Coupling Granular Activated Carbon Adsorption with Membrane Filtration for Chemical Oxygen Demand Removal from High Strength Wastewater

*Aida Isma M.I., Ali Abdo Saad, Rachid Ali A., Munira M., Kenneth Yeoh

Centre for Water Research, Faculty of Engineering and the Built Environment,

SEGi University

* aidaisma@segi.edu.my

Abstract: Combined granular activated carbon adsorption with membrane filtration for high strength wastewater treatment have been carried out. Raw oleo-chemical wastewater and leachate were used as sample. Ultrafiltration is also relatively low cost, easy to backwash and operates up to 3 barg. Experiment was carried out by passing through the sample to an adsorption column for 10 minutes followed by membrane filtration at different trans-membrane pressure of 1, 2 and 3 barg. Oleo-chemical samples were analysed for chemical oxygen demand, turbidity, suspended solid and leachate samples were analysed for chemical oxygen demand and ammonia nitrogen according to APHA method. Results showed that the best chemical oxygen demand, suspended solids and turbidity removal for oleo-chemical samples achieved at 2 bar with 64%, 93% and 97%, respectively. Leachate showed the best removal of chemical oxygen demand and ammonia nitrogen achieved at 3 bar, with 76% and 87%, respectively. The adsorption process combined with membrane filtration is feasible as an alternative for conventional biological treatment for high strength wastewater. However, GAC exhaustive breakthrough point requires monitoring.

Keywords: hollow fibre membrane, granular activated carbon, high strength wastewater

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1. Introduction

Oleo-chemical is a chemical compounds derived industrially from animal or vegetable oils or fats. Oleo-chemicals industry is one of the major industries in Malaysia that is producing glycerine, fatty acid and ester from oleo-chemical. The wastewater discharge produces high chemical oxygen demand (COD) with value between 6,000 mg/L to 20,000 mg/L (Abbasi M. *et al.*, 2013). Conventional biological treatment that is commonly used for treatment of oleo-chemical wastewater is not sufficient to treat high COD concentration which will caused environmental pollution. Leachate is a by-product of sanitary landfills containing complex waste composition with high organic content, high ammonia nitrogen concentration and strong biodegradability (Renou *et al.,* 2008). Due to its large concentration of pollutants, it must be properly treated before being discharged. Directly discharging leachate into the surrounding environment would cause irreversible harm, especially to groundwater systems. High strength wastewater from such as oleo-chemical wastewater and leachate is often contaminated with various compounds such as phenol, chromium, suspended solids and dissolved organic compounds, which leads to high chemical oxygen demand in wastewater and it is crucial that it should be treated to an environmental acceptable limit (Ismail *et al.,* 2016). Effective treatment is required before the effluent can be discharged to the environment. Without proper treatment, wastewater effluent produced eventually contributes serious and long lasting consequences to human and life.

The activated carbon which is known commercially as activated charcoal is made of carbon which is processed to have very small pore sizes. Furthermore, this small sizes of pores results in creating very high surface area which is used for higher adsorption of the chemical particles in the effluent, which is important for the high strength wastewater treatment particularly from industrial wastewater treatment process. Much of the available literature focused on the removal of trace organic contaminants by GAC from surface water (Kim et al., 2010; Deegan et al., 2011; Grover et al.,2011; Hernández-

Leal et al., 2011) and only a few have investigated the use of GAC adsorption for the removal organic contaminants from industrial wastewater (Xing W. et al, 2008). Nguyen et al., 2012 reported that membrane treatment or granular activated carbon on its own will not adequately remove all trace organics of concern in the high industrial wastewater. This study aimed to investigate the reduction of chemical oxygen demand from high strength wastewater using combined GAC adsorption-membrane ration treatment.

2. Materials and Method

2.1 Experimental Setup

The physical properties of the hollow fiber membrane are listed in **Table 1.** The granular activated carbon used were purchased from the local shop (brand: CALGAN). Experiments were carried out in room temperature by allowing the wastewater to pass through granular activated carbon filter for 10 minutes. Then the solution was introduced to the shell side of the hollow fibers membrane filter at different trans-membrane pressure of 1, 2, 3 bar in 10 minutes. Treated effluent were collected in the reservoir for further analysis. Experiments were repeated after membrane cleaning has been carried out. Membrane was backwashed and rinse with distilled water.

Table 1. Details specification of hollow fiber membrane

Materials	Polyvinylidene fluoride
Thickness, (μm)	220
Inner diameter, (μm)	610
Average pore size, (μm)	0.2
Porosity, (%)	50-60

The wastewater samples used in this investigation was collected from the collection sump of the respective wastewater treatment plant. Samples were stored at 4°C to prevent biodegradation of pollutants. A cylindrical hollow fiber membrane module was prepared for the study as shown in **Figure 1.**

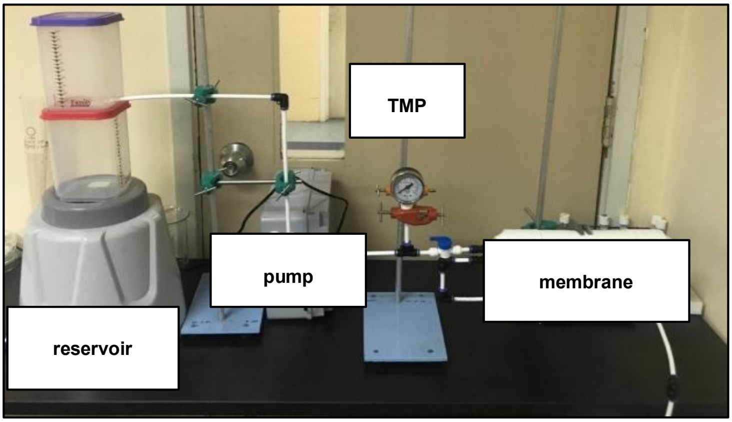

Figure 1. Experimental setup for the membrane filtration system

2.2 Analytical Procedures

Conventional wastewater quality parameters, namely pH, temperature, turbidity, chemical oxygen demand, ammonia nitrogen, and suspended solids were analyzed according to the Standard Methods for the Examination of Water and Wastewater (APHA, 2017). Calorific method with HACH DR/2800 spectrophotometer (set at 620 nm wavelength) was used in measuring COD concentration. Ammonia concentrations was measured by Nesslerization Method (4500 NH_3) using HACH DR2800 spectrophotometer (set at 425 nm wavelength).

3. Results and Discussions

3.1 Removal From Oleo-Chemical Sample

The pH, COD, turbidity and TSS of raw oleo-chemical wastewater collected at the consumption sump tank of the industry were 3.86, 2496 mg/L, 29.26 PtCo and 90 mg/L, respectively as showed in **Table 2.** After adsorption by GAC at pH 3.86, results observed a reduction in COD, TSS and turbidity of 18%, 42% and 43%, respectively. Adjusting pH to 7.58 resulting to better absorption for COD, turbidity and TSS of 23%, 53% and 56%, respectively.

Table 2. Raw and treated by GAC for oleo-chemical sample

No	pH	COD (mg/L)	Turbidity (PtCo)	TSS (mg/L)
I	3.86	2496	29.26	90
	After passing through GAC in 10 min			
II	3.86	2027 (18%)	16.58 (43%)	52 (42%)
III	7.58	1910 (23%)	13.86 (53%)	40 (56%)

Experimental results from the combined GAC-membrane at different trans-membrane pressure (TMP) was tabulated in **Table 3.** Adsorption on GAC followed by filtration from membrane have lead to high initial removal of organic contaminants. It should be noted that the best COD removal achieved at TMP pressure of 2 bar, where most of the pollutants passed through the membrane pore resulting to reduction of COD, TSS and turbidity of 64%, 99% and 96.3%, respectively.

Table 3. Effluent quality at different trans-membrane pressure

TMP (bar)	COD removal (%)	Turbidity removal (%)	TSS removal (%)
1	52.07	84.60	88.15
2	64.02	99.35	96.30
3	62.13	97.07	93.50

It can be seen in **Figure 2** that the color of the treated effluent is clear.

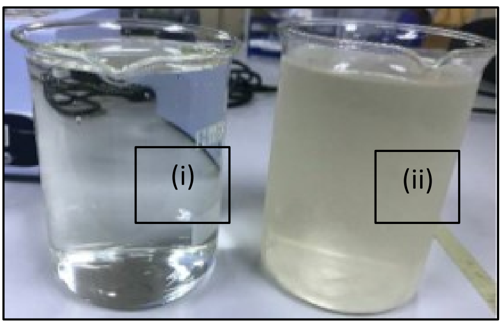

Figure 2. Oleochemical wastewater (i) treated, (ii) untreated

3.2 Removal From Leachate Sample

The removal efficiency of COD and ammonia nitrogen from leachate was investigated using combined GAC-membrane treatment. In this study, three set of experiments on raw leachate at different absorption time and TMP of 5 min, 10 min and 1 bar, 2 bar, 3 bar, respectively as shown in **Figure 3** and **Figure 4** were conducted. Each samples were collected and analysed after the absorption and filtration treatment.

Leachate sample collected from Kuala Selangor was characterized as showed in **Table 4.** Results observed a reduction in COD and ammonia nitrogen in 5 min absorption time to be 15% and 40%, respectively. Increasing absorption time to 10 min resulting to better removal of COD and ammonia nitrogen of 25% and 52%, respectively.

Figure 3. Raw leachate used in the study

Table 4. Raw leachate characteristics

COD (mg/L)		Ammonia-Nitrogen (mg/L)	pH
2853		1570	7.71
	After passing through GAC in 5 min		
COD (mg/L)		Ammonia-Nitrogen (mg/L)	pH
2418(15%)		950(40%)	8.30
	After passing through GAC in 10 min		
COD (mg/L)		Ammonia-Nitrogen (mg/L)	pH
2150(25%)		757.5(52%)	8.41

Experimental results from the combined GAC-membrane at different trans-membrane pressure (TMP) was tabulated in **Table 5.** High removal rates were observed at high TMP. It should be noted that the best removal achieved were at TMP pressure of 3 bar, with COD and ammonia nitrogen reduction of 76% and 87%, respectively. Membrane filtration driven by pressure lead to separation through the semi permeable membrane. Organic.

Table 5. Effluent quality at of combined GAC-membrane at different trans-membrane pressure (TMP)

TMP (bar)	COD removal (%)	Ammonia nitrogen removal (%)
1	43	65
2	47	73
3	76	87

GAC adsorption may lead to high organic matter removal over time, however the adsorption capacity will eventually become shattered due to exhaustive breakthrough that reduces the capabilities of organic matters being absorbed and regeneration of the spent carbon will be needed. This will result in higher organic matter to be loaded onto membrane treatment. It is important to monitor the performance in order to detect the breakthrough point of the biodegradable compounds prior to the membrane treatment. Regeneration of GAC or fresh GAC is needed to ensure pre-treatment prior to membrane filtration. Raw landfill leachate in a very dark colored liquid. **Figure 4** shows the color reduction after treatment by combined GAC-membrane at different TMP. By physical observation, sample (i) with GAC-TMP of 3 bar shows lighter color due to the combined treatment.

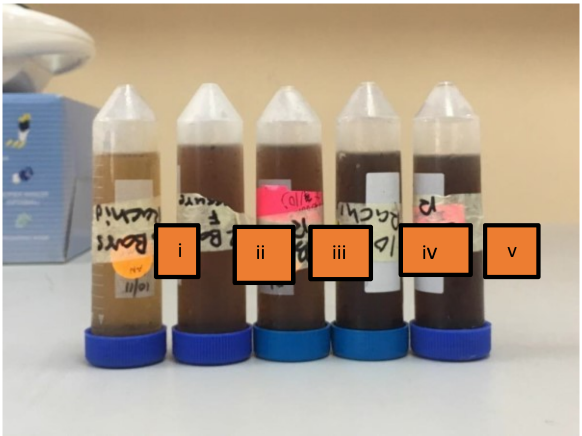

Figure 4. Leachate treatment at various condition: (i) GAC-TMP of 3 bar, (ii) GAC-TMP of 2 bar, (iii) GAC-TMP of 1 bar, (iv) with GAC-10 min., (v) with GAC-5 min

4. Conclusion

From the present study, the following conclusion can be drawn:

1. The best COD removal achieved using combined GAC-membrane for high strength wastewater specifically oleochemical wastewater and leachate were 64% (at 2 barg) and 76% (at 3 barg), respectively.

2. GAC pre-treatment initially complement membrane filtration treatment very well, however results reaffirm that strict monitoring should be in place over the lifetime of the GAC column to detect the breakthrough point of biodegradable compounds.

Acknowledgement

The authors would like to thank Centre for Water Research of SEGi University for the financial and research support.

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