

Treatment of petroleum refinery effluents by a hybrid system of activated sludge and rotation biological reactor, followed by the sand filter

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Abstract

In this study, a novel hybrid treatment system was designed to increase the removal efficiency of petroleum refinery wastewater. The hybrid system is a pilot scale including an activated sludge combined with a rotating biological contractor (RBC) and sand filter. Four vertical rotating polyurethane disks in the aeration tank combined activated sludge-rotating biological contactor pilot. The influent wastewater for this system was the effluent from the DAF unit in the Shahid Tondgooyan Oil Refining Co's wastewater treatment plant. The rotation rate of disks and retention time has been evaluated for their impact on the removal efficiency of total dissolved solids (TDS), dissolved oxygen (DO), pH, total suspended solids (TSS), turbidity (TU), chemical oxygen demand (COD), biochemical oxygen demand (BOD), ammonia (NH₃). According to the results, enhancing the rotational speed of disks (from 4 rpm to 8 rpm) and increasing the retention time (from 6 hours to 10 hours) can improve COD, NH₃, TSS, BOD₅, TU, TDS, and oil removal efficiency of this system to 100, 98.52, 84.21, 100, 99.25, 13.32 and 100% respectively. Escalating the rotational speed beyond 8 rpm had reverse effects on the performance of this hybrid system. The rotational speed of 8 rpm and a retention time of 10 hours were the optimum conditions for removing the abovementioned parameters. It is worth mentioning that the high TU removal efficiency of the system was due to the presence of a sand filter. This system performed well in removing pollutants compared to other biological wastewater treatment systems.

Keywords: Oily Wastewater, Rotating Biological Contractor, Activated Sludge, Sand Filter.

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1. Introduction

Water is one of the most important resources used in petroleum refinery. The amount of water used in the refining process is estimated to be two to three times that of crude oil [1], and the wastewater produced contains oil, suspended solids, heavy metals, dissolved organic matter, chemical substances, and dissolved gases. [2]. Due to their difficult degradation and high biological toxicity, these pollutants are problematic [3]. The discharge of oil refinery wastewater into the environment without treatment not only poses a danger to human health and the environment [4] but also prevents the industry from recycling water. As a result, efficient treatment methods are needed to treat oily wastewater. In recent years, various chemical, biological, and physical treatment methods have been proposed to reduce and eliminate organic and inorganic pollutants from water [5-7] also an extensive studies has been conducted focusing on petroleum refining wastewater [4, 8, 9]. Due to inefficiency of most of proposed approaches in treatment of various kinds of pollutants in oil refinery wastewater, a combination of treatment approaches is required [10, 11]. For example, Seyyedi and Ayati proposed a novel and innovative wastewater remediation approach by which a lower energy consumption and higher treatment efficiency was achieved relative to the conventional systems [12].

In petroleum refinery industries such wastewater treatment methods as coagulation, flocculation, adsorption, membrane, and chemical oxidation are employed as a pretreatment, but the problem is that these approaches are not able to eliminate small suspended oil particles and dissolved elements lonely, and here the biological methods as a secondary treatment can be a solution for this challenge [13]. Under biological treatment methods, remaining dissolved oils and organic pollutants will be degraded by microbial activities[10, 14]. Biological wastewater treatment processes are classified as either attached growth or suspended growth. activated sludge process (ASP), rotating biological contactor (RBC), sequencing batch reactors (SBRs), and membrane bioreactors (MBRs), powdered activated carbon treatment (PACT) process, continuous stirred tank bioreactor (CSTB) are among various biological wastewater treatment methods[15].

The activated sludge process has been widely used for oil refinery wastewater treatment because of its high efficiency, cost-effectiveness, and environmental friendliness [16]. In this system, suspended bacterial biomass (activated sludge) takes the responsibility of elimination of pollutants. Depending on the application, this method can remove nitrogen, phosphorus, and organic carbon substances [17].

Rotational biological contractor represents an attached biological growth approach for treating wastewater from petroleum refineries. This method involves immersing nearly 35–45% of the circular discs in wastewater to form an active thin layer of microorganisms known as a biofilm [18]. RBC systems use rotating discs to promote oxygen transformation, reducing the need for additional aeration. Also, this system has a low energy consumption rate, is easy to maintain and operate, contains a high concentration of biomass, and has a short hydraulic retention period [19, 20]. The main application of this system was for the nitrification and denitrification of organic matter due to the steady and decent growth of the dominant microorganism on the biofilm [18].

Sand filters are one of option as a tertiary treatment system in petroleum refineries to meet the allowable suspended solids level of 15 mg/l. As the effluent after biological treatment still contains 25–80 mg/l of suspended solids, sand filters can be used to solve this problem. This system involves passing wastewater through a filter bed with filter media [19]. Sand filters are able to eliminate a wide range of contaminations in wastewater and thus promotes removal efficiencies of wastewater treatment systems concerning disinfection as well as esthetic aspects.

Employing sand filtration after biological treatments has shown the capacity to reduce BOD₅ and TSS to 10 mg/L [21].

Due to the low chemical oxygen demand (COD) and nutrient levels (N) in petroleum refinery wastewater, sensitive bacteria in conventional activated sludge (CAS) are exposed to toxic levels of inorganic and organic compounds, and this process lacks the efficiency necessary to satisfy the discharge criteria. [22]. Hence, a hybrid system of activated sludge, RBC, and sand filter was tested for the treatment of petroleum wastewater of Shahid Tondgooyan Oil Refining Co, Tehran, Iran. This study evaluated the performance of this hybrid system in removing COD, NH₃, TSS, BOD₅, TDS, oil, and TU in operational conditions including hydraulic retention time (HRT), and disk rotational speed.

2. Experimental procedure

2.1. Wastewater sampling and characterization

The influent of our hybrid system in this research was taken from the effluent of the dissolved air flotation (DAF) unit of Shahid Tondgooyan Oil Refining Co, Tehran, Iran. Before designing the pilot plant, accurate analyses of the influent were conducted over a period of four months. Oil and petroleum are separated by the paddles in DAF units in such a way that the effluent from this unit almost contains no oily substances suspended in it. DAF effluent characteristics and contents varied due to various operational conditions within the petroleum refinery as well as different executive conditions within the triple API and DAF units. Table 1 shows the general characteristics of wastewater.

Settlement tank and sand filter were hired after the adoption period.

Table 1. Wastewater characteristics

Parameter	Maximum Value	Minimum Value	Average Value
pH	8.2	7.1	7.6
COD (mg/l)	330	122	226
BOD ₅ (mg/l)	83	30	56.5
NH ₃ (mg/l)	15	3.2	3.9
TSS (mg/l)	67	28	47.5
TDS (mg/l)	2100	1305	1702
DO (mg/l)	1.5	0.2	0.85
Oil (mg/l)	87	23	55
TU (NTU)	29.9	17.7	23.8

2.2. Pilot plant

This study was carried out on a hybrid biological pilot plant under continuous-flow conditions at Shahid Tondgooyan Oil Refining Co. The hybrid system contains four distinct sections: a feed storage tank, a combined activated sludge lagoon with RBC, a settlement tank, and a sand filter for tertiary treatment, see Fig. 1. Fig. 2 illustrates the three zones of treatment: contact, stabilization, and settling tank. In Figure 2, the experimental set-up is shown, and Table 2 shows the configuration information of the pilot plant.



Fig. 1. The hybrid system of activated sludge, RBC, settlement tank, and sand filter

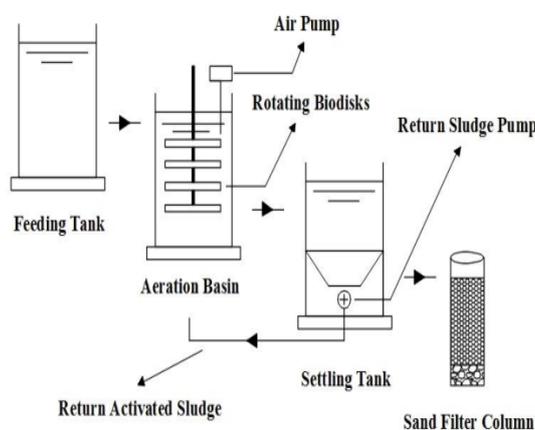


Fig. 2. Configuration of the pilot plant and the process used in this study.

Table 2. Configuration of pilot plant

	Length (cm)	Width (cm)	Height (cm)	Radius (cm)	Volume (L)
Feeding tank	-	-	96	32	300
Aeration Tank	35	35	40	-	50
Settlement tank	30	16	25	-	12
Sand filter column	-	-	90	16	18

We installed a feed storage tank above the pilot plant, and the wastewater flowed gravitationally toward the combined activated sludge tank. As it is shown in Fig.2, or the combination of activated sludge and RBC methods, an aeration tank was used. At each end of a covered aerobic unit with a 50-liter working volume, a vertical shaft passed through four fully immersed Plexiglas acrylic discs with a 25-centimeter diameter mounted on a bearing. The layers of polyurethane foam (PUF) between bio discs provided a suitable environment for the growth of microorganisms and organic matter degradation. High porosity and specific surface area make PUF an ideal media [23]. Table 3 presents the characteristics of RBCs used in this study.

Table 3. Characteristics of rotating biological contactors used in this study

Parameter	value
Number of disks	4
Diameter of disks (cm)	25
Thickness of disks (cm)	4.5
Surface material	PUF
Spacing between disks (cm)	5
Total surface area of disks (m ²)	0.4
Porosity of disks (%)	85
Disk submergence (%)	100

Compressed air with a flow rate of 30 L/min was injected through a porous diffuser located at the bottom of bioreactors to control substrate feeding rates and aerate wastewater. Additionally, the lid of the aeration tank had some 8-centimeter holes for air circulation. The temperature of the wastewater was adjusted at 30°C using two aquarium heaters with temperatures ranging from 25°C to 35°C. Trapezoidal settlement tanks are designed to accelerate sludge and suspended solids settlement, and a pump is provided at the bottom of the settlement tanks to return settled sludge to aeration basins.

Following the biological treatment section, there is an 18-liter settlement tank, which is designed based on the aeration tank's outlet flux. We then used a slow sand filter as a tertiary treatment to enhance the treatment process. The sand filter is constructed in cylindrical form; there are two layers of gravel for drainage, a 30-centimeter layer of sand for filtration improvement, and a 40-centimeter layer of 98% pure silica sand with a uniformity coefficient of 1.7 and an effective size of 0.18 mm. There are three ways that sand filters remove pollutants; the first is through physical transport to the grain's surface, the second is through electrostatic attraction, mass attachment, and adhesion, and the third is through collection around the grain [24, 25].

2.3. Experimental Set-up and experimental procedure

With the installation of rotating contractors, the aeration basin was partially filled (50% of the whole aeration tank capacity) with return activated sludge collected from the aerobic basin at Shahid Tondgooyan Oil Refining. An overview of the characteristics of returned activated sludge can be found in Table 4. The remaining working capacity of the combined aeration system was filled with wastewater collected from the DAF unit of the petroleum wastewater treatment plant. During the first week, no influent was entered into the aeration basin, and DO, pH, and temperature were measured every day to ensure they were within standard ranges. At the end of the first week, the reactor start-up involved daily adding wastewater and activated sludge to the aeration basin to feed microorganisms with required organics and nutrients, and biomass grew on the discs for 25 days. During this adaptation period, temperature and pH varied from 25°C to 30°C and 6.5 and 8.5, respectively, and dissolved oxygen (DO) and COD of the wastewater were checked. The results showed that COD concentrations in aeration basins increased after 7.5 hours.

It is due to the decrease in mixed liquor volatile suspended solids (MLVSS) that this phenomenon occurs. According to the wastewater flow rate and the dimensions of the settling tank, the HRT of the aeration tank was 2.5 h during start-up and 10 h for the whole hybrid system. A settlement tank and sand filter were hired after the adoption period. Influent wastewater was injected into the aeration basin with a 100 mL/min flow rate from the 1000-liter feed tank, and it was tested to determine its characteristics. Wastewater flows through the

settlement tank after being treated biologically in an aeration basin. For tertiary treatment, a small percentage of the settled sludge was returned to the aeration tank, and the wastewater was passed through a sand filter. The aeration tank combines activated sludge and RBC to remove contaminants through both suspended growth and attached growth. Furthermore, both biological and physical-chemical processes play a role in the removal of contaminants in sand filters [26].

In this study, the influences of HRT (9.6, 12.8 and 16 h) and rotational speed (4, 8, 12 rpm) on the efficiency of the hybrid system were assessed in two steps. In addition, the rotational speed of disks was determined according to similar research by VGKhondabia et al. [27]. In the first step, different HRTs were applied under fixed rotational speed. In the next step, considering the optimum HRT from the previous step, the effect of disk rotational speed on the reactor performance was investigated.

Table 4. Characteristics of return activated sludge

Parameter	value
Temperature (°C)	34
COD (mg/L)	259
volatile suspended solids (MLVSS)(mg/L)	240
mixed liquor suspended solids (MLSS)(mg/L)	1142
pH	7

2.4. Analytical method

Secondary effluents (after clarification) and tertiary effluents (after filtration) were analyzed statistically. In this study, COD, BOD₅, DO, TSS, TDS, MLSS, MLVSS, temperature, TU, pH, oil, and NH₃ were evaluated. TSS, MLVSS, MLSS, BOD₅, DO, and oil were measured according to APHA standard methods [28]. A digital pH meter was used to measure temperature and pH. NH₃ and COD concentrations were measured using a spectrophotometer (Loviband laboratory spectrophotometer). The TDS was measured with an AZ8371, and the turbidity was measured with a PC CECKIT Loviband. The removal efficiency of pollutant concentrations was calculated accordingly using the formula given below [29]:

$$R\% = \left(1 - \frac{C}{C_0}\right) \quad (1)$$

where C_0 and C are concentrations of wastewater before and after treatment in the hybrid system.

3. Results and discussion

To assess the effect of HRT and rotational speed of RBC's discs on the efficiency of this system in the removal of COD, NH₃, TDS, TSS, BOD₅, TU, and oil, this study was conducted in two stages. It is worth mentioning that pollution levels vary from day to day.

3.1. Effect of Hydraulic Retention Time.

HRT is one of the most influential parameters in biological adsorption. To evaluate the effect of HRT on the removal efficiency of pollutants, three different HRTs of 6, 8 and 10 h were applied aeration basin. The corresponding HRTs are 2.16, 2.88, and 3.6 h in sand filter, and the total corresponding HRTs are 9.6, 12.8, and 16 h in the hybrid system. The results are shown in Fig. 3.

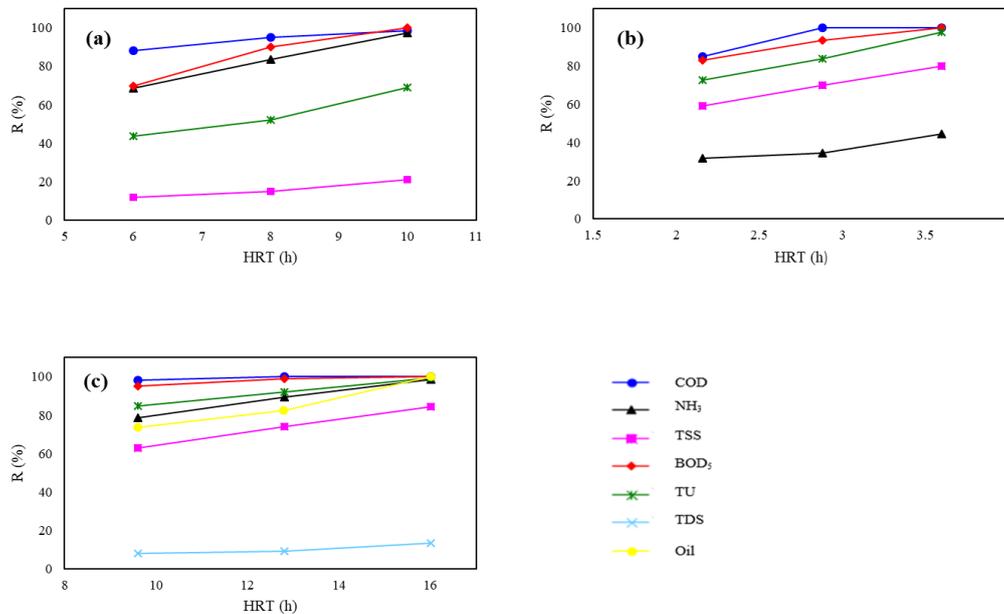


Fig. 3. pollutants removal efficiencies in the (a) aeration basin, (b) sand filter, (c) and hydrate system vs HRT at 4 rpm

3.1.1. COD and BOD₅ removal

As shown in Fig. 3, with increase of HRT, COD and BOD₅ removal efficiencies enhanced in each reactor individually and consequently in the hybrid system. When HRT is increased, the contact time between substrate and microorganisms and the number of microorganisms on the aeration basin increases [30, 31], and microorganisms in the combined activated sludge have more time to engage in biological activities. In addition, in the settling tank, sludge has more time to settle. As the aeration basin is a combination of activated sludge system and RBC, increase of HRT can provide an opportunity for better oxygen transfer [32]. [32]. As HRT increased from 9.6 to 16, COD removal efficiency of the settling tank, sand filter, and hybrid system increased from 83.23 to 100%, 85 to 100%, and 98.23 to 100% at 4 rpm, and these results demonstrated the effectiveness of this hybrid system in COD removal. Yoong and Lant [33] used SBR for oil refinery wastewater treatment and achieved 97% COD removal efficiency at HRT of 10 h. Our hybrid system had better performance in COD removal compared to the SBR system. With an increase in HRT, microorganisms were given more time to be active, which caused BOD₅ removal to rise from 94.99 to 100%.

3.1.2. TSS removal efficiency

TSS is removed primarily by the settling tank and sand filter in the hybrid system. According to Fig. 3, the longer the HRT is, the more efficient the settling tank and sand filter are in TSS removal. Longer HRT allows sludge to settle more slowly, thus creating this result. 84.21% of the TSS in the wastewater was removed by the hybrid system.

3.1.3. NH₃ removal efficiency

As shown in Fig.3 and 4, a greater HRT resulted in better NH₃ removal by the combined activated sludge, sand filter, and hybrid system because microorganisms had more time at their disposal. At 4 rpm, the combined activated sludge, sand filter, and hybrid system removed NH₃ by 97.35, 44.44, and 98.52%, respectively. In a study, Ghalekhondabi et al. [20] removed 98.89% of ammonia from refinery wastewater at an optimum HRT by a four-stage RBC. A comparison reveals that our hybrid system was as efficient as Ghalekhondabi et al.'s system in ammonia removal.

3.1.4. Turbidity removal efficiency

Based on the results, the sand filter is the most influential part of the hybrid system for removing turbidity. As HRT increases, wastewater flows slowly through sand filters, improving their efficiency in removing TU. Additionally, aeration plays a key role in the removal of average-sized particles through the degradation of organic substances, so, with the increase of HRT, the efficiency of the aeration basin in TU removal was enhanced [34]. 99.25% of the TU of the wastewater was successfully removed by the hybrid system.

3.1.5. TDS removal efficiency

The hybrid system removed 13.32 % of the TDS from the wastewater. The TDS removal efficiency of the hybrid system was very low, and this minor amount was mainly removed by sand filter. Increase of HRT had negligible effect on TDS removal efficiency as this system is not suitable for TDS removal.

3.1.6. Oil removal efficiency

As shown in Fig. 3, the hybrid system had significant ability to remove oil and its efficiency reached 100%, thereby meeting the requirements of USEPA for the concentration of oil or total organic components (TOC) in drinking or treated water of 2 mg/L [35]. According to the results, 16 hours of HRT led to the best performance of the hybrid system. In activated sludge and RBC, higher HRTs provide a ground for more effective aeration of wastewater and longer contact time between microorganisms and organic compounds. This condition had a hand in improvement of the efficiency of the hybrid system in oil removal. Our hybrid system represented 100% oil removal efficiency in HRT Of 16 h, which is higher than the oil removal efficiency (88%) of an anaerobic baffled reactor (ABR) for treating heavy oil produced water used by Ji et al [22].

3.2. Effect of the rotational speed of discs

Another contributing factor to the system's removal efficiency is the rotational speed of the discs in the RBC reactor. System efficiencies and biofilm activity are affected by rotational speed. Essentially, by adjusting the rotational speed of the discs, the thickness of biofilm and dissolved oxygen can be controlled, and oxygen and nutrients can be transformed into microorganisms in the biofilm [36].

To assess the effect of this parameter on the efficiency of the hybrid system, three different rotational speeds were examined (4, 8, 12 rpm). The rotational speed effect on reactor performance in COD, NH₃, TSS, BOD₅, TU, TDS, AND oil removal is represented in Fig. 4.

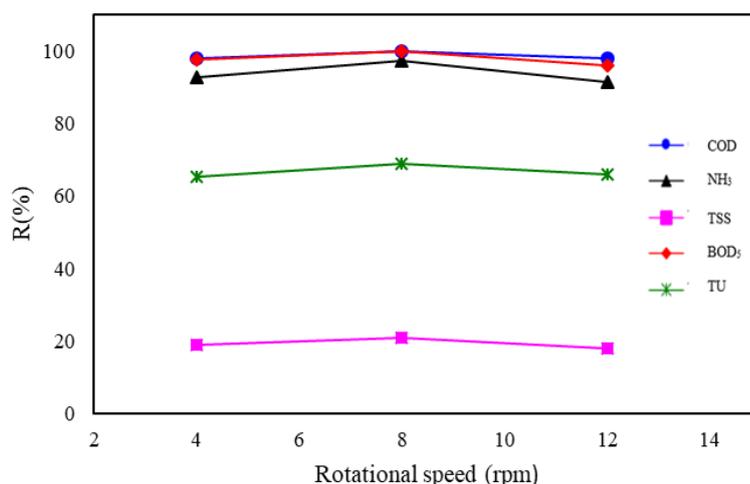


Fig. 4. pollutants removal efficiencies in the aeration basin at HRT=16 h.

3.2.1. COD and BOD₅ removal

The removal efficiencies of aeration basin increased from 98 to 100% for COD and 97.5 to 100% for BOD₅ with the increase of rotational speed from 4 to 8 rpm. The main reason was that dissolved oxygen (DO) mounted to the higher values, more turbulence and better flow mixing was occurred, and higher contact ration between substrate and microorganism were provided when the rotational speed increased [37]. On the other hand, increase of the rotational speed from 8 to 10 rpm had reverse effect, and COD and BOD₅ removal efficiency fell from 100 to 97.85% and 1 from 100 to 96 respectively. This was the direct result of increase of the fluid shear stress over the mass and the detachment of the produced biofilm [30]. It is worth mentioning that in low rotational speed, the process of oxygen transfer for the formation of biofilm may face difficulties [38]. Hence, in our research rotational speed of 8 was the most suitable speed that prevented from all of the mentioned problems. Najafpour et al. [39] assed the performance of a three-stage aerobic RBC reactor for COD removal at rotational speed varying between 3 and 11 rpm. They were successful to remove 93.7% COD in rotational speed of 11 rpm, and the better performance of our hybrid system is obvious compared to Najafpour et al.'s study.

3.2.2. TSS removal efficiency

According to the results, increase of rotational speed from 4 to 8 rpm ended up with increase of TSS removal efficiency from 19 to 21%, but this trend is reverse when rotational speed rose from 8 to 12 rpm, and the TSS removal efficiency diminished from 21% to 18.05 %. Detachment of biomass at high rotational speed may be responsible for this result. As the detached biofilm remains suspended and does not easily settle, the TSS removal efficiency decreased [40].

3.2.3. NH₃ removal efficiency

Fig. 4 indicates that when discs rotational speed increases from 4 to 8 rpm, ammonia removal efficiency increase from 92.6 to 97.35 %. Increase of rotational speed made a better mixing process, and as a result, microorganisms adsorb more oxygen and nutrients. On the contrary,

with further increase of rotational speed to 12 rpm, ammonia removal efficiency of aeration basin declined to 91.3% because of a rise in biofilm detachment rate. To be more precise, the thickness of viscous biofilm diminishes and the region with higher shear stress expands, and consequently ammonia removal efficiency decrease [20].

3.2.4. Turbidity removal efficiency

According to the Fig. 4, TU removal efficiency followed the same trend as TSS removal efficiency and at high speed biofilm detached from discs, and this caused decrease in TU removal efficiency [41]. Irfan et al. [40] used RBC reactor to remove TU from wastewater. In this study, they showed that increase of rotational speed from 30 to 50 rpm caused the decrease of TU removal efficiency from approximately 82 to 71%. While, the TU removal efficiency of the aeration basin in our study was lower than that of Irfan et al, our total hybrid system had better performance and was able to remove 99.3% of TU from the refinery wastewater.

3.2.5. TDS removal efficiency

Just like previous step, the TDS removal efficiency of this hybrid system was not considerable and was not being affected with the rotational speed's changes.

3.2.6. Oil removal efficiency

In a study, Khondabi et al. [1] used a RBC to treat petroleum refinery wastewater. In this study, ammonia, COD, BOD₅, TDS, TSS, and TU respectively were under the 40, 89, 87, 76, 85, and 58%, in the optimum conditions. Our integrated system with COD, BOD₅, NH₃, and TU removal efficiencies of 100, 100, 98.52, 99.25% showed better efficiency compared that system.

4. Conclusions and prospective

The activated sludge combined with rotating biological contractor and sand filter was successful in COD, NH₃, TSS, BOD₅, TU, and oil removal. The optimum HRT and rotational speed of discs were 16 h and 8 rpm, respectively, and in this optimum condition COD, NH₃, TSS, BOD₅, TU, and oil removal efficiencies were 100, 98.52, 84.21, 100, 99.25, and 100%. Increase of HRT positively affected biological performance, and with increase of HRT, the pollutants removal efficiencies were enhanced. There was a direct relationship between rotational speed and the pollutants removal efficiencies when rotational speeds varied between 4 and 8 rpm, but at disk rotational speeds higher than 8 rpm, biofilm detachment occurred because of increased shear rate resulting in decreased biological performance and loss of microbial biomass. Due to the presence of sand filter in the structure of this hybrid system, this system was managed to remove turbidity more effectively compared to such other systems as single RBC and activated sludge. This work addresses problems regarding conventional activated sludge systems in wastewater treatment of petroleum refineries. Adding an RBC to the aeration tank increases the removal efficiency without needing to rebuild a new treatment system, making it very cost-effective.

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