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CHARACTERISTICS OF REFINERY AND CAR WASH WASTEWATERS PLUS POTENTIAL TREATMENT BY COMBINED DAF-MBBR TECHNIQUES AND REUSING

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Abstract

This study intended to characterize raw refinery and car wash wastewater samples. Samples were taken from Kewrgosk oil refinery and car wash location in Erbil City, Kurdistan Region, Iraq. Wastewater samples were analyzed for many parameters such as oil and grease, ammonia-nitrogen, total suspended solids (TSS), chemical oxygen demand (COD) and nitrite, etc. The results were compared to Iraqi standard effluent regulations. Moreover, the research aimed to suggest a suitable treatment for types of wastewaters. Detailed about dissolved air flotation (DAF) and moving bed biological reactor (MBBR) were explained and described to treat wastewater samples. Results indicated that the wastewater parameters exceed standard limits and need treatment. Results illustrated that DAF removed 70-75%, 92.5%, and 92 % of oil and grease, COD, and TSS, respectively. While, MBBR removed 99%, 55-100% and 85 of oil and grease, COD, TSS, respectively. Combined treatment of DAF and MBBR was effective to remove oil and grease and organic compounds.

Keywords: Car wash, refinery, reusing, treatment, wastewater.

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1- Introduction

An increasing number of cars on roads due to the increase in population and economic status results in an increasing the intake of water in car wash stations for washing cars. These car wash service locations produce a high amount of wastewater. If not properly treated the wastewater, it will threaten the environment. Previously, people used to wash cars in the courtyards of their houses. Nowadays, new technologies and mechanical devices are designed to implement this task (Kumar and-Chauhan, 2018). The water used for washing care contributes pollutants from the environment, car wash chemical cleanings, traffic pollutants and road pollutants. According to Lau et al. (2013), average quantity of water used to wash a car is 150 L to 600 L. The type of car wash affects how much water is used. Brown, (2002) stated water consumption variations between car wash stations in different countries. Water usage varies depending on wash equipment and schedule. According to Al-odwani et al. (2006), 185-370 liters of water are used per car wash in Kuwait when in-bay washing is used. While in Belgium due to the high capacity of washes, the average amount of 400 liters per wash water is used.

On the other hand, one of the major constituents of wastewater discharged is oily wastewater from industry. petroleum refineries from petroleum industries produce large quantity of wastewater. This wastewater is a complex of organic compounds and phenols which are toxic compounds and can remain long period of time. wastewater from refinery contains high amount of COD and oil and grease and phenols and have risk to natural environment. Based on the process configuration, wastewater quantity and characteristics are determined. About 3.5-5 m³/ton of wastewater is produced during the recycling of cooling water (Aziz and Ali, 2021).

Components of refinery wastewater depends on the type of crude oil and natural gas (Aljuboury et al., 2017). According to Jain et al. (2020) the amount of produced wastewater from petroleum refineries about 1.4 to 1.6 times the amount of crude oil produced. Generally, many methods and treatment techniques have been used for refinery wastewater treatment such as flow anaerobic sludge blanket (UASB), up-flow anaerobic fixed bed (UAFB) reactor, membrane bio-reactor (MBR), activated sludge process (ASP), moving bed bio-reactor (MBBR), anaerobic-aerobic-biofilm reactor (A/O-BR), membrane filtration, catalytic ozonation, ...etc. Petroleum effluents from refineries contain large amount of toxic substances and biodegradable compounds. Moreover, type of oil being processed and operation procedures and plant configuration can greatly affect the variation in petroleum wastewater compounds (Wang et al., 2015). Constructing many car wash stations and a huge amount of refineries in Erbil city without an appropriate treatment result in environmental problems and health risks. The objective of the present study is to characterize and treat both WWs and potential treatments by combined DAF-MBBR methods.

2. Materials and methods

2.1 . Site description

2.1.1 Car wash WW

Wastewater from a big car wash service location called (Diyari) located in Erbil city and situated at 36° 9' 23'' N 44° 0' 43'' E. Figure 1 shows the location of the car wash WW. Additionally, cars were washed manually by full hand service and large amount of fresh water was used per washed cars. Daily, tankers of fresh water were used to bring water for washing cars. it is different from car wash stations in terms of the type of washing, the approximate number of cars washed, and the amount of water used per car washed. Based on the interview with the owner of the car wash station, all the information was obtained. It required full labor intensive of cleaning. The effluents collected into a long drained canal and discharged into sewerage system of the city (figure 2).

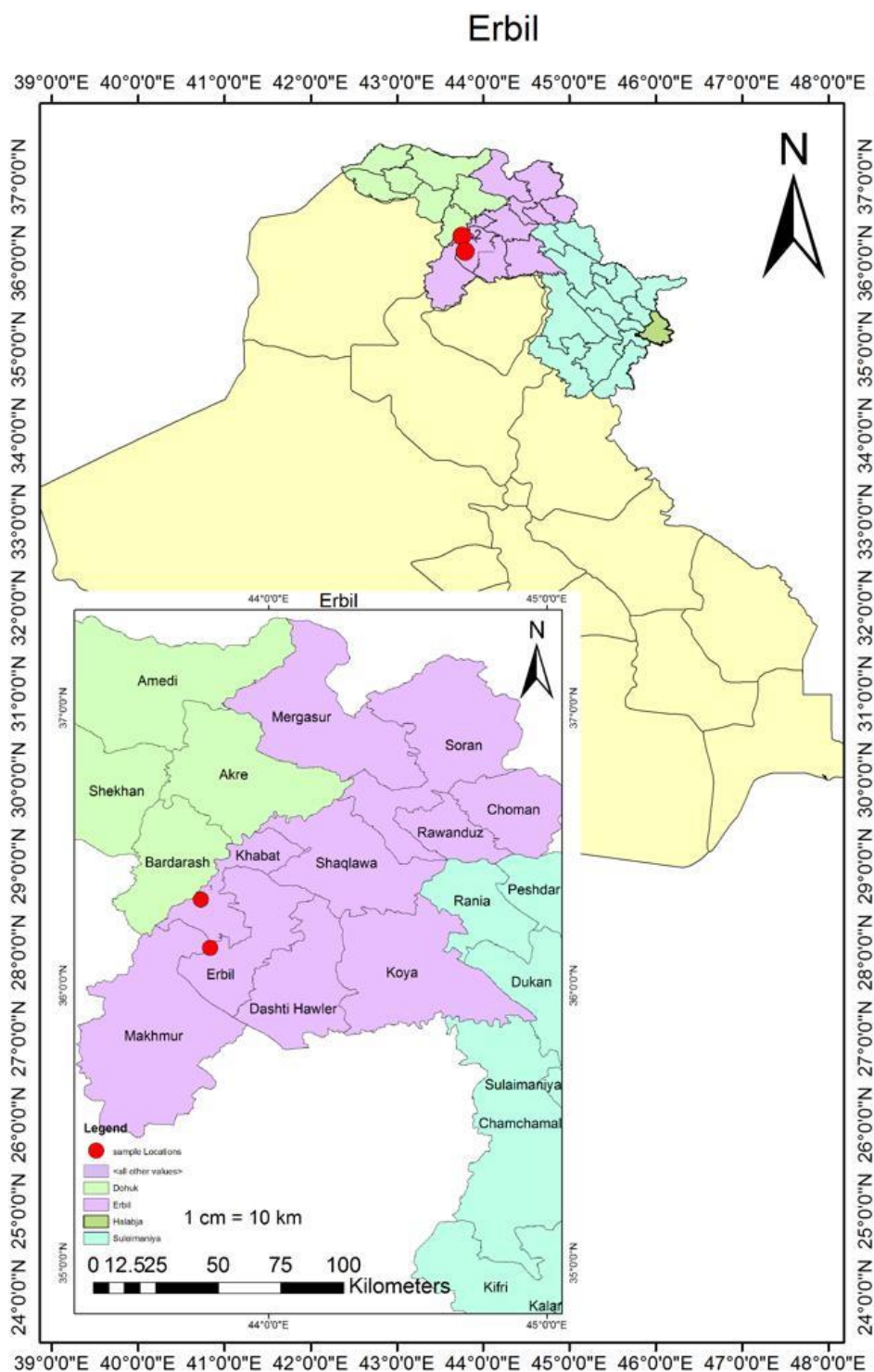


Figure 1 Map location of car wash and Kewrgosk oil refinery wastewaters

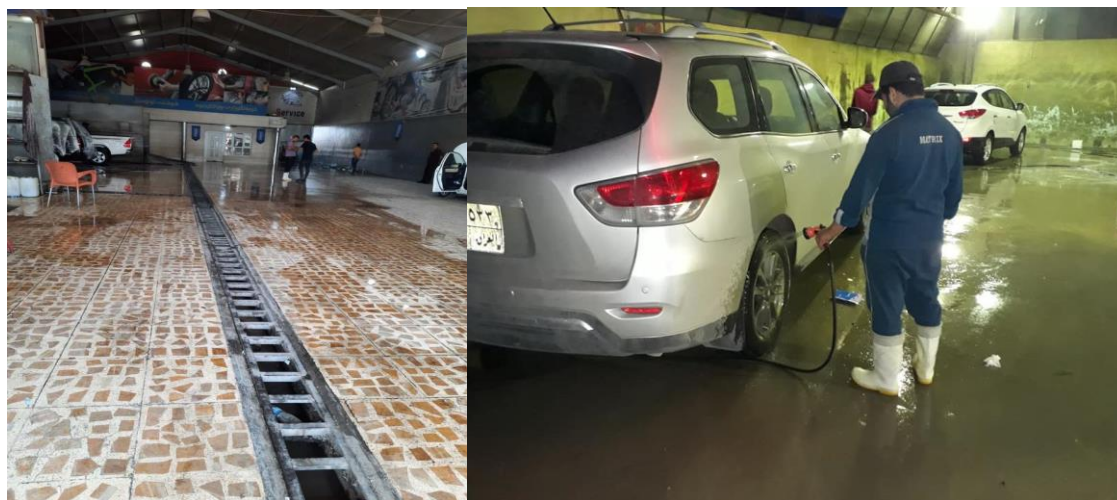


Figure 2 car wash activity at car wash station

2.1.2. KAR oil Refinery WW

Kewrgosk oil refinery located in Khabat District, near Kewrkosek Sub-District. It is situated at $36^{\circ} 8' 8''$ N $43^{\circ} 47' 23''$ E. (Figure 3). The refinery is 40 Km far from west of Erbil. And has been occupied by a land of 2.5 km². The refinery is the biggest petroleum refinery in the Kurdistan Region-Erbil, Iraq. The capacity of Kewrgosk refinery is 185,000 barrel per day. WW comes from the plant operations and the process of oil productions such as kerosene, benzene, gasoline, fuel oil and it is composed of heavy metal and various organic compounds mainly oil and grease. Large amount of wastewater is produced in the refinery. The wastewater goes to steps of physical, chemical and biological treatments. The outlet of the treated effluent discharged to greater Zab river.



Figure 3 location of Kewrgosk oil refinery

2.2 . Sampling process

Samples of WW were collected from November 2021 to April 2022. In three period of time. Samples were collected in plastic bottles and transported immediately to the laboratory and analyzed for many WW quality parameters to know their characteristics (APHA, 2012). The parameters were pH, turbidity, total dissolved salts(TDs) (mg/L), temperature (°C), total solids (TS) (mg/L), total suspended solids (TSS) (mg/L), COD (mg/L), turbidity (FTU), color (Pt.Co), electrical conductivity (EC)(μ s/cm), nitrite (NO₂) (mg/L), nitrate (NO₃) (mg/L), and oil and grease (mg/L).

All experiments were conducted in Sanitary and Environmental Engineering Laboratory, Civil Engineering Department, College of Engineering, Salahaddin-University-Erbil, Erbil, Kurdistan Region, Iraq. Figure 4 illustrates wastewater samples from car wash and Kewrgosk oil refinery wastewater.

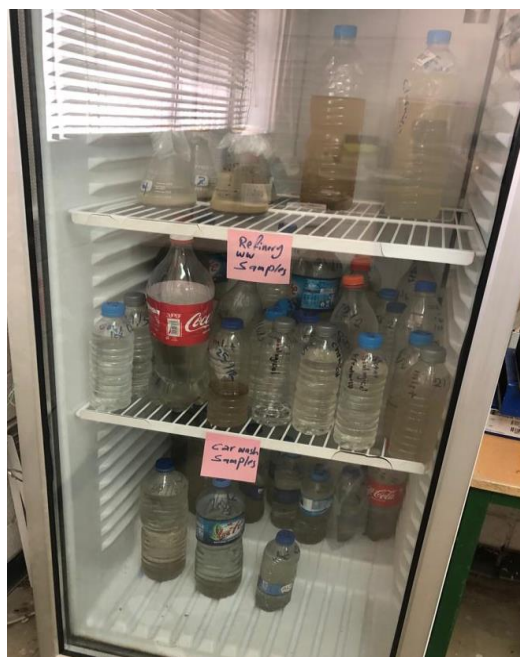


Figure 4 wastewater samples stored at 4C° in refrigerator

2.3. Analytical methods

Based on the standard method for Examination of Water and Wastewater (American Public Health Association (APHA) 2012) samples were analyzed and measured. Tests of color, COD, NO₂, and NO₃ were measured by a spectrophotometer (Hatch DR3900). pH of the samples was measured by pH meter device. Turbidity and EC were taken by HANNA Turbidity meter-LP 200 and Wissenschaftlich-Technische Werkstätten-LF-42 respectively. Additionally, separation funnels were used to determine the concentration of oil and grease inside the samples. Temperature and EC were measured by EC device. filter paper, muffle furnace, oven and digital weighing instruments were used to conduct TS and TSS tests.

2.4. DAF and MBBR Treatment Techniques

Physical, chemical and biological treatments are available for the treatment of refinery wastewater. As the most advanced and efficient method of reducing impurities and suspended particles within wastewater, dissolved air flotation (DAF) is one of the many techniques used to treat water and wastewater. By reducing the pressure that is exposed to atmospheric pressure (30-65 psi) in the DAF process, small bubbles are produced (Aziz and Ali, 2021). DAF is primarily influenced by air pressure, recycling ratio, and air to solid ratio.

3. Results and discussions

3.1. Car wash WW

Results of the car wash samples were listed in Table 1. The results were compared to the Iraqi standard for effluents. Overall, COD, oil and grease parameters, and TSS reported high concentrations of 970 mg/l, 1760mg/l and 5800 mg/l and all the samples exceeded standard limits. Details and discussions about measured parameters are given below.

3.1.1 pH

Generally, pH in car wash stations comes from maximum pH of 8.41 has slightly alkaline. The pH of WW is a critical parameter that needs to be measured. pH is optimal in the range of 7.5-8.5 because bacterial growth in MBBR is optimal at those pH values (Sosamony and Soloman 2018). pH 7 generally resulted in high removal of colour, COD, and turbidity when using alum dosage for optimum pH (Palaniandy, et al., 2010).

3.1.2 Temperature

Every chemical and biological reaction is affected by temperature. All organisms are affected either in the laboratory or in the nature by exposure to these contaminants, including bacteria needed for biological oxidation. The temperature of the samples was within the Iraqi standard for effluents. High temperature does not effect on treatment efficiency therefore; it is not necessary to raise the temperature above 20C° (Baddor, et al., 2014). Parameters such as pH, COD, total count of bacteria, BOD and EC are measured with temperature which can be seen the importance of temperature.

Table 1 Pollutant characteristics of car wash WW

No	Parameters	Unit	Oct. 2021	Oct. 2021	Nov. 2021	Dec. 2021	Jan. 2022	Feb. 2022	Mar. 2022	Apr. 2022	May 2022	Min.	Max.	Iraqi standards
1	pH		5.97	5.9	7.7	8.2	7.86	8.41	6.39	7.65	7.84	5.9	8.41	6-9.5
2	Temperature	C°	24.3	23.4	20.1	10.5	14.7	14.1	19.9	17.5	23.6	10.5	24.3	<35
3	EC	µs/cm	900	314	1007	862	895	847	651	827	986	314	1007	
4	Turbidity	FTU	167.7	319	518.8	137.55	477.625	1050	241	730	320	137.5	1050	
5	Color	ptCo	810	2440	27000	5950	2320	1630	1310	3633		810	27000	100?
6	TS	mg/L	1300	1500	2300	2500	1300	1900	727	1500	1100	727	2500	
7	NO ₃	mg/L	31	60	9	2.6	9.7	5.3	38	3.33	19.2	2.6	60	
8	NO ₂	mg/L	75	196	795	175	95	77	134	119	77	75	795	
9	TSS	mg/L	200	1000	1600	5800	1000	1400	400	2000	400	200	5800	60
10	Oil and grease	mg/L	0.4	1.34	1250	600	8	400	120	440	80	0.4	1250	Not permitted
11	TDS	mg/L	1100	500	700	430	448	419	327	414	493	327	1100	
12	COD	mg/L	480	970	788	144	423	743	482	714	162	144	970	<100

3.1.4 COD

In wastewater, COD is a measure of how much oxygen it takes for dissolved organic matter to become oxidized in the presence of a powerful oxidant and a strong acid medium. COD value in car wash wastewater ranges between 423 and 970 m/l during the period of study.

3.1.5. Oil and grease

An oil gravity separation technique is used to remove oils and grease by floatation or abrasion. After the chemical fracturing of emulsified oils, dissolved air flotation is used to remove the oil emulsion system. In all cases, oils and grease must be removed before the discharge of wastewater (Farhoud, et al., 2014). One of the most important parameters in car wash wastewater is oil and grease pollutant. In the literature less than 5 mg/l to more than 100 mg/l were reported. In this research, the minimum value of 0.4 mg/L and the maximum value of 1250 mg/L were observed. The efficiency of removal of O&G is 70-75% was achieved by Mujumdar, et al., (2020) when used five steps of integrated treatments such as Collection, Screening with O&G removal, Chemical coagulation and flocculation, sedimentation and filtration for raw car wash.

3.1.6. TDS

It is crucial to know the TDS values, especially when using treated effluent for irrigation. TDS value is proportional to the electrical conductivity of a sample. Moreover, electrical conductivity changes due to the reactions and external conditions such as pH and temperature (Abdul hakim and Farhoud, 2012). The values of TDS for car wash samples range from 327 to 1100 mg/l.

3.2- Refinery ww

Table 2 explains the results of Kewrgosk oil refinery wastewater samples. Parameters were compared to the standards for effluents. The composition of refinery effluents

No	Parameters	Unit	Oct. 2021	Nov. 2021	Dec. 2021	Jan. 2022	Feb. 2022	Mar. 2022	Apr. 2022	May 2022	Min.	Max.	Iraqi standards
1	pH		6.03	8.79	10.45	7.88	8.4	7	8.6	8.64	6.03	10.45	6-9.5
2	Temperature	C°	25	19.6	8.8	14.1	14.2	13.8	25	26.9	8.8	27.9	<35
3	Ec	µs/cm	46000	1113	1685	1493	1097	1859	1849	853	853	46000	
4	Turbidity	FTU	184.14	169.9	120.55	190.02	260	30.2	181.5	131.4	30.2	361.53	
5	Color	Pt.Co.	3030	1055	3875	1040	2010	1310	1550		1040	3875	100?
6	NO ₂	mg/L	220	225	127	68	90	223	112	83	68	740	
	NO ₃	Mg/L	12.3	95	13.8	17.6	13.9	139	75	50.5	12.3	139	
7	TSS	mg/L	10,000	400	200	1200	600	200	1400	400	200	10000	60
8	Oil and grease	mg/L	80	460	296	140	320	40	1600	80	6.14	1600	Not permitted
9	TS	mg/L	30400	800	1100	1400	900	700	1500	600	600	30400	
10	TDS	mg/L	19200	400	846	745	547	875	933	426	400	19200	
11	COD	mg/L	766	908	1547	1001	968	1165	1244	196	196	2496	<100

3.2.1 COD

The effluent COD varies between 196 and 2496 mg/l during the study period. COD concentration in cold weather is high compared to hot weather. This result is because in cold weather the refinery produces kerosene for demand of the city. According to study of Yan et al., (2011) in China, another petroleum refinery wastewater was found to contain up to 1021 mg/L of COD. High COD content inhibit the growth of plant and animals when the effluent discharged to natural environment.

3.2.2 Oil and grease

Similarly, petroleum refinery wastewater contains a high amount of oil and grease. In anaerobic conditions, oil and grease tend to aggregate, clog drain pipes and sewer lines, and cause unpleasant odors and corrosion of sewer lines (Mitra, et al., 2022). According to Iraqi standards, it is not permitted to dispose wastewater contains oil and grease into natural water bodies.

3.2.3 TSS

During the first three months, there were fluctuations in the TSS values. The higher the TSS value, the higher turbidity was obtained. Influencing TSS value by pH was documented by Syafrudin, et al. (2015) they observed that pH values do not significantly influences TSS concentrations in effluent.

3.3. Treatment Methods

In the literature, there are many techniques used to treat wastewaters, particularly DAF and MBBR as well as hybrid processes were used for oily wastewater treatment by applying many methods and

techniques. Additionally, operational parameter for design operation conditions were also explained in table 3 used by researchers.

Table 3 treatment techniques using DAF and MBBR

No.	WW type	WW treatment technique	Type of media	Operation conditions	HRT	%removal	Reference
1	Food industry	DAF+Hybrid MBBR with ASP	polyethylene	148 MBBR hybrid tank, COD=7800 mg/l %35 filling media	-	COD: 97 Oil&grease:99	Falletti et al., 2014
2	Oily wastewater	DAF	-			COD removal rate is 92.5%	Hami et al., 2007
3	Petroleum wastewater	MBBR	85% of the reactor was filled with Polyurethane elements	Volume 550L, MLSS = 1400–1700 mg/L	4 hr	Phenol- 55 to 90% COD- 62 to63%	Jain et al., 2020
4	Municipal ww	MBBR+ reverse osmosis	cylindrical polyethylene	Four different carriers (Ø25-12, Ø 25-4, Ø 15-15, and Ø 10-7mm) pH:7.5-8.1	12 hr	MBBR C with F15 _ 15 polyethylene as the carrier had the best removal performance on NO3-N (78.0 _ 15.8%),, NO2-N(43.79 _ 9.30%),, NH4-N(55.56 _ 22.28%),	Wang et al., 2020
5	Wastewater	MBBR+ coagulation	Kaldnes K1 and K2	Two reactor with different Filling ratio 60% and 70%. Loading rate 10-120 g COD m ² /d	380, 52, 27 and 18 min.	85-90% COD	Ødegaard H. et al., 2000
6	Urban ww	Floating carrier FC	Polysterene, polyvinyl chloride, polyvom, bioballs and polyurethane foam	Filling ratio 10, 20 and 30%	8 hrs	BOD: 95-96 %, NH ₄ : 78- 86%	Makisha, N., 2021
7	Oily ww	An MBBR, ACR	AnoxK™5 carriers	Filling ratio 50%, 30%, 20%	83-104 d, 30 d	COD; 60% COD; 65%	Morgan S., et al., 2019
8	Synthetic phenol ww	MBBR	Anoxkaldnes K1 carriers	pH: 3-7.5	24-48 hr	COD: 90%	Zhou H. et al., 2020
9	Pre-treated textile ww	MBBR	PVC corrugated cylinder	pH: 7, filling ratio: 62%,	2.4 d	BOD: 68.9% , COD: 68.9%	Sosamony and Soloman, 2018
10	Synthetic ww contains glucose and ammonia	MBBR(anoxic, anaerobic)	Kaldness (k1)	Anoxic %40 and anaerobic %50 filling ratio.	5.2 and 14.8 hr	TN: 98%, NH ₄ -N: 99%, SCOD: 99%	Zafarzadeh, A., 2010
11	Petroleum refinery ww	Sequential A/O MBBR	Ceramic bio carrier	Anaerobic/aerobic reactor	72-18 hr	COD and NH ₃ -N>85% at 72 and 36 h HRT, COD < 120 mg/L, NH ₃ -N < 25 mg/L at 18 hr HRT	Lu M, et al., 2013
12	Refinery oil ww	MBBR with granular activated carbon	Bee-cell media	HRT, filling ratio, AC concentration	22hr	Total hydrocarbon removal 95, 91 and 57 % respectively	Sayyahzadeh A, et al., 2016

3.1 Treatment by DAF

Oil-contaminated WWs are generally treated by a combination of Physico-chemical and biological methods. The use of coagulation prior to sedimentation to enhance primary treatments has been limited. In addition, DAF is being used as an alternative process in drinking and industrial wastewater treatments. In this method, the air saturated is injected into the tank. This results in producing small bubbles which push the solid impurities to reduce their density and cause them to float to the surface where they are removed (Edzwald, 2010).

In particular, DAF technology is effective with low-density solids to illustrate the removal efficiencies. In the DAF process, the air is mixed with water a high pressure, allowing the air to dissolve in the water. In comparison with gravity system, DAF yields higher-quality treated water (Abuhasel et al., 2021). DAF and coagulation processes were used to treat landfill leachate and FeCl_3 was added as per the experiments. Thus, an initial concentration of 2610 mg/L of COD was reduced by 78% (Adlan, et al., 2011). The DAF with Nano bubbles is capable of removing more than 90% of the oil from wastewater (Azevedo et al., 2016). Recent work has mainly concentrated on developing DAF systems for reducing the surface tension of oil by adding a surfactant (Bürger et al., 2020). Two processes are involved in bubble formation; first, a nucleation process begins once the pressured water is released with air through the nozzle. As a second step, the excess air in the saturated water is transferred to the flotation tank as gas (Palaniandy et al., 2017). Oily wastewater has been the focus of most DAF research. Furthermore, DAF was applied to papermaking in white waters, highly protein contaminated wastewater and various types of wastewater by controlling some operational parameters such as saturator pressure, flow rate, retention time, bubble size, ..etc. (Palaniandy et al., 2010).

3.3.2. Treatment by MBBR

The MBBR was very effective in treating industrial pollution wastewater. Biofilms grow on both the small carriers and the media moving in the MBBR, a kind of completely mixed biofilm reactor. A major aspect of MBBR systems is aeration rates and biological carriers used in reactors that provide surfaces for bacterial colonization. In an aeration tank operated as shown in Figure 5, the suspended porous biofilm carriers are continuously mixed, activated biomass will develop into a biofilm on the surface of these carriers, whose density is low compared to water. The MBBR also removes organic matter, ammonium nitrogen and phenols were removed at 75 and 95% percent respectively by the MBBR respectively (Dias, et al., 2012).

The advantage of MBBR is that the proportion of carriers containing biofilm can be controlled. According to Rusten et al., (2006) filling fractions below 70% are recommended so that the carrier suspension can move freely. Zafarzadeh, et al., (2010) according to the study, the partial nitrification/denitrification process in the moving bed biofilm reactors system can treat wastewater with a high load of organic nitrogen compounds and organic carbon. Media with a large specific weight need more energy to remain suspended, while media with a smaller specific volume will have more space for wastewater in the reactor. There are various biomass carriers on the market today, but the most commonly used is the K1 type bio-carrier. Recently, polyvinyl alcohol PVA gel was introduced which has been proved effective in the growth and high enrichment of bacteria (Gani, et al., 2014). Mallick, et al., (2017) studied synthetic refinery wastewater treatment by applying aerobic MBBR reactor for optimum HRT of 16 hours, a complete removal of phenol, COD ammonia-nitrogen with nitrification was observed in the performance of reactor. Rahmat, et al., (2016) concluded that salinity changes affect system performance more than phenol changes, as indicated by MBBR reactor removal efficiency for salinities 2% and 3%, were 86-89% and 82-85.5% respectively.

The MBBR operates continuously, so it does not need to be backwashed or maintained. MBBR systems have much higher specific surface areas for biofilm than fixed film systems (Bertino, et al., 2011). Moreover, it was found that MBBR is a better alternative to conventional biological treatment to remove organic matter and ammonia. Also a high efficiency with reduced HRT values can also be achieved by upgrading existing facilities (Schneider, et al., 2011). According to a study performed by Zhang, et al., (2013) on MBBR with polyethylene carriers, COD removal efficiency averaged 75.7, 91.1, 85.5, and 79.6% at 10, 20, 30 and 40% filling rates, respectively. According to these results, the MBBR system

also had a higher COD removal efficiency at a 20% Polyethylene carrier filling rate under the same conditions as influent organic loading rates. Study of MBBR followed by sand filter with the hydraulic retention time of 9 hours was achieved by Dias, et al., (2012). Results showed that the MBBR was able to remove 90% chemical oxygen demand (COD), 75% NH₄, and 95% phenols.

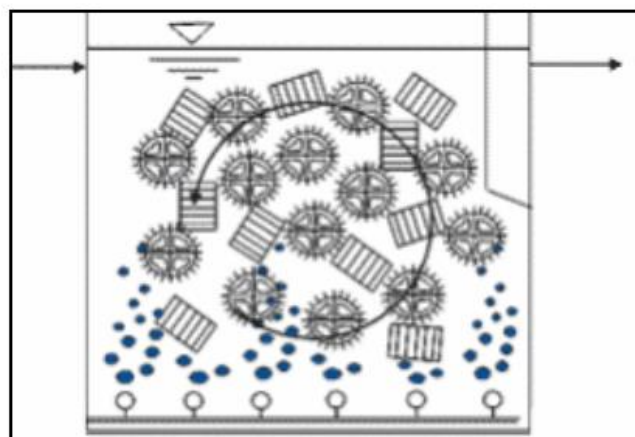


Figure 5 MBBR reactor process (source Lariyah, et al., 2016)

3.4 Reusing

In order to recycle and reuse the WW, it must first be treated and processed to remove impurities, then applied to a wide range of industrial, agricultural, and residential purposes. Water consumption from refineries goes to steam boilers, cooling towers, processes and services, potable water, and other uses (Souza, et al., 2011). Recycled effluent from refineries can be used as cooling water in cooling systems for plants. This can be cost-effective in meeting supplementary water needs. Technologies such as DAF, MBBR, ion exchange, micro/ultra-filtration, and many other biological techniques have the potential to reuse/recycle wastewater in refineries (IPIECA, 2010). Nikazar and Jamshidi, (2008) reported an analysis of treated WW and the city water used in cooling towers showing that the reverse osmosis technique is equally suitable for these purposes. Moreover, water consumption for car washing would increase significantly, which will lead to worsening urban water shortages. For several years, regulators and manufacturers have been paying more attention to reclamation as a method of water conservation and quality control (El-Ashtoukhy, et al., 2015). Brown, (2000) noted that the reclamation of car wash wastewater requires the removal of oil and grease and grit before it can be used. To improve the quality of reclaimed water for use in different wash stages, additional treatment processes can be employed. Some of these techniques such as reverse osmosis, Nano filtration, biological treatments and flotation appears to have a greater potential than any of these technologies and have shown advantages (Zaneti, et al., 2011). To reuse treated car wash wastewater for washing cars, it should not contain particles larger than 10 mm recommended by Brown (2000). Thus, washing cars with high-pressure pumps will damage the skin of the cars if there are larger particles in the treated water.

4- Conclusions

Utilizing the integrated system i.e. DAF followed by MBBR turned out to be a successful solution that might encourage a high level of treatment in oily effluents. Almost all the measured parameters exceed the standards and need to be treated properly. However, all samples for organic compounds such as COD exceed the standard discharge limit of less than 100 mg/L. DAF and MBBR as combined methods have the potential to treat the WWs. Particularly for removing oil and grease and COD in oily wastewaters. In order to ensure that wastewater is released safely and conserve the environment for the greater good of all beings, a proper post-treatment procedure should be applied to the wastewater produced.

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