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WATER REUSE FROM CAR WASH WASTEWATER TREATED BY A MEMBRANE BIOREACTOR SYSTEM WITH CHEMICAL ADDITION

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ABSTRACT

In this study, a lab-scale membrane bioreactor (MBR) system with the ferrous sulfate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) addition was conducted to treat oily wastewater from a car wash station. The system was operated for over 120 days, in which 60 days of the stabilized phase and 60 days for the $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ addition. Based on the effluent characteristics of the system, the removal efficiencies of higher than 90% of chemical oxygen demand (COD) and 70% oily and grease (O&G) without the chemical addition. Subsequently, 98% COD and 93% O&G removal efficiencies with the chemical addition were obtained. The recalcitrant components were removed by the polishing effect of the $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ addition. Interestingly, adding $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ into the MBR system could be helpful in minimization of membrane fouling because the relative small particles, which are considered as a cause of the membrane fouling, could become bigger particles having less fouling tendency. A long operating period without the membrane fouling was achieved when $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ was added into the MBR system. The results of this study pointed out that the oily wastewater from the car wash station could be treated effectively by the MBR system combined with the chemical addition. The MBR system has a good potential for scaling up and is very promising alternatives for water reuse from the car wash station.

Keywords: oily wastewater, ferrous sulfate, membrane bioreactor, membrane fouling, water reuse

1. INTRODUCTION

Car wash service stations play a very important role in maintaining vehicles and keeping them in good condition. Washing activities may include, removal of coarse material, debris and dust using high pressure cleaning water and detergent mixture (Fakhrul-Razi et al., 2009; Razavi and Miri, 2015; Yu et al., 2017). The detergent is applied to the surface that is followed by high pressure water cleaning. A car wash at the garage will take about 115 to 170 L, and after the wash of vehicles, some water will also be used to wash floors and washing equipments (Zaneti et al., 2011). Lot of water is wasted daily for the vehicle washing and servicing (Uçar, 2018). Yasin et al., (2012) studied oily wastewater from different petrol pumps and services stations and found pH, BOD, COD and O&G concentration in the average value of 8.3, 520 mg/L, 1330 mg/L and 1070 mg/L, respectively. COD, O&G and pH were ranged between 255 to 445 mg/L, 400 to 800 mg/L and 6.4 to 7.0 respectively for heavy vehicle sources (Lau et al., 2013). Meanwhile, the COD, O&G concentrations and pH for wastewater from light vehicles sources were ranged from 227 to 378 mg/L, 150 to 700 mg/L and 7.1 to 7.6, respectively (Bujang et al., 2012). High amounts of the organic pollutants in the car wash wastewater cause a dissolved oxygen depletion in the receiving stream (Alsahy et al., 2016; Asha et al., 2016). O&G can coat the fish gills and take to death. The toxic hydrocarbons are fatal to the aquatic life and humans, and grease can cause a loss of the hydraulic capacity of the sewers and fouling of wastewater treatment plant. Suspended solids cause turbidity in water, impair photosynthesis, clog the fish gills, and damage their productivity (Jamaly et al., 2015). Besides, sludge from the car wash activity can clog the sewer and municipal sewers if it is not separated from wastewater stream from the car services stations. Therefore, oily wastewater treatment has become an urgent problem, and it must be treated before discharging.

Several methods could be used for the treatment of oily wastewater, such as flotation, coagulation, biological treatment, membrane separation technology, combined technology, and advanced oxidation process. Among treatment methods, membrane bioreactor (MBR) has a high potential for the removal

of petroleum and oil pollution from oil refinery wastewater treatment (Kumar et al., 2014; Ma et al., 2015; Razavi and Miri, 2015). Scholz and Fuchs (2000) evaluated the treatment of oily wastewater by MBR process, reporting that a high removal rate of 99% was achieved for fuel oil and lubricating oil. Rahman and Al-Malack (2006) investigated the use of a cross-flow membrane bioreactor in treating the wastewater discharged from a petroleum refinery. The average removal efficiency of lubricating oil throughout the MBR operation was observed to be 97%. It should be noted that, during treatment in MBR, biodegradation is a natural process by which microbes alter and breakdown oil into other substances (Viero et al., 2008). Moreover, during the membrane filtration operation, membrane fouling is a major disadvantage, restricting the widespread application of MBR, especially in the oily wastewater treatment (Gkotsis et al., 2014). Numerous attempts have been made in order to prevent and control fouling in MBR. The most common methods to achieve this include the application of conventional physical or chemical methods, the optimal operation of the MBR process by permeate flux reduction and aeration increase (Abu Seman et al., 2011; Hu et al., 2013; Khirani et al., 2006). More recent methods focus on the addition of specific chemicals (Capodici et al., 2017). It should be noted that, one of the requirements in the car wash industry is that water recycling must be in accordance with the present and upcoming environmental laws. These positive evaluations imply that membrane processes can be useful in recycling wastewater in the car wash industry (Vinoth Kumar et al., 2016). However, little attention has been paid to the treatment of oily wastewater from the car wash industry (Uçar, 2018).

This study, therefore, is aimed to examine the combination of membrane bioreactor system with the chemical addition to treat oily wastewater treatment from a car wash station for water reuse. In particular, the system performance on removing the COD and O&G, and the membrane fouling were studied.

2. MATERIALS AND METHODS

2.1 Experimental system

A lab-scale MBR system in this study was presented in Figure 1. The working volume of the MBR system was 10.5 L. A hollow fiber membrane module (PVDF), with a membrane pore size of 0.22 μm and effective membrane area of 0.065 m^2 , was submerged in the bioreactor. The MBR system was operated under a constant flux (8.5 $\text{L}/\text{m}^2\cdot\text{h}$) mode with 10 min of suction, followed by 2 min of relaxation. The transmembrane pressure (TMP) was continually monitored. An aeration system was placed below the membranes to provide dissolved oxygen for biological processes and to minimize membrane fouling due to cake layer formation.

In this study, car wash wastewater samples were collected daily from a car wash station at Le Duan street (Hanoi, Vietnam). It contained a significant amount of large debris and sand. Therefore, it was kept in a 20-L plastic can for 2 h to let the sand settle down. Subsequently, the supernatant was filtered by a standard sieve (0.5 mm) to remove coarse debris before adding it to the reactor. The wastewater characteristics vary in a wide range, TSS 320-640 mg/L, COD 540-780 mg/L, and O&G 60-125 mg/L, and pH 7.6-7.8.

The experiment lasted for almost 120 days, including 60 days for the steady-state period and 60 days for the ferrous sulfate ($\text{FeSO}_4\cdot 7\text{H}_2\text{O}$) addition. In order to add a proper chemical dosage, the preliminary tests were carried out by a standard "Jar Test" apparatus (Model SJ-10, Young Hana Tech Co., LTD). The experimental procedure consisted of three phases: a period of 1 min was allowed for the flash mixing of the coagulant, at 120 rpm. It was followed by a further slow mixing at 40 rpm for 30 min and in the final stage the flocs were allowed to settle for 90 min. The supernatant obtained after 90 min of settling was subjected to analysis.

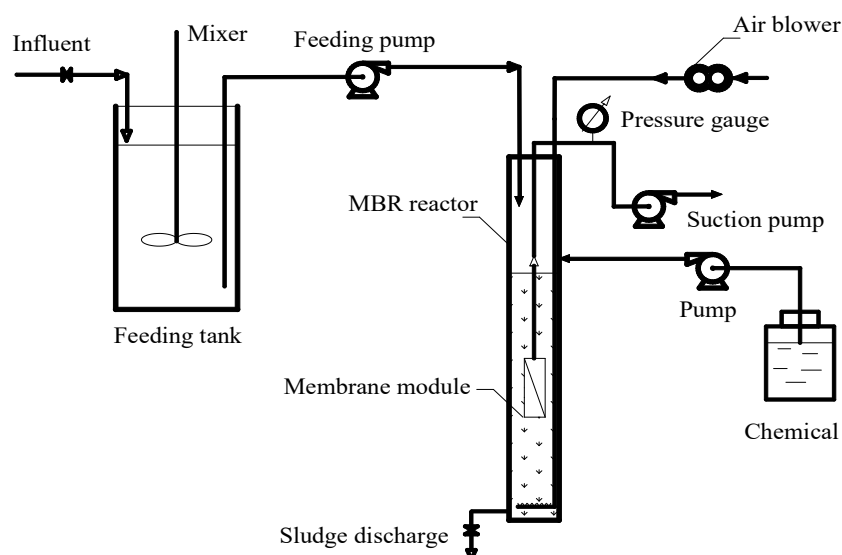


Figure 1. Schematic diagram of the experimental MBR system.

2.2 Analytical methods

The MLSS, MLVSS, and COD were measured based on the Standard Methods (APHA, 2005). MLSS and MLVSS were measured by using the 2540D and 2540E methods, respectively. COD was determined by the closed reflux, colorimetric method (Method 8000). The total oil and grease concentration was determined by using n-hexane as the extraction solvent (USEPA Hexane Extractable Gravimetric Method 1664, adapted from Standard Methods, Section 5520B). The transmembrane pressure (TMP) was monitored using a pressure gauge.

3. RESULTS AND DISCUSSIONS

3.1 Variation of biomass in the system

The seed sludge obtained from a drainage receiving wastewater from a car wash station was used in this study. Figure 2 shows the variation of MLSS and MVSS during operation. In this study, the MLSS was aimed to maintain in the range of 10000 mg/L as reported in MBR systems (Buer and Cumin, 2010). However, in this study, the MLSS was varied largely from about 3000 mg/L to over 6000 mg/L which is much lower than the values from previous studies (Buer and Cumin, 2010, Capodici et al., 2017). The results show that the native microbial biomass cultivated from the sludge obtained from a car wash wastewater drainage could developed to degrade the oily wastewater. The fraction of hydrocarbons probably remained difficult to degrade resulted in limitation of biomass growth. One interesting observation is that in stabilization period, the volatile fractions of the mixed liquor solids (MLVSS/MLSS ratio) in the system was almost identical in the range of 71-76%. However, when $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ was added in the system, the MLVSS/MLSS ratio was decreased down to the range of 63-67%. Thus, a significant reduction in MLVSS/MLSS ratio of about 10% was observed. This could be explained due to the contribution of inorganic solids such as $\text{Fe}(\text{OH})_3$, associated with sludge in the reactors.

3.2 Preliminary test of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ on O&G removal

The O&G concentrations in car was wastewater were varied largely. Therefore, the samples with the O&G concentrations of 100 mg/L were used for the preliminary experiments.

Figure 3 shows that removal of O&G takes place with a fast rate for the ferrous sulfate doses ranging within 60 mg/L. After that the removal rate of O&G were became stable. $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ dose as low as 25 mg/L was required to cause a removal of 50% of O&G. It has been observed that coagulation process can remove O&G with high efficiency and even 100% removal is also possible. However, high chemical dosage will be consumed, resulted in increasing the treatment cost. Besides, a significant volume of sludge could be produced, resulted in increasing the cost for sludge management and disposal (Bujang et al., 2012). Therefore, in this study, 25 mg/L of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ was selected to add directly into the MBR system.

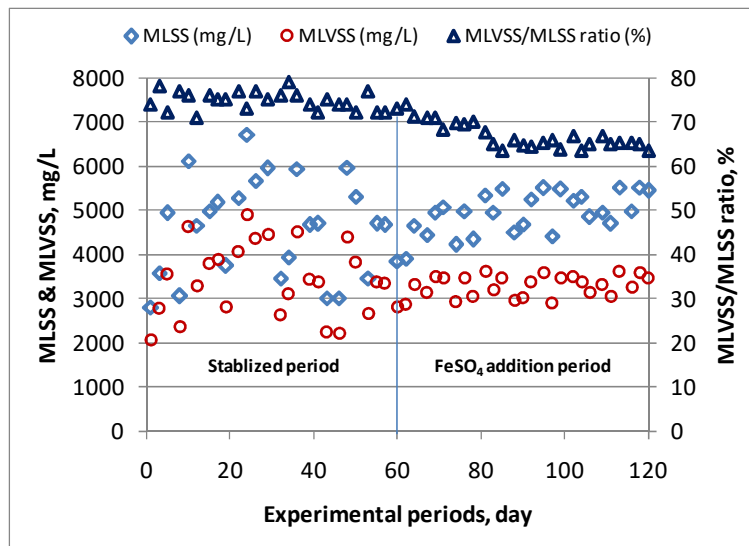


Figure 2. Variation of MLSS, MLVSS and its ratio during experimental operation.

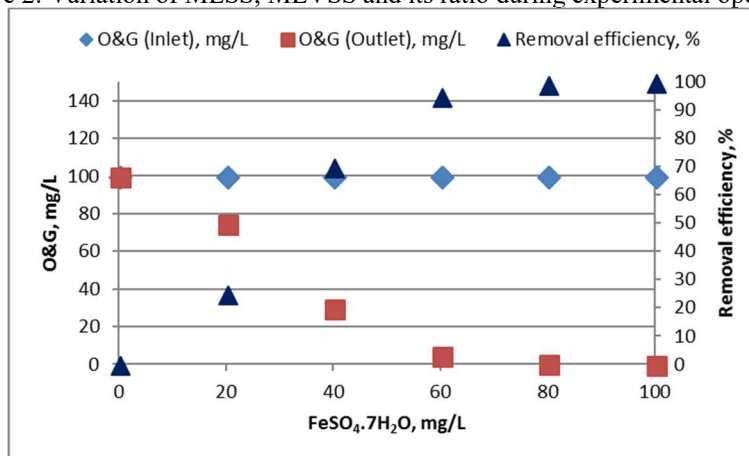


Figure 3. Preliminary tests of ferrous sulfate on O&G removal.

3.3 COD removal in the MBR system

COD removal in the MBR system was presented in Figure 4. It is clear that the influent COD concentrations were varied in a wide range. The results obtained indicated that after the first 20 days of operation, the MBR system could achieve the COD removal efficiency of 90%. In submerged MBRs, the membrane filtration plays an important role in maintaining a high and stable COD removal. It serves as an additional purification phase to retain the remaining particulate COD and biomass, thus improving the quality of the treated wastewater (Kumar et al., 2014). It should be noted that the oily wastewater was not easily biodegradable. Therefore, the biomass could not be fully acclimated to this wastewater (Zaneti et al., 2011). As seen from during stabilized period, the typical COD values of the permeate were about 100 mg/L, showing that the presence of the recalcitrant components were not properly disposed with biological treatment. It was interesting that the effluent COD concentrations were almost lower than 10 mg/L when FeSO₄.7H₂O was added into the MBR system. COD removal was improved from 90% to over 98%. This could be due to that the recalcitrant components were removed by the polishing effect of the chemical addition.

3.4 O&G removal in the MBR system

The oil and grease (O&G) concentration of the influent was approximately 60 to 125 mg/L. As seen from Figure 5, the O&G concentration measured in the effluent showed that it was slowly removed during first 30 days, following which its concentration declined significantly to lower than 35 mg/L.

The average O&G removal rate of 70% was achieved after 60 days of operation. The O&G removal was significant improved upto 93% when FeSO₄.7H₂O was added directly into the MBR system. The effluent &G concentrations were almost lower than 9 mg/L in this operational period. It should be noted that the car wash wastewater containing oil, grease, and chemical surfactants, is susceptible to microbial degradation processes (Capodici et al., 2017). Moreover, a large quantity of oil was present in both,

emulsified and non-emulsified forms (Viero et al., 2008). Therefore, biological treatment of the car wash service station wastewater is feasible with a long retention time (Hu et al., 2013). It should be mentioned that the detergents surround oil droplets with a layer of the detergent molecule to create a water-soluble coating (Vinoth Kumar et al., 2016). During treatment, the oil uptake by a microbial biomass in a bioreactor can be carried out by two mechanisms: uptake by direct interfacial contact of microorganisms with oil drops and uptake by emulsified forms of oil (Jamaly et al., 2015). Scholz and Fuchs (2000) studied the MBR using an ultrafiltration membrane unit, with an oil removal rate higher than 99%. A cross-flow microfiltration process was applied to treat the oily wastewater. An oil removal efficiency of higher than 92% was achieved (Razavi and Miri, 2015).

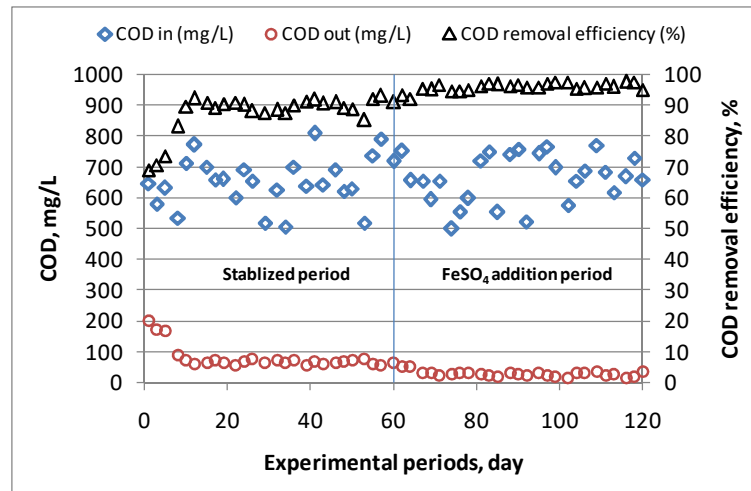


Figure 4. Variation of the influent, effluent COD and its removal efficiency.

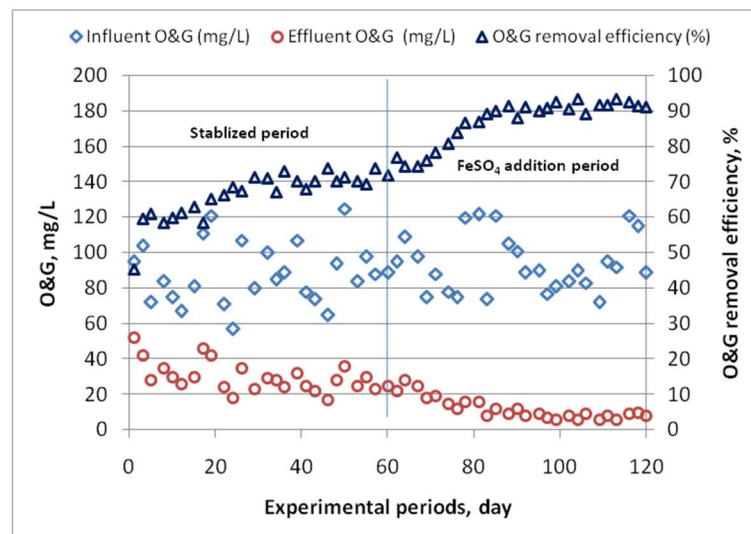


Figure 5. Variation of the influent, effluent O&G and its removal efficiency.

3.5 Membrane fouling during operation

It is known that fouling problem is usually encountered in MBR operation (Buer and Cumin, 2010). One of the methods to determine the threshold activity of MBR is to monitor its transmembrane pressure (TMP) during operation. As shown in Figure 6, in stabilized period the TMP dramatically increased from 1 to over 40 cm Hg within 60 days. The oil-in-water emulsion with a small droplet size could be adsorbed into the sludge. The high hydrophobicity of the sludge was caused due to the strong connection of the highly dispersed oil droplets to the sludge. It should be noted that the membranes used in MBRs are typically made hydrophilic to improve their water permeability. Low hydrophobicity of sludge flocs may cause high fouling due to stronger interactions with the membrane surface (Gkotsis et al., 2014). The fouled membrane module was subjected to physical cleaning (rinsing with tap water) and chemical cleaning (by immersing the membrane module for 2 h in sodium hypochloride solution, followed by 2 h in citric acid solution characterized by a pH of 2.5). It was interesting that during the period of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ addition, the TMP was not significant increased. It seems that the small particles which can be the cause of membrane fouling were getting bigger due to the precipitant addition. It has

been reported that the specific cake resistance and membrane fouling increased due to colloidal particles (Rahman and Al-Malack, 2006). Therefore, the $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ addition could be helpful in minimization of membrane fouling. Thereby, a long operating period without the membrane fouling was achieved.

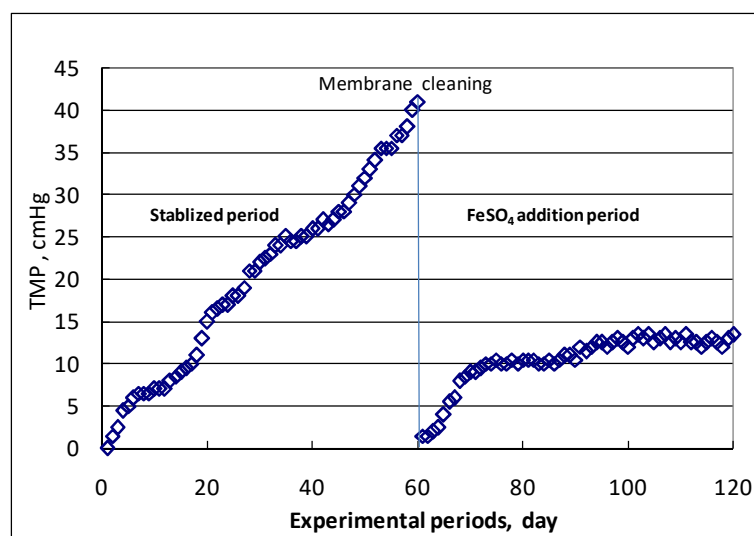


Figure 6. Variation of TMP during the system operation.

4. CONCLUSIONS

From the results obtained it can be concluded that the MBR system was used effectively for treating car wash wastewater. Membrane separation played an important role in ensuring excellent and stable effluent quality. The removal efficiencies of higher than 90% COD and 70% O&G without the chemical addition. The COD and O&G removal efficiencies were enhanced up to 98% and 93%, respectively with the chemical addition. Although membrane fouling was encountered during operation, the $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ addition could minimize the membrane fouling. Therefore, the MBR system showed the good performances for the car wash wastewater. Specially, adding $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ directly into MBR system could maintain the effluent O&G at low concentration. In conclusion, the combination of MBR system and $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ addition offers a potential alternative treatment process for the oily wastewater from the car wash service station. The MBR system combined with chemical addition could supply an effluent of satisfactory quality for water reuse in the car wash industry.

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