for the removal of different waste-water contaminants [29, 27]. Petroleum-based products have also had suitable capability for treating wastewater. For instance, the use of the Moving Bed Biofilm Re-actor (MBBR) system has led to over 73 percent COD removal at different loading rates for treat-ing the waters produced in the oilfields [10]. Also, over 80 percent COD removal for phenol and hydroquinone with concentrations of 700 to 1000 mg/L have been reported [4]. 90 percent of the initial COD of 2000 mg/L including aniline was removed using MBBR af-ter three days by Delnavaz et al. [9]. In Brazil, wastewater from a crude oil refinery was treated by a three-stage system. MBBR as the first stage had a COD removal efficiency of 70 to 90 percent [33].

In recent years, more attempts have been made toward developing innovative treatment methods with higher removal efficiency by many research-ers. Although the biological systems can remove a wide variety of soluble organics, there are many hard-degradable compounds in the wastewater of some industries that cannot be removed effectively [6, 11, 34]. Therefore, different hybrid systems have been investigated for better treatment [1, 22, 31, 33, 38].

In recent years, the use of UV-LED technolo-gy for water and wastewater treatment process-es has increased noticeably [19]. For this research, a photolysis system was utilized for the degradation of hard degradable compounds into biodegradable com-pounds as a form of pretreatment for the biological system. UV photolysis and photo-initiated oxidant ions have a proper potential for the inactivation of microorganisms and destruction of a wide variety of contaminants in an aqueous medium [35] which the outcomes of the re-search are the evidence of this claim. For instance, by application of UV-C and without any catalyst, Yang et al. [39] could decrease the concentration of paracetamol at an initial concentration of 4mM up to 12% within 12.5 days from the liquid me-dium. Furthermore, the application of ultraviolet rays led to the degradation of riboflavin contained in water. By conducting some research, Hirahara et al. [14] as well as Minamidate et al. [23] stated that UV-C can degrade pesticides remaining in water. However, very little research has been conducted on petroleum hydrocarbon wastewater treatment through UV treatment and existing re-search was no success [12]. The use of a hybrid system using biological reactors and UV treatment for petroleum hydrocarbons is also new due to its very low biodegradability [16].

In this research, the capability of individual pho¬tolysis systems and MBBR were investigated to treat wastewater containing petroleum hydrocarbons.

2 Material and Methods

In this study, a plastic rectangular cube pilot (L: 24 cm, W: 17 cm, H: 9 cm) and a plexiglass cylindrical pilot (H: 50 cm, ID: 10cm, an operational volume of 5 L) were used as photolysis and biological systems, respectively (Figure 1).

As shown in Figure 1 (a), to prevent harmful emission of UV-C rays and also ascend the effect of radiation on removal efficiency by reflection, the internal and external walls of the reactor were covered by aluminum foils.

50% of the biological reactor was filled with Kaldness-K1 packings (Figure 1 (b)). An aquarium aeration pump manufactured by RESUN Company (AC-9906) was used for supplying the required oxygen and moving the packing. To prevent deviation of the biofilm carriers from the reactor, a sieve was placed inside the reactor.

To have a similar composition as the Tehran Refinery effluent, a mixture of gasoline (C_{16}-C_{20}) and crude oil (C_8-C_{37}) by the ratio of 1 to 2 was prepared and aerated for 48 hours. So, the lighter compounds with water vapors were stripped and removed from the medium and the heavier hydrocarbons (> C_{35}) remained on the reactor walls.

Given the use of synthetic wastewater and the

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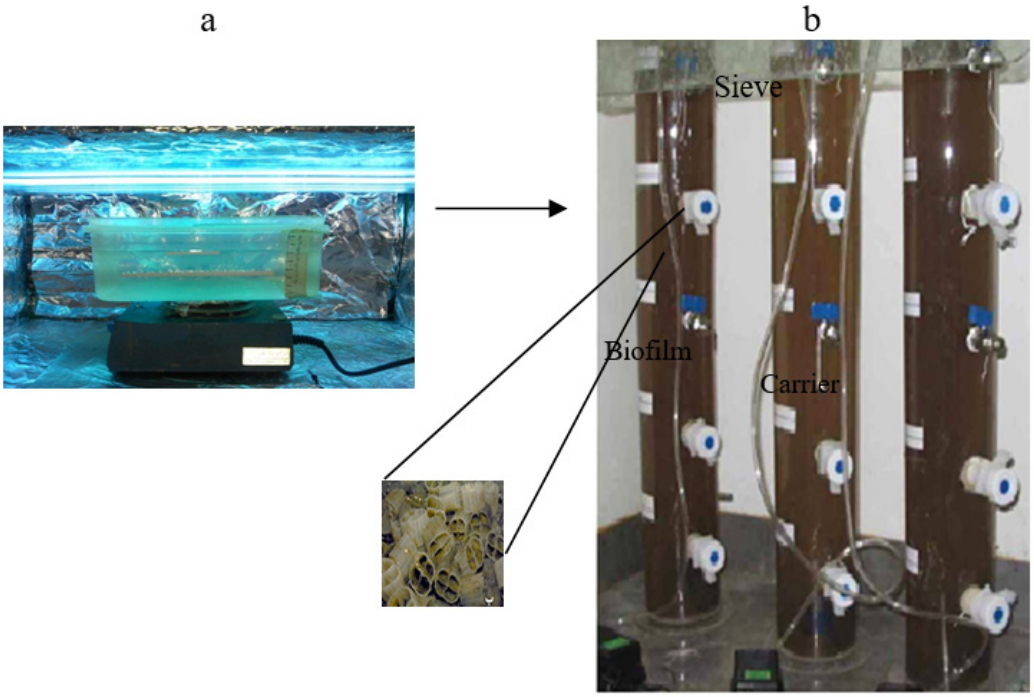

Figure 1. Studied Pilot a) Photolysis system; b) MBBR system

hard degradability of some petroleum hydrocarbons, the adaptability stage of the microorganisms is of great significance (Delnavaz et al. 2009). To prepare synthetic feed from glucose ($C_6H_{12}O_6$-H_2O) as a source of carbon, ammonium bicarbonate (NH_4HCO_3) and potassium dihydrogen phosphate (KH_2PO_4) were used as the sources of nitrogen and phosphorus, respectively to adjust the ratio of 100:5:1 of COD: N:P- existing in the feed. Furthermore, magnesium sulfate ($MgSO_4.7H_2O$), calcium chloride ($CaCl_2.2H_2O$), ferric chloride ($FeCl_3.6H_2O$), copper sulfate ($CuSO_4.5H_2O$), potassium iodide (KI), manganese chloride ($MnCl_2.H_2O$), sodium molybdenum ($Na_2MoO_4.2H_2O$), zinc sulfate ($ZnSO_4.7H_2O$), cobalt chloride ($CoCl_2.7H_2O$) and boric acid (H_3BO_3) were used as micronutrient for making synthetic wastewater (Qaderi et al. 2011, Dale & Biggar., 2008, Kishida et al. 2006).

In the photolysis system, the same wastewater was exposed directly to a UV-C ray, and concentration parameters (50, 100, 250, 350, and 500 mg/L), as well as radiation power (60, 80, 100, and 120 W), were examined. First, the optimum concentration was determined given a radiation power of 60 W and later given the optimum concentration, the optimum radiation power was determined. The parameters of radiation power and the concentration were optimized according to the minimum energy consumption by the industry as determined earlier. Reactor temperature as a controlling parameter was monitored and an insignificant vaporization from the surface of the sample was observed and this vaporization was compensated by distilled water before sampling.

During MBBR operation, about one-third of the volume of the bioreactor was filled by the sludge prepared by the returned flow of the activated sludge tank of the Ekbatan wastewater treatment plant, and the remained volume of the bioreactor was filled by water and glucose solution with COD of 100 mg/L. During the adaptation stage of the microorganisms, COD equal to 100 mg/L (TPH equal to 0- 27/4 mg/L) was injected into the system. During the first loading of contaminant compounds into the reactor, the shares of organic loads of hydrocarbon compounds and glucose were selected as 10% and 90%, respectively. After reaching a stable removal efficiency, 9 other loadings were tested, progressively reducing the proportion of glucose to zero, by steps of 10%.

During the operation period, input organic load with COD equal to 1000 mg/L by a stepped increase of 200 mg/L during each loading period and COD equal to 2500 mg/L by a stepped increase of 500 mg/L during each loading period were injected into the reactor.

To examine the percentage of petroleum hydrocarbons removed by a hybrid system consisting of a photolysis process and moving bed biofilm reac-

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tor (MBBR), some tests were conducted and the optimized parameters of the systems were studied and assessed accordingly. As mentioned earlier, after the determination of optimum conditions for two systems operated in parallel, the hybrid system was studied under optimum conditions for two systems. In a hybrid system, a sample of synthetic wastewater as prepared earlier was exposed to ultraviolet ray radiation. It was intended that the output optimum parameters of this system get close to the input parameters of the biological system and then the sample was injected into the MBBR system and examined.

2.1 Sampling and Test Analysis

During the direction period of the MBBR system and to control the biological reactions under aerobic conditions as well as to provide a suitable medium for the microorganisms, pH parameters, and dissolved oxygen were measured and controlled daily and in this way, input feed and optimum conditions for better growth were determined. To regulate an appropriate pH as necessary for growing the bacteria (range of 7±0.2), sodium hydroxide (NaOH) and sulfuric acid (H_2SO_4) were employed.

For infiltration of the samples, a membrane filter of thickness of 0.45 micrometer was used and then solution parameters were measured. To measure the COD parameters, a spectrophotometer, DR4000 (Model: Carry50) was employed. To measure TPH, TOG/TPH Analyzer (Model: Infracal) was used. A digital pH meter, Metrohm (Model: 691) was employed to measure the pH of the solution. COD reactor manufactured by HACH company (Model: DRB200) was applied for COD measurement. A digital scale manufactured by Kern company (Model: PLS360-3) with, an accuracy of 0.001g was employed for weighing the materials. A DO meter manufactured by HACH company (Model: HQ30d) was used for measuring dissolved oxygen and a magnetic stirrer manufactured by Ika company (Model: RH-Basic2) was employed. To measure COD, the Closed-reflux method was applied as per Directive 5220B [3].

3 Results and discussion

3.1 Photolysis System

For this study, the contact surface of radiation and the distance of the sample were assumed as constant, and the effect of UV radiation individually as well as the effect of concentration on the efficiency of contaminant removal in the Photolysis process were examined.

3.2 Determination of Optimum Concentration

The results acquired during the investigation of the effect of UV light individually at different CODs and radiation power of 60 W are given in Figure 2. Initially, a progressive trend of COD level was observed which may be attributed to the nature of the hydrocarbon-rich contaminant and in the following and after some time (within some days), this chart reached a maximum value of COD corresponding to input concentration for each COD concentration.

As shown in the chart, by increasing the concentration of the contaminant, the radiation period of the UV rays to meet constant removal efficiency will be increased. So, given the CODs of 50, 100, 250, 350, and 500 mg/L, the radiation period of UV is 5, 6, 9, 11, and 14 days, respectively. Furthermore, by raising the concentration of the contaminant, the removal efficiency declined under a steady state. Maximum values of removal efficiency of the contaminant due to radiation of UV of 60 W for the above-mentioned CODs are 50, 37, 28, 25, and 16%, respectively. Increasing the COD value as shown in the chart may be attributed to the degradation of hard degradable petroleum compounds into other petroleum compounds.

For determining of optimum concentration, the period of tests and consumed energy shall be cost-effective. Energy consumption against removal of each unit of COD has been given in chart 2. As shown in the chart, the lowest values of energy consumption, i.e. COD equal to 350 mg/L are related to the stirrer and the lamp which is regarded as an optimum concentration in this research.

3.3 Determination of Radiation Power of Optimum UV

The effect of radiation power on COD removal is indicated in figure 4. As shown, by increasing the radiation power and the number of photons emitted, the efficiency remained almost constant after a long time.

To determine the optimum power, the variations of energy consumption per COD at different UV powers are shown in Figure 5. According to the figure, a radiation power of 80 W with energy consumption of 0.24kWh/mg/L was considered as the optimum amount because of the insignificant difference with the results of 100 W.

3.4 Results for Biological System

The outcomes for COD removal efficiency

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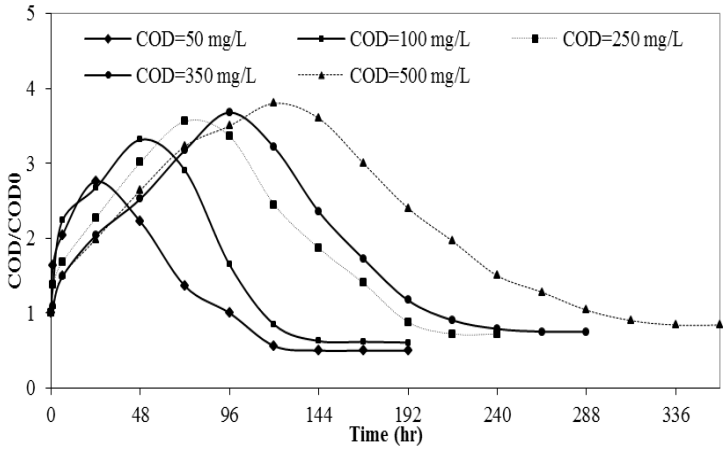

Figure 2. Variation of COD removal in photolysis system (COD0= 50, 100, 250, 350 and 500 mg/L; Puv= 60 W)

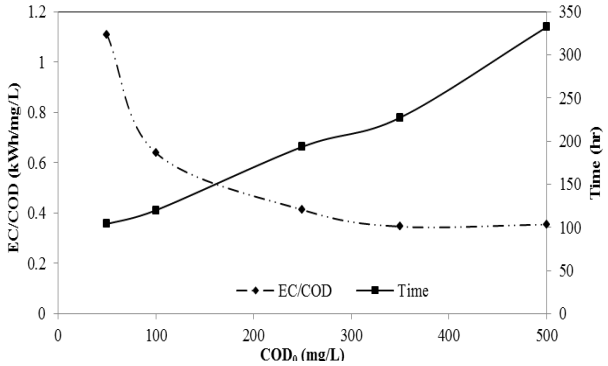

Figure 3. Energy consumption at different initial CODs (COD0= 50, 100, 250, 350 and 500 mg/L; Puv= 60 W)

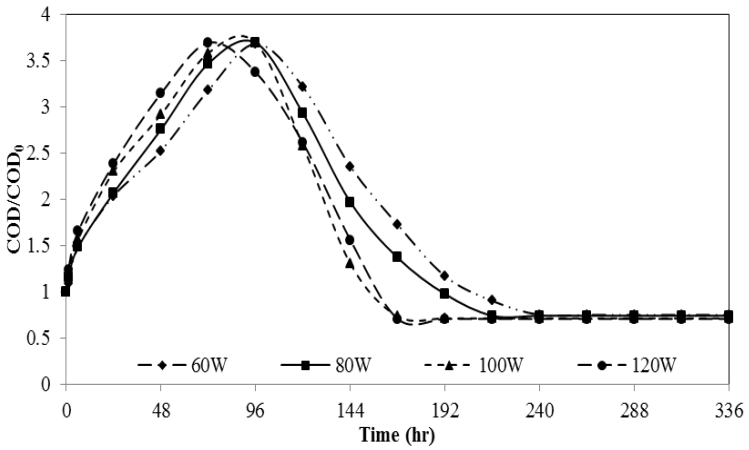

Figure4. The effect of different UV radiation powers at COD of 350 mg/L (COD0 = 350mg/L; PUV= 60, 80, 100, and 120 W)

during the loading stage at COD of 200 to 2500 mg/L (TPH of 52 to 400 mg/L) are given in Figure 6. As indicated, the maximum removal efficiency of hydrocarbon compounds has reached 85 percent within the resident time of 72 hours at a COD of 1000 mg/L (TPH= 270 mg/L). As shown, the removal trend of organic compounds from the end of the adaptation stage (COD= 100 mg/L to COD= 1000 mg/L) at resident times of 8, 12, and 24 hours has been changed between 62 and 48 percent. In some research conducted on the MBBR system, the removal efficiency for aniline, phenol, and hydroquinone have been reported 50, 65, and 55 percent 24 hours, respectively (Delnavaz et al. 2009,

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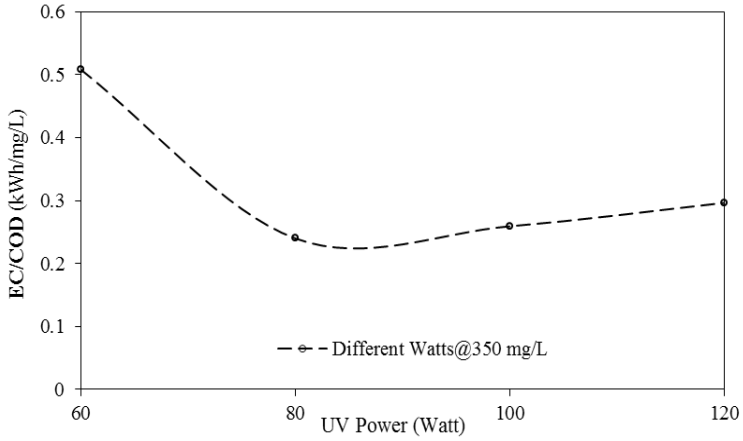

Figure 5. Energy consumption level against different powers (COD0 = 350mg/L; PUV= 60, 80, 100 and 120 W)

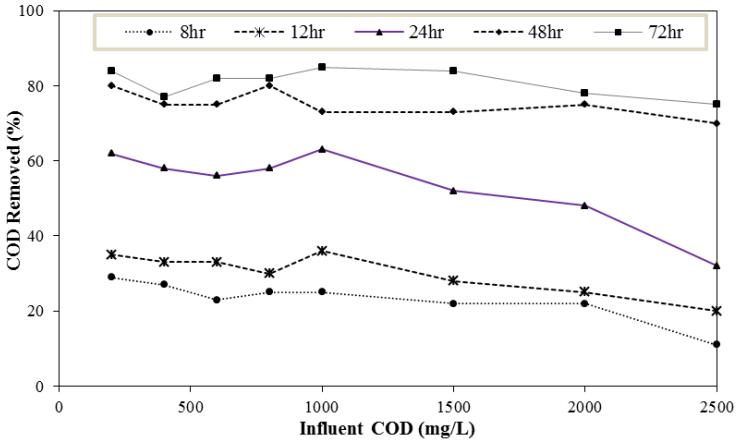

Figure 6. Changes in COD removal efficiency against different CODs

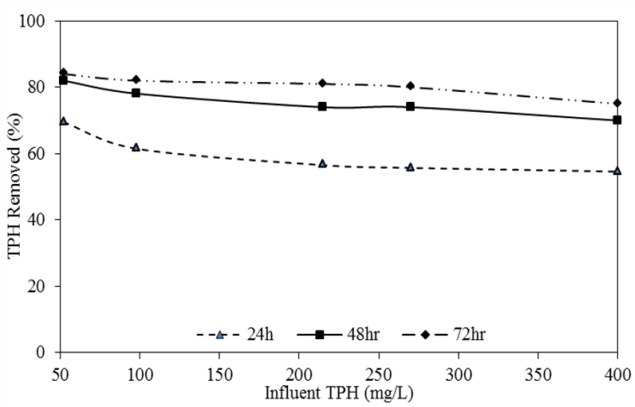

Figure 7. Changes in TPH removal efficiency at different TPHs

Ayati, et al. 2007).

According to Figure 7, a moderate trend for the efficiency decrease from 84% at TPH=50 mg/L to 75% at TPH=400 mg/L was observed at the resident time of 72 hours. The ratio of COD to input TPH is 3.82 and the range of ratios of COD to output TPH at resident times of 24, 48, and 72 hours are 3.09-4.80, 9.00-13.89 and 10.80-15.63, respectively which the results have been given in Table 1.

3.5 Hybrid Process

To study the capability of a hybrid process on hydrocarbon removal, the tests were designed at the optimum condition of both systems. In all tests related to photolysis, the input pH of the system was

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Figure 5. Energy consumption level against different powers (COD0 = 350mg/L; PUV= 60, 80, 100 and 120 W)

Input TPH (mg/L)	COD (mg/L)	Input COD/ TPH	Output COD/TPH		
			24(hr)	48(hr)	72(hr)
52	200	3.84	4.80	13.89	15.63
98	400	4.08	4.45	12.48	15.19
215	800	3.72	3.60	10.02	13.71
270	1000	3.70	3.09	9.12	11.85
400	1500	3.75	3.97	9.00	10.80

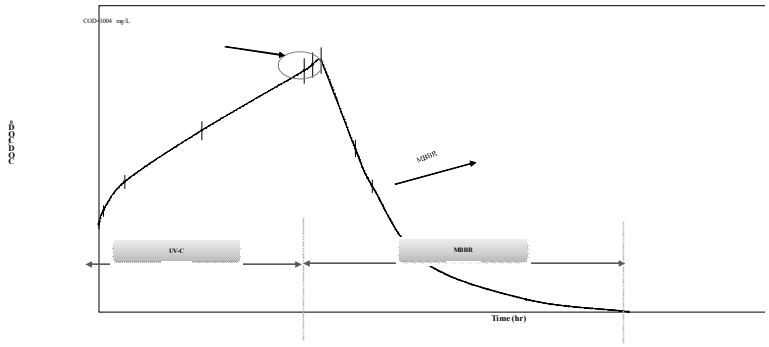

Figure 8. COD variation in hybrid system

Table 2. Comparison of the effect of the photolysis process on the removal percentage of the biological process (Concentration= 1000 mg/L)

Resident Time	8(hr)	12(hr)	24(hr)	48(hr)	72(hr)
MBBR	25	36	62	73	85
Hybrid	35	50	80	95	99

equal to 7 and since the output pH of the photolysis process (approximately 6.5) is similar to the input pH of the biological reactor, the determination of the optimum pH parameter for this process was found unnecessary.

The removal efficiency variation at an initial COD of 350 mg/L in the hybrid system is given in Figure 8. These conditions are the best fit for the photolysis system and in terms of the results obtained the ratio of COD/COD0 will be increased up to 52 hours by increasing the resident time. Thus, at this resident time, COD/COD0 of 0.35 was reached and its equivalence COD was equal to 1004 mg/L. Given the results related to the MBBR system, such a COD value is almost equal to the optimum input COD of a biological system. Then, treatment of the contaminant was completed at the moving bed biofilm reactor. As indicated in the chart, 80% of the contaminant was removed within 24 hours by the MBBR part of the system and a removal efficiency of 99% was met by the MBBR system at the resident time of 72 hours. The standard output COD in Iran is 60 mg/L (periodically) and 100 mg/L (instantaneously). Ultimate COD resulting from this system is 50 mg/L which is lower than its standard.

According to the results obtained, the removal efficiency of the contaminant in the hybrid system at a resident time of 12 hours after entering wastewater into the biological system is 14% which is greater than of removal efficiency in the MBBR system. The trend of COD removal from the wastewater in biological systems (individually or hybrid) is given in Table 2.

As shown, due to the degradation of cyclic compounds of petroleum contaminant during the pho-

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tolysis process and more degradable simple compounds existing in the wastewater entered into the biological system, the total efficiency of the hybrid system has been acceptably increased and treatment of the wastewater of greater COD has been feasible.

4 Conclusion

Parameters like high toxicity, stability, ability to accumulate in the organisms, and longer viability in the environment, made petroleum hydrocarbons a serious threat to the environment. The effectiveness of Photolysis and Moving Bed Biofilm Reactor (MBBR) for the treatment of petroleum hydrocarbons-laden synthetic wastewater has been studied in this paper. In the photolysis system, the impact of different concentrations (50, 100, 250, 350, and 500 mg/L), as well as the power of distinct radiations of UV-C bulbs (60, 80, 100, and 120 W), were studied. A concentration of 350 mg/L and a strength of radiation of 80 W were obtained as optimum values, respectively. In the MBBR system, CODs equivalent to 200-2500 mg/L (range of TPH between 52 to 400 mg/L) were injected into the reactor at different resident times, and subsequently efficiency of disposal of TPH and COD was determined. Maximum efficiency of removal for COD equivalent to 1000 mg/L by 85% during the resident time of 72 hours and filling capacity of 50% was reached. The removal efficiency of petroleum hydrocarbons reached 99% after 5.1 days by a hybrid system. Although the operation time of the hybrid system is too high, this system can be considered an advanced and suitable treatment process in comparison with other methods for the removal of hard degradable compounds of petroleum hydrocarbons in research. More research needs to be conducted for the application of LED-UV coupled with advanced oxidation for more complicated wastewater.

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