



A Hybrid Activated Sludge-Moving Bed Biofilm Reactor System for the Treatment of Petroleum Refinery Effluents

Nuha Muhsen Ali¹, Eynas Muhamad Majeed²

^{1,2} Middle Technical University, Polytechnic College of Engineering Specializations – Baghdad, Iraq.

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ABSTRACT

A novel hybrid biological treatment system was developed to enhance the efficacy of effluent from petroleum refineries, as demonstrated in this experimental study. In the proposed pilot system, activated sludge is mixed with a sand filtering unit and a moving bed biofilm reactor (MBBR). There was a mix of attached and suspended microbes in the hybrid reactor, which consisted of an aeration tank containing activated sludge and polyurethane biofilm carriers. Shahid Tondgooyan Oil Refining Company's dissolved air flotation (DAF) discharged water was used to collect the influent wastewater. Researchers looked into how treatment efficacy was correlated with carrier mixing intensity and hydraulic retention time (HRT). Quantities of total dissolved solids (TDS), dissolved oxygen (DO), pH, total suspended solids (TSS), turbidity (TU), chemical oxygen demand (COD), biochemical oxygen demand (BOD₅), and ammonia (NH₃) were among the water quality metrics examined. The results demonstrated that expanding the HRT length to 6 to 9 hours and increasing the carrier mixing intensity to a moderate range improved the system's carrier mixing intensity. The COD, NH₃, TSS, BOD₅, TU, and TDS removal efficiencies were approximately 98.6, 97.1, 82.4, 99.2, 98.7, 15.1 and 99.4 percent, respectively, when working circumstances were good. In contrast, increasing the mixing intensity to even higher levels reduced treatment efficiency slightly, probably due to biofilm dissociation and decreased microbial stability. Following a 9-hour HRT and a medium carrier mixing regime, the optimal operating conditions were identified. The major reason for this was the inclusion of the sand filtering unit, which greatly improved the effectiveness of turbidity removal. It appears that the hybrid system of sludge-MBBR-sand filter might be utilized to treat effluent from petroleum refineries, as it outperformed conventional biological treatment methods in eliminating a variety of contaminants.


1. Introduction

Among the most important resources that are used during petroleum refinery processes is water. It is estimated that the amount of water involved in the refining processes is two to four times the quantity of the refined crude oil [1]. As such, petroleum refineries produce large volumes of oil and grease, suspended solids,

heavy metal, dissolved organic solvents, chemical additives, and dissolved gases wastewater [2]. These contaminants pose severe treatment problems since they are not biodegradable, and their toxic effects on the body are also very high [3]. The untreated wastewater released by refineries into the environment does not only pose a threat to human health and the aquatic life in the

Corresponding author E-mail address: inas.mohammed@mtu.edu.iq
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environment [4], but also restricts water reuse in the industry. Thus, it is necessary to develop effective and robust technologies of the treatment of oily wastewater.

Over the last few years, there has been a big variety of chemical, physical, and biological treatment techniques that have been explored to eliminate organic and inorganic contaminants in industrial wastewater [5 -7]. There are a great number of research studies devoted to petroleum refinery effluents due to their multifaceted structure and variable nature [4, 8, 9]. Nonetheless, the ineffectiveness of most standalone treatment methods to eliminate various pollutant groups has attracted the increased attention to the integrated and hybrid treatment systems [10, 11]. As an example, Seyyedi and Ayati proposed a new type of remediation that was more efficient in terms of its treatment and consumed less energy than the traditional systems [12].

Rather common processes used as pretreatment steps in petroleum refineries include coagulation/flocculation, adsorption, membrane separation, and chemical oxidation. However, such methods cannot be used alone to eliminate small oil drops and dissolved organic substances [13]. Biological treatment processes have also been extensively used as secondary treatment processes in this regard, and in which residual organic pollutants and dissolved hydrocarbons are broken down by the activity of microorganisms [10, 14].

There are broadly applied biological wastewater treatment systems, which are suspended-growth and attached-growth processes. Typical ones are the activated sludge process (ASP), sequencing batch reactors (SBRs), membrane bioreactors (MBRs), powdered activated carbon treatment (PACT), continuous stir tank bioreactors (CSTBs), and moving bed biofilm reactors (MBBRs) [15]. The activated sludge process is one of them, and it has been widely used in treating petroleum refinery wastewater because of its relatively high efficiency, flexibility of operation, low costs, and environmentally friendly nature [16]. During this process, the removal of organic matter, nitrogen and phosphorus compounds are

achieved through suspended microbial biomass based on the operating conditions [17].

Moving bed biofilm reactor (MBBR) is a sophisticated attached-growth biological treatment technology which utilizes free floating plastic carriers with high specific surface area of biofilm proliferation. In contrast to fixed biofilm systems, MBBR carriers are freely moving in the reactor when there is aeration or mechanical mixing, which facilitate the even distribution of biomass and enhanced mass transfer [18]. Some of the benefits of this technology are increased retention of biomass, shock load resistance, small reactor design, decreased sludge generation, and consistent operation in nitrification [19, 20]. MBBR systems have been shown to be used successfully to eliminate organic and ammonia content in industrial and petroleum wastewater as a result of the well-established and diverse microbial communities on the biofilm carriers [18].

Tertiary filtration units are usually used to polish and meet the discharge standards after biological treatment. In petroleum refineries, multimedia or dual-media filters (e.g., anthracite sand filters) are common to achieve the removal of residual suspended solids to less than 1020 mg/L since effluents of biological units usually have 3090mg/L suspended solids in them [21]. Such filtration systems promote the clearness of the effluent, effectiveness of disinfection, and aesthetic quality of the treated wastewater. Past research has shown that BOD 5 and TSS levels can be brought to a lower range of less than 1015 mg/L after the first biological filtration; hence, the discharge or reuse of the effluent is possible [22].

The comparatively low nutrient levels and toxic organic and inorganic chemicals found in the petroleum refinery wastes cause the inhibition and low treatment efficiencies of the conventional activated sludge systems [23]. Thus, hybrid systems that incorporate activated sludge and MBBR along with the tertiary filtration have become more and more popular as the potential solutions to the refinery wastewater treatment.

The current research paper examines the performance of a hybrid activated sludge-MBBR system and subsequent multimedia filtration of

the petroleum refinery wastewater of the Al-Dorra Oil Refinery, Baghdad, Iraq. Their removal efficiencies were determined at different operating conditions such as hydraulic retention time (HRT) and biofilm carrier mixing intensity, such as chemical oxygen demand (COD), ammonia (NH_3), total suspended solids (TSS), biochemical oxygen demand (BOD₅), total dissolved solids (TDS), oil, and turbidity (TU).

2. Experimental Procedure

2.1. Wastewater Sampling and Characterization

Influent wastewater that was used in this study was taken to the effluent of the dissolved air flotation (DAF) unit of the Al-Dorra Oil Refinery. To characterize physicochemically the influent variations in the quality of wastewater, physicochemical characterization was performed on the influent over a time interval of about four months before the design and operation of the pilot-scale treatment system.

The design of the DAF unit is to help eliminate free and dispersed oil fractions by the flotation method and leaves behind an effluent with insignificant suspended oil in it. Nevertheless, the nature of the DAF effluent was different under the circumstances of the refinery working and the performance of the upstream

treatment at the API separator and DAF units. In this study Table 1 shows the mean physicochemical properties of the influent wastewater. After completion of the system acclimation period, a clarification tank and a tertiary filtration unit was added.

Table 1: Properties of petroleum refinery wastewater

| Parameter | Maximum Value | Minimum Value | Average Value |
|-------------------------|---------------|---------------|---------------|
| pH | 8.3 | 7.0 | 7.6 |
| COD (mg/L) | 345 | 135 | 238 |
| BOD ₅ (mg/L) | 88 | 32 | 58.2 |
| NH_3 (mg/L) | 16.2 | 3.5 | 4.4 |
| TSS (mg/L) | 72 | 30 | 49.8 |
| TDS (mg/L) | 2250 | 1420 | 1785 |
| DO (mg/L) | 1.6 | 0.3 | 0.9 |
| Oil (mg/L) | 92 | 25 | 58 |
| TU (NTU) | 31.5 | 18.5 | 25.2 |

2.2. Pilot Plant Description

A pilot continuity hybrid biological study was carried out in an experimental experiment that is installed in the Al-Dorra Oil Refinery. The treatment regime was a combination of activated sludge-moving bed biofilm reactor (AS-MBBR) as shown in Fig. 1.



Figure 1: combined activated sludge-moving bed biofilm reactor(AS-MBBR)

The treatment process involved three operating areas, which consisted of contact, stabilization, and clarification (Fig. 2). The feed storage tank was placed above the pilot unit such

that the influent flowed into the biological reactor through gravity. The ASMBBR unit was the central treatment unit, it offered both suspended and attached microbial growth.

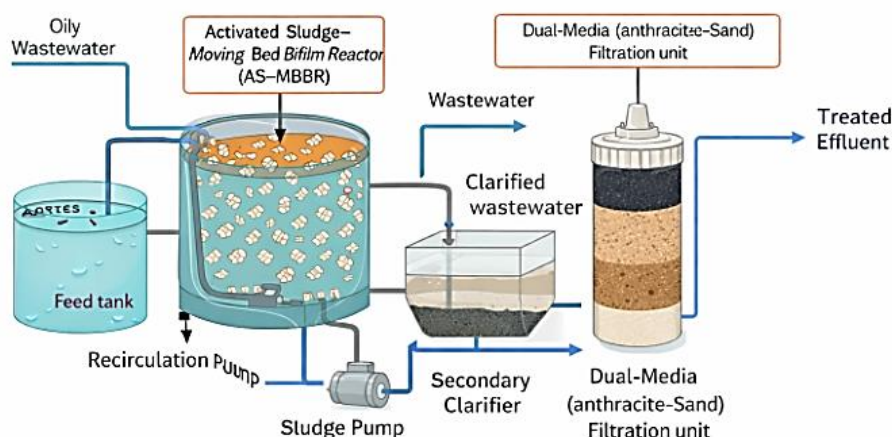


Figure 2: Design of the pilot plant and the procedure applied in this study.

The aeration tank was partially filled (estimated between 40 and 45 percent of the total reactor volume) with high-density polyethylene (HDPE) biofilm carriers of high density. These carriers also offered a large area of specific surface to promote growth of biofilms whilst being suspended freely under aeration. High porosity polyurethane-based carrier material was chosen because of its favourable biofilm adhesion property and mass transfer efficiency [23].

Compressed air was fed into the reactor as a diffuser with bubbles that were in the fine range; this was done at a rate of 25-30 L/min to transfer enough oxygen and mix the carriers. More ventilation sources were also availed in the reactor cover to ensure that there were aerobic conditions. The temperature of the wastewater was maintained at 28-30 °C by means of thermostatically controlled heaters in aquarium. In Table 2, major configuration specifications of the pilot plant are provided.

Table 2: Pilot plant set-up at Al-Dorra Oil Refinery.

| Unit | Length (cm) | Width (cm) | Height (cm) | Radius (cm) | Volume (L) |
|------------------------------|-------------|------------|-------------|-------------|------------|
| Feeding tank | – | – | 100 | 35 | 320 |
| Aeration tank (AS-MBBR) | 38 | 38 | 45 | – | 55 |
| Secondary settling tank | 32 | 18 | 28 | – | 15 |
| Dual-media filtration column | – | – | 95 | 18 | 20 |

Table 3 shows the features of Moving Bed Biofilm Reactor (MBBR) that was applied in this study.

Table 3: The properties of MBBR biofilm carriers in this experiment.

| Parameter | Value |
|------------------|---|
| Carrier type | Cylindrical moving bed biofilm carriers |
| Carrier material | Polyurethane-based polymer (PU/HDPE) |

| | composite) |
|--|------------|
| Average carrier diameter (mm) | 22 |
| Average carrier height (mm) | 8 |
| Specific surface area (m ² /m ³) | 520 |
| Effective surface area (m ² /m ³) | 420 |
| Carrier porosity (%) | 88 |
| Bulk density (kg/m ³) | 95 |
| Filling ratio (% of reactor volume) | 45 |
| Carrier submergence (%) | 100 |

2.3. Set-Up and Operating Procedure Experimental.

The aeration tank at start-up was seeded with return activated sludge that was collected at the aerobic treatment unit of the Al-Dorra refinery wastewater treatment plant. The starting mixed liquor concentration was equated to that of a working volume of say 45-50 percent of the aeration tank. Table 4 gives characteristics of the inoculated sludge. The rest of the reactor was filled with wastewater of DAF effluent.

Table 4: Features of return activated sludge utilized in this study

| Parameter | Value |
|---|-------|
| Temperature (°C) | 33 |
| COD (mg/L) | 268 |
| Mixed liquor volatile suspended solids (MLVSS) (mg/L) | 255 |
| Mixed liquor suspended solids (MLSS) (mg/L) | 1,180 |
| pH | 7.2 |

The initial 7 days period was used to run the system as batch mode without influent feeding to allow the microbial to acclimate. The pH, dissolved oxygen (DO), and temperature were checked on a daily basis during this time to maintain the operating condition. The reactor was then slowly switched to a continuous mode through the addition of wastewater and activated sludge every day to ensure the growth of microbes and biofilm on the MBBR carriers. The period of acclimation took approximately 25 days during which the pH varied between 6.8 and 8.3, temperature was kept between 26-30 °C and the concentration of DO keep at its optimum level of more than 3 mg/L.

There was a short-lived rise in the COD concentration in the initial operation which was explained by variation in the mixed liquor volatile suspended solids (MLVSS) as biomass

became adapted. The hydraulic retention time (HRT) of the aeration tank at the start-up in relation to the rate of influent flow and reactor volume was about 3 h, whereas the hydraulic retention time of the hybrid system was 9-10 h in the steady-state situation.

After the biological treatment, a stream of wastewater was directed to a secondary clarifier (20 L) where suspended solids and biomass were removed. Part of the settled sludge was recirculated to the aeration tank to ensure that there was a concentration of the biomass and the clarified effluent was sent to the tertiary treatment unit.

A dual-media filtration system was used in tertiary polishing which had an upper anthracite layer and a bottom silica sand layer provided with graded gravel as a drainage medium. This filtration unit better removed the remaining suspended solids, turbidity and related organic matter by the physical straining, adsorption and attachment processes [24, 25].

2.4. Operational Parameters

How hydraulic retention time (HRT) and the intensity of biofilm carriers mixing influence the development of system performance was assessed during two experimental phases. The HRT of 10, 13 and 16 h were tested under a constant aeration condition. After that, three carrier mixing regimes (low, moderate and high) which is equivalent to carrier velocity induced by aeration were used at the optimum HRT [26]. The intensity of excessive mixing was not reached to avoid the detachment of biofilms, according to the findings of such like studies [27].

2.5. Analytical Methods

After the clarification, secondary and after the filtration process were sampled and

analyzed. Such parameters were assessed as COD, BOD 5, DO, TSS, TDS, MLSS, MLVSS, temperature, turbidity (TU), pH, oil and grease, and ammonia (NH₃). The determination of TSS, MLSS, MLVSS, BOD 5, DO, and oil content were done by Standard Methods for the Examination of Water and Wastewater (APHA) [28].

A digital calibrated pH meter was used to measure temperature and pH. The concentrations of COD and NH₃ were measured in Lovibond laboratory spectrophotometer, whereas TDS was measured on AZ8371 conductivity meter. The turbidity was measured by the Lovibond PC-CHECKIT turbidimeter. The degree of eliminating pollutants was determined using the equation. (1) [29]:

$$R(\%) = (1 - C / C_0) \times 100 \quad (1)$$

where C₀ and C represent influent and effluent pollutant concentrations, respectively.

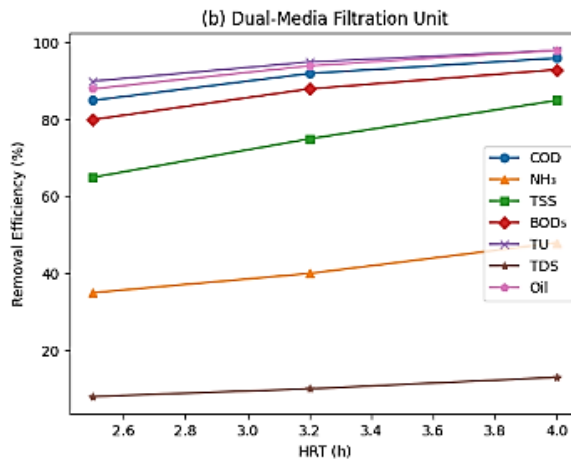
3. Results and Discussion

In order to assess the effect of hydraulic retention time (HRT) and the biofilm carrier

mixing intensity on the functionality of the hybrid activated sludge-MBBR-dual-media filtration system, the experimental research was performed in two consecutive phases. The efficiency of the removal of chemical oxygen demand (COD), ammonia (NH₃), total dissolved solids (TDS), total suspended solids (TSS), biochemical oxygen demand (BOD 5), turbidity (TU), and oil were examined. It is noted that there were daily variations in the influent pollutant concentrations because of the variability in the refinery operations.

3.1. Hydraulic Retention Time Effect

One of the most important operation parameters that have an impact on the biological wastewater treatment performance is the hydraulic retention time. Three concentrations of 10, 13 and 16 h of HRTs were introduced into the hybrid system to evaluate its effect. The biological reactor and tertiary filtration unit corresponding HRTs were adjusted and the total system HRTs were approximately 10.0, 13.2 and 16.0 h respectively. The findings are demonstrated in Fig. 3.



a)

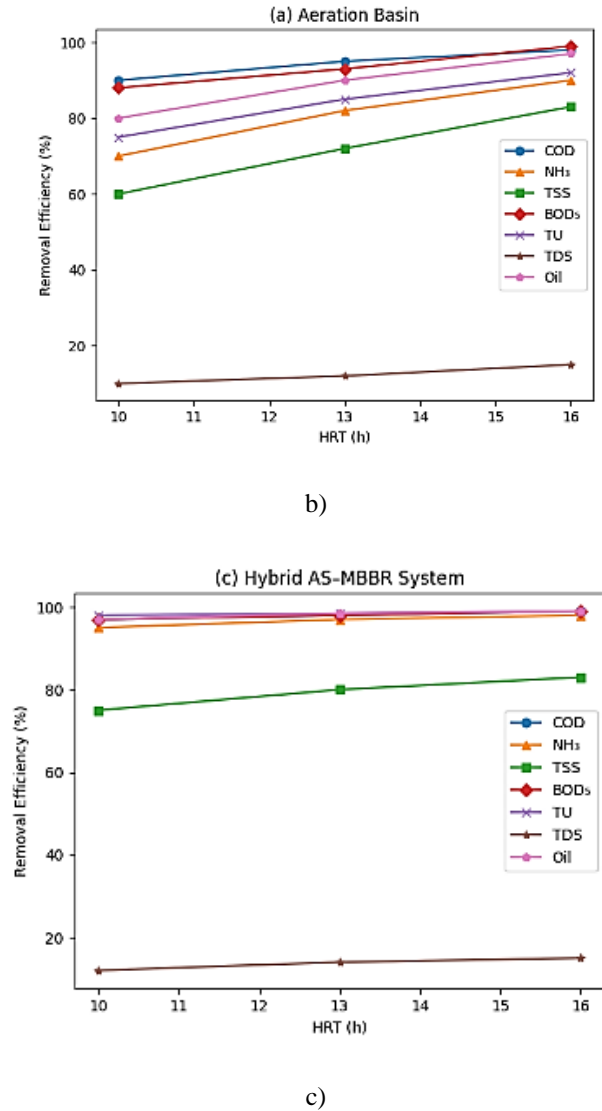


Figure 3: (a). Influence of hydraulic retention time (HRT) on the efficiencies of pollutant removal in the aeration basin (AS -MBBR). (b). Influence of hydraulic retention time (HRT) on the efficiencies of pollutants removal in the dual-media filtration unit. (c). Influence of hydraulic retention time (HRT) on the efficiencies of the total hybrid AS system in the MBBR.

3.1.1. COD and BOD₅ Removal

In Fig. 3, the addition of HRT considerably boosted COD and BOD₅ removal in the biological reactor and, therefore, in the hybrid system as a whole. Longer HRTs offer more time of exposure to microorganisms and organic substrates to biodegrade more [30,31]. Besides this, increased biomass concentration and the efficiency of oxygen transfer in the activated sludge-MBBR reactor led to better removal of organic matter [32].

The COD removal efficiency of the hybrid system rose as the total HRT (10-16 h) rose with

about 96.8 to 98.9 per cent; the BOD₅ removal rose with about 97.2 to 99.1 per cent. These outcomes prove the great efficiency of the hybrid system to remove organic pollutants. According to Yoong and Lant [33], a sequencing batch reactor (SBR) demonstrated COD removal efficiencies of about 97 percent of refinery wastewater at HRT of 10 h. The marginally better performance of the current system indicates the advantage of integrating suspended and attached growth processes.

3.1.2. TSS Removal Efficiency

The hybrid system was found to remove the TSS during the second clarifier and the dual-media filtration unit. Sludge settling and filtration efficiency were noted to be enhanced with an increase in HRT. The hybrid system was able to effortlessly remove the TSS with an average of 81-83 at the maximum HRT. The longer HRET gives time to form floc, compacted sludge and trapping of particles in the filtration media; hence, the solids removal is enhanced.

3.1.3. Efficiency of Ammonia (NH₃) Removal.

The efficiency of removal of ammonia rose significantly as HRT rose as a result of increased nitrification activity. The biological reactor and the hybrid system had the NH₃ removal efficiencies of around 96.4 and 97.8 at the optimum HRT of 16 h as well. Prolonged HRT gives good conditions to nitrifying bacteria by escalating the duration of contact of the substrates and maintaining the dissolved oxygen [20]. These findings can be compared to those found by Ghalekhondabi et al. [20] who obtained high ammonia removal efficiencies when using a multistage biofilm system in the treatment of refinery wastewater.

3.1.4. Removal Efficiency of turbidity.

The tertiary dual-media filtration unit was highly significant in turbidity removal. An augment in HRT diminished hydraulic loading rates across the filter enabling higher retention of the particles. The hybrid system had turbidity removal efficiencies of about 98.6 to 98.9 at the highest HRT. Also, there was improved biodegradation of colloidal organic matter in the biological reactor which led to low levels of turbidity before filtration [34].

3.1.5. TDS Removal Efficiency

The hybrid system was characterized by poor TSS removal capacity with average efficiencies of 12-15 and was not very dependent on HRT. Such a small decline has been largely explained by partial elimination of dissolved

solids associated with particle in the filtration unit. As anticipated, biological treatment mechanisms do not tend to work well in the dissolved inorganic salts removal and a higher HRT did not make any difference in the TDS removal performance.

3.1.6. Oil Removal Efficiency.

The removal of oil was increased with an increase in the HRT because of an increase in biodegradation and physical separation. The hybrid system had oil removal efficiencies of about 98.5 99.2 at optimum HRT of 16 h, which was equivalent to international discharge limit on treated refinery effluents [35]. An increase of retention enables better microbial degradation of remaining hydrocarbons as well as enhances separation of oil droplets during clarification and filtration. The current hybrid system performed better than anaerobic baffled reactors (ABR) that usually have a lower removal efficiency of oil below 90 percent [22].

3.2. Both Effect of Biofilm Carrier and Mixing Intensity.

The intensity of carrier mixing, which is mainly controlled by the rate of aeration, is important in regulating the biofilm thickness, oxygen transfer and mass transfer in MBBR systems. Three mixing regimes (low, moderate and high) at the optimum HRT were considered to measure its effect. The findings are given in Fig. 4.

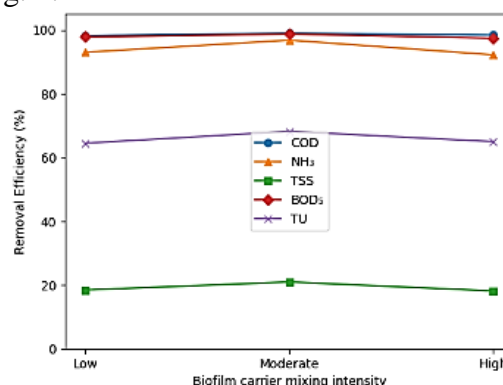


Figure 4: Efficiencies of pollutant removal in the AS-MBBR aeration basin at HRT = 16 h at high, moderate and low biofilm carrier mixing intensities.

3.2.1. COD and BOD₅ Removal

An intensification of carrier mixing between low to moderate carrier mixing levels improved COD and BOD 5 removal because of elevated oxygen exchange, enhanced substrate biomass contact, and even biofilm activity [36,37]. At moderate mixing conditions, COD and BOD 5 removal efficiencies were about 99.0% and 98.7 respectively. Nevertheless, an additional rise in mixing intensity resulted in a minor decrease in removal efficiency, which was due to too high shear stress and the partial detachment of biofilm [30]. This observation is in line with earlier researches, which showed decreased biofilm stability at high hydrodynamic stress [38].

3.2.2. TSS Removal Efficiency

The efficiency of TSS removal was low at high mixing conditions but high when there was moderate mixing. Over shear forces favored biofilm shedding, augmented suspended solids in the effluent and decreased settling effectiveness [39]. The highest TSS removal efficiency of about 22 percent in the biological unit was found to be at moderate mixing intensity.

3.2.3. Ammonia Removal Efficiency

The removal of ammonia increased to 96-97 percent when the intensity of mixing rose to moderate levels as opposed to low values indicating increased nitrification owing to increased oxygen distributions. NH₃ removal efficiency was 92 (of maximum mixing intensity) and probably associated with thinner biofilm and that the nitrifying biomass was lost [40].

3.2.4. Removal Efficiency of Turbidity.

The trend of Turbidity removal was the same as the TSS. Intermediate mixing improved degradation and aggregation of particles and extreme mixing promoted detachment of biofilm

and turbidity of effluent [41]. The hybrid system had a turbidity removal efficiency of about 98.8 99.0 percent, which was a reflection of the efficiency of the dual media filtration stage even in case of higher turbidity removal in the biological unit by high mixing intensity.

3.2.5. TDS Removal Efficiency

As is accurate with HRT experiments, mixing intensity changes did not have a significant effect on TDS removal. Mean removal efficiencies were lower than 15 which proved to be not that the hybrid biological system can be used to remove dissolved salt [20].

3.2.6. Oil Removal Efficiency

Low intensity carrier mixing led to the greatest oil removal efficiency (approximately, 99) owing to a better biodegradation and dispersion of oil droplets. Over mixing had a slight negative impact because free floating biofilm and re-entrained oil droplets enhanced effluent levels. The current hybrid system was found to have a better performance in terms of overall oil removal compared to biofilm-based refinery wastewater treatment systems that were reported before [1].

4. Conclusions.

This paper has shown that a hybrid type of activated sludge-moving bed biofilm reactor (AS-MBBR) followed by dual-media filtration is an efficient treatment plan of petroleum refinery wastewater in the Al-Dorra Oil Refinery, Baghdad, Iraq. The combined system had high removal efficiencies of major pollutants, including, COD, NH₃, TSS, BOD 5, turbidity and oil, which validate the merits of integrating suspended-growth and attached-growth biological treatment and tertiary filtration.

The optimum operating parameters were determined to be at a total hydraulic retention time (HRT) of around 16 h and an intermediate biofilm carrier mixing intensity. In such circumstances, COD, NH₃, TSS, BOD 5, turbidity, and oil removal efficiencies of 98.9, 97.8, 82.6, 99.1, and 99.0 were observed, respectively, in the hybrid system. The presence

of HRT was found to have a positive effect on the performance in biological treatment, showing consistent positive effect on microbial degradation and enhancing the sludge settling.

The intensity of carrier mixing was critical in performance of the system. The moderate mixing increased the oxygen uptake, substrate supply, and biofilm activity, which led to better elimination of pollutants. But over mixing caused higher hydrodynamic shear, biofilm detachment, and the partial loss of active biomass, which in turn caused a decrease in the efficiency of the treatment. Such results emphasize the fact that the hydrodynamic conditions of MBBR based hybrid systems are important points that need to be carefully optimized.

The presence of a dual-media filtration device greatly enhanced the removal of turbidity, and suspended solids, which are important factors of biological systems (traditional activated sludge or single stage biofilm reactor) in isolation. The step of polishing improved the quality of the effluents, and met the discharge requirements of petroleum refinery wastewater.

Practically, the suggested hybrid AS -MBBR system has a number of disadvantages over traditional activated sludge processes such as sensitivity to load changes and low nitrification rates. Notably, the retrofit of aeration tanks with MBBR carriers is an efficient strategy of upgrading at a low cost that may be undertaken without significant structural changes.

The future researches must be on the stability of the systems in the long run, nutrient recovery, energy optimization, and the possibility of incorporating an advanced oxidation or membrane-based polishing systems to improve further on the quality of the effluents and assist in the reuse of water in petroleum refinery processes.

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