Malaysian Journal of Chemical Engineering & Technology 1 (2018) 28–32

Malaysian Journal of Chemical Engineering & Technology

Journal Homepage: www.mjcetfkk.uitm.edu.my

Removal of ferum ion from wastewater by using thin film composite membrane

Norin Zamiah Kassim Shaari*, Nurul 'Izzati Mohamad, Nurul Aida Sulaiman

Faculty of Chemical Engineering, Universiti Teknologi MARA, Selangor, Malaysia

*Corresponding email: norinzamiah@salam.uitm.edu.my

Abstract

In this study, the thin film composite membrane (TFC) was fabricated to remove ferum ion from wastewater. The composite membrane was fabricated using polysulfone as the support membrane with two formulations of barrier layer; membrane from polymer blend of polyvinyl alcohol (PVA)/Chitosan and membrane from hybrid formulation of the polymer blend with tetraethyl orthosilicate (TEOS). Prior to the membrane filtration process, the wastewater was characterised in term of pH, turbidity, chemical oxygen demand (COD), biological oxygen demand (BOD), total suspended solid (TSS), and concentration of ferum ion. By comparing with Standard B effluent quality for discharge, the wastewater qualities were not conformed to the standard in term of COD, pH and ferum concentration. After pH adjustment was conducted on the wastewater, it was filtered through the two types of the thin film composite, respectively. The effect of pH and formulation of the barrier layers on the percentage removal of ferum ion was investigated. Based on the result, higher removal of ferum ion (99.89%) could be achieved by using TFC with hybrid membrane especially at pH 10 as compared to TFC with polymer blend.

Article Info

Article history:

Received date: 10 September 2017
Accepted date: 24 January 2018

Keywords:

Wastewater
Hybrid membrane
Ferum ion
pH
Chitosan

1.0 Introduction

Globalisation and industrialisation become one of the factors that cause the environmental issues rises rapidly. All the environmental issues caused by chemical industries such as air pollution, water pollution, greenhouse effect, and soil sediment. Thus, variety methods are needed to treat all the problems that causes by the electroplating industries. The methods are important to reduce the emissions of uncontrolled chemicals produced from industries and cause the environment to be polluted. One of the major concerns of the environmental issues caused by chemical industries is the presence of heavy metal in wastewater and in industrial effluent. Over the last few years, the increase in the level of contamination of heavy metals in water had become a major concern.

Heavy metals are the elements that have relatively high density and can become toxic at low concentrations. They also give bad effect to environment and human if produced uncontrollably. Thus, to reduce the emission of heavy metals in wastewater, there are various methods can be used effectively. Reducing heavy metal contain in wastewater is not only to minimise the pollution that may occur, but also to follow the guidelines of Environmental Quality Act 1974, an act that relates to control environment from being polluted (Gunatilake, 2015).

The wastewater produced by industries which normally contains heavy metals, hence it is important that the concentration of heavy metals are within control prior to waste disposal. Examples of heavy metals produced are mercury, iron, copper, zinc, cadmium, lead and aluminium. Some heavy metals are important for human's body to maintain body's metabolism however, if consumed in excess, it can cause serious health effects and may lead to death (Anon, 1971).

As the effluent produced different from one to other industries, different treatments were needed to treat waste produced such as ion exchange, chemical precipitation, membrane filtration, adsorption on new adsorbents, electrodialysis, photocatalysis, and biological methods. By comparing various methods to remove heavy metals from wastewater, membrane filtration possessed a lot of advantages such as requires low energy, high separation efficiency, no phase changing required, simple, heavy metal ions can be recycled and environmental friendly (Huang et al, 2016).

Furthermore, nowadays the most popular method to treat heavy metals from wastewater is membrane filtration as polymer can enhanced the ultrafiltration on membrane to remove the contaminants from wastewater. Polymer may enhance the ultra-filtration as the blended polymer will form the interaction between hydrophobic side chain combination with intramolecular and intermolecular hydrogen bonds and this interaction improved the membrane's filtration. Thus, it also will improve the ultrafiltration process on the membrane (Ariyaskul et al, 2006).

This study focuses on removing heavy metals using thin film composite membrane. Generally, the hybrid membrane may improve the thin film composite membrane's efficiency to remove heavy metals from wastewater. Furthermore, adjusting pH of solutions could increase the percentage of removal of heavy metal using the hybrid membrane.

2.0 Methodology

2.1 Materials characterisation of wastewater

In the preparation to determine the characteristics of wastewater, materials used were comprised of two types of wastewater obtained from Company A; Fe(I): wastewater from equalisation tank and Fe(II): wastewater from settling tank, distilled water, deionized water, digestion solution for COD, ferric chloride, magnesium sulphate, calcium chloride, and phosphate buffer solution. All the reagents for wastewater characterisation were purchased from Merck, Malaysia.

2.2 Materials for preparation of thin film composite membrane

In the preparation of thin film composite membrane, solution from polymer blend of chitosan with polyvinyl alcohol, hybrid membrane solution (polymer blend cross linked with tetraethylorthosilicate, polysulfone resin pellet (MW: 44000–53000), 99% purity of 1-methyl-2-pyrrolidone (NMP), and deionised water were used. All of the chemicals involved in the preparation of hybrid membrane solution and polysulfone were obtained from Sigma Aldrich, Malaysia. 1-methyl-2-pyrrolidone (purity 99%) was obtained from Merck, Malaysia.

2.3 Determination concentration of ferum ion in wastewater

One millilitre of wastewater was poured into 100 ml volumetric flask. Then, distilled water was filled in the volumetric flask until full volume. The solution was shaken until it was completely dissolved. Next, the concentration of ferum (Fe) in the dilution solution was analysed by atomic absorption spectroscopy (AAS). The reading of concentration of Fe was recorded.

2.4 Basic wastewater analysis

pH test: Wastewater of 50 ml was poured into a beaker. Then, the electrode of pH meter was immersed in the wastewater for about 30 seconds until a stable pH reading was obtained. The pH reading was recorded.

Chemical oxygen demand (COD) test: COD reactor was set to be heated to 150 °C. Four vials were labelled according to the source of the wastewater while one vial was labelled as the blank sample. The vials were labelled as Vial A: Fe(I), Vial B: Fe(I) diluted in 250 ml, Vial C: Fe(II) and Vial D : Fe(II) diluted in 100 ml. The digestion solution was filled in all the vials that containing sample whereas the blank sample was filled with 2 ml of deionized water. The vials were mixed well. Next, all the vials were put into the COD reactor for two hours. After two hours, the samples were let to cool in the COD reactor until the temperature reached 120 °C. Then, it was continued with placing the samples on the cooling rack to let the vials cooled down to room temperature. The spectrometer was set with program code 435. The samples were tested through the spectrometer and the COD readings were recorded for all vials. For vial B and D, the reading was multiplied by each dilution factor respectively.

Biochemical oxygen demand (BOD) test: Firstly, the dilution water was prepared. 2000 ml of distilled water was filled in a beaker. Then, 2 ml of each ferric chloride, magnesium sulphate, calcium chloride, and phosphate buffer solution were dropped into the beaker using pipette. Approximately 50 ml of each type of wastewater, Fe (I) and Fe (II) were filled in two BOD bottles respectively. The BOD bottles were labelled as A: Fe (I), B: Fe (I), C: Fe (II), and D: Fe (II). After that, the dilution water was added into each bottle until full volume. Bottle A and C were stored into a thermostatic cabinet for five days. Then, the DO meter was used to measure the initial dissolved oxygen (DO) concentration in bottle B and D. The reading of initial DO for both bottles was recorded. After five days, the reading for final DO was taken using the DO meter with bottle A and C as the sample. The DO reading was recorded, and BOD was calculated.

Total suspended solid (TSS) test: Spectrometer was set with program code 630. About 10 ml of distilled water was filled into the sample cell as the blank cell while 10 ml for each Fe (I) and Fe (II) was filled into each sample cell. The blank cell was put into the spectrometer to get the zero reading. Next, sample cell filled with Fe(I) was put into the spectrometer and the TSS reading was recorded. The steps were repeated with sample cell containing Fe (II).

2.5 Preparation of porous support membrane

Thirteen grams of polysulfone pellet was dissolved into 1-methyl-2-pyrrolidone to prepare 13 wt.% polysulfone. The mixture was stirred continuously at 60 °C for five hours until homogenous. The mixture was then left at room temperature to remove air bubbles. Then the polymer solution was cast onto a glass plate by using casting membrane applicator by adjusting the thickness to 90 µm. The polysulfone membrane was left overnight in the coagulation bath at room temperature (Gunatilake, 2015). It was dried under the sun for a day.

2.6 Preparation of thin film composite membrane

The polysulfone membrane was placed on a glass plate. Next, the hybrid membrane solution was cast on the polysulfone as a thin layer to form a thin film composite membrane. Glass rod was used for the casting process. The composite membrane was left at room temperature for 24 hours. Then, the composite membrane was heat-cured at 45 °C in an oven for one hour (Gunatilake, 2015).

2.7 Performance testing

Prior to the treatment process by using the thin film composite, pH of the wastewater, Fe (I) was adjusted to 7 and 10. Then, the thin film composite membrane was placed in the casing section of the membrane filtration rig. The pressure was applied at 12 bars by using nitrogen gas. The permeate sample was collected at 30 minutes time interval. The permeate flux was calculated. Each sample collected was sent to analysis for ferum ion concentration by atomic absorption spectroscopy (AAS) (Sulaiman et al., 2016). The percentage removal of the metal ion was calculated and recorded.

3.0 Results and discussion

3.1 Characterisation of wastewater

As the company A is an electroplating company, thus the wastewater produced must comply with Standard B of effluent standard by Environmental Quality Act 1974 (EQA). Based on characteristics of two types of wastewater as shown in Table 1, both waste water requires a treatment process as the COD, pH and ferum concentration are exceeded the allowable limit (Effluent Standard, 2017).

Table 1: Characteristics of the wastewater

Parameter	Wastewater	Standard B
pH	1.95	5.5–9.0
COD (mg/L)	666.7	200
BOD (mg/L)	50.94	50
TSS (mg/L)	11.70	100
Fe concentration (mg/L)	281.91	5.0

However, for the treatment process by using the fabricated thin film composite membrane, wastewater Fe(I) had been selected as it contained higher concentration of Fe (II) as compared to Fe (I). Based on the result in Table 1, it can be concluded that wastewater Fe (I) needs the highest oxygen than wastewater Fe (II) to degrade organic matter. Thus, it requires membrane filtration to remove the contaminations it efficiently.

3.2 Effect of pH and formulation of barrier layer on percentage removal of ferum ion from the wastewater

Fig. 2 depicts that at pH 7, the percentage removal of ferum ion from the wastewater increases with time for both types of composite membranes. However, the percentage removal is higher for composite with hybrid membrane layer as compared to that with the polymer blend. It could be observed during the third hour of the treatment process, the removal of Fe ion by using composite with hybrid membrane is 99.22% as compared to 95.77% through the polymer blend.

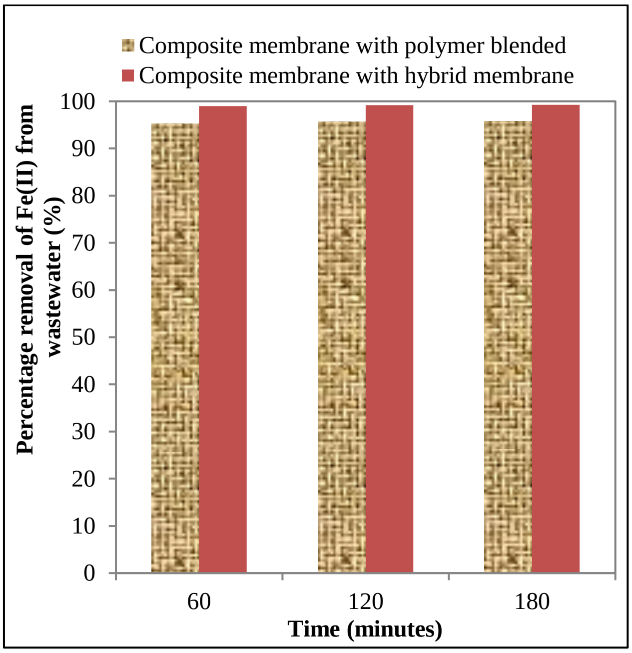

Fig. 2: Percentage removal of Fe ion from wastewater at pH 7 against time.

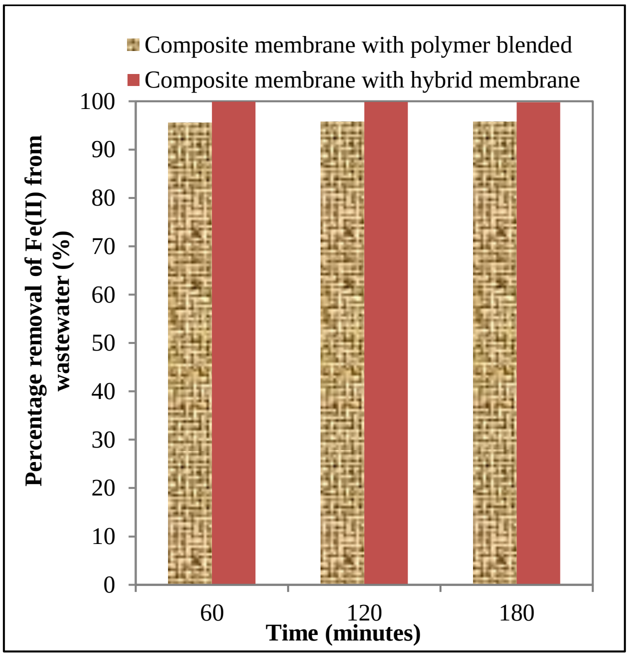

Fig. 3: Percentage removal of Fe ion from wastewater at pH 10 against time.

As shown in Fig. 3, by increasing the pH of feed solution to pH 10, the percentage removal of ferum ion increased as compared to pH 7 for both types of composite membrane. Composite membrane with hybrid membrane has the highest removal at the first hour with 99.89% and the value keep constant until three hours as compared to the percentage removal of ferum ion using composite membrane with polymer blended. A slight increase in the percentage removal was observed by using composite with polymer blend after three hours of treatment process.

Basically, the actual wastewater contains stable ferum ions, which are dissolved in aqueous form and unable to form solids. The insoluble precipitates will form after pH adjustment (pH 7 and 10) was conducted on the feed solution. The precipitation process had caused the concentration of metal in wastewater become lower and easy to be removed.

3.3 Effect of pH and formulation of barrier layer on the permeate flux

Based on Fig. 4 and 5, less membrane fouling occurred during the experiment as the flux increased with the increase in the filtration time. This situation was due to the presence of polymer blend or hybrid membrane layer which had reduced the fouling process unlike polysulfone membrane (Miller et al., 2014a).

Membrane fouling is easy to occur when the membrane was made up using the phase inversion method.

This is due to the phase inversion method where water was used as the non-solvent with hydrophobic polymer as the solvent to fabricate the membrane. As the hydrophobic membrane solutes in the water, the water caused the membranes to foul through strong hydrophobic interactions that occur in the membrane (Miller et al., 2014a). As both membranes contain surface modification, it can help in controlling the membrane fouling from occurs during the experiment. The surface modifications help to control membrane fouling as the polymer membrane was hydrophobic and the modification on membrane’s surface usually directed the surface of membrane to hydrophilic (Miller et al., 2014a).

If the membrane contains hydrophilic surfaces, the membrane will attract a strongly bound layer of water molecules. This layer of water molecules will act as the buffer on the adhesion of hydrophobic foulant. Thus, as the membrane having hydrophilic surface modifications, it will form a thin and highly permeable dense film. This film will help in reducing the overall mass transfer resistance of the membrane (Miller et al., 2014a).

Based on Fig. 4 and 5, as both composite membranes showed an increase value in membrane flux throughout the filtration time, it shows that the membrane mass transfer resistance for both composite membranes was lower where only membrane itself was acted as the resistance as there was no foulant accumulate on the membrane’s surface (Miller et al., 2014b).

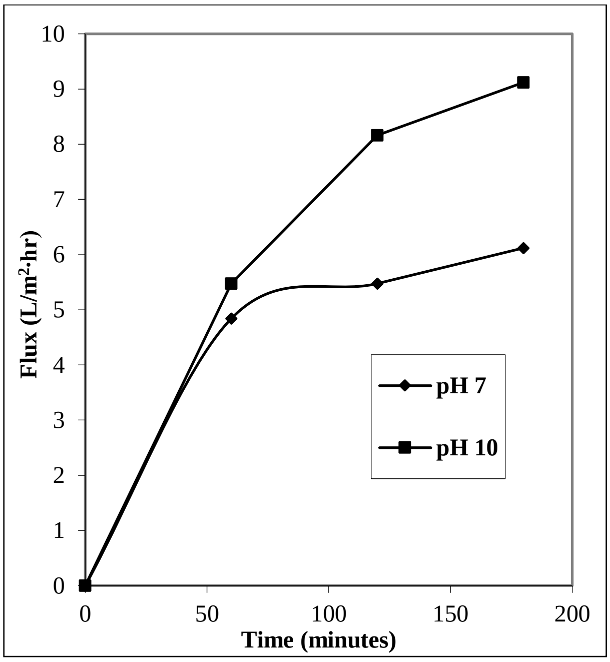

Fig. 4: Flux versus filtration time for composite with polymer blend.

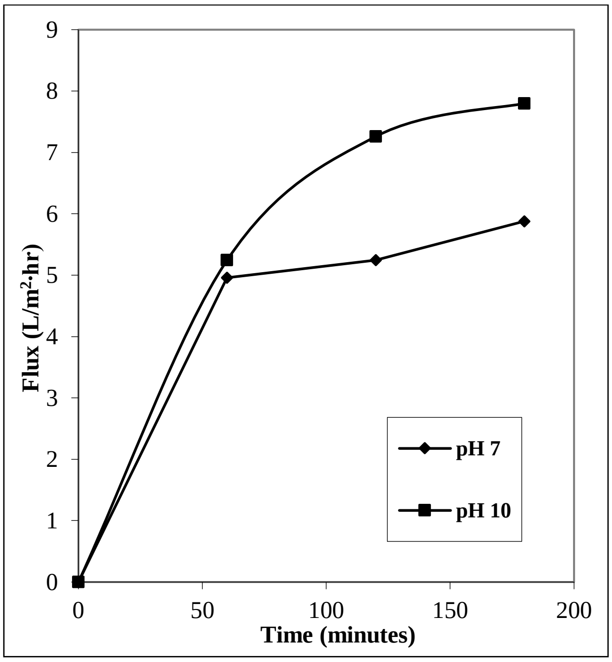

Fig. 5: Flux versus filtration time for composite with hybrid membrane.

4.0 Conclusion

From the experiment, wastewater Fe(I) was chosen as the sample test for performance testing of membrane due to its contamination present in the wastewater in term of BOD and ferum concentration, was higher than wastewater Fe (II). Based on the results obtained, comparable performance of composite with polymer blend was observed at pH 7 and 10. However, a slight improvement on the percentage removal of ferum ion was depicted by using composite with hybrid membrane at pH 10 as compared to pH 7. The duration of treatment process did not affect much on the percentage removal as the values are constant throughout three hours filtration process. Furthermore, less fouling was expected by using the thin film composite membrane (TFC) as the volume of the permeate increases along with the filtration time. Finally, the fabricated TFC, which represent the integrated complexation process, is suitable to treat the wastewater containing ferum ion because the final concentration after the treatment process is within the acceptable limit of Standard B of the effluent discharge of EQA (1974).

Acknowledgment

We would like to thank Faculty of Civil Engineering, UiTM Shah Alam for allowing us to conduct wastewater analysis in the laboratory. Thank you also to Ministry of Higher Education Malaysia for the sponsorship of the research through Fundamental Research Grant Scheme with file no. 600-RMI/FRGS 5/3 (92/2014).

References

A.S. Ariyaskul, R.Y.M. Huang, P.L. Douglas, R. Pal, X. Feng, P. Chen, L. Liu, (2006). Blended chitosan and polyvinyl alcohol membranes for the pervaporization dehydration of isopropanol. *Journal of Membranes Science*. *280.* 815-823.

Anon, (1971). Trace metals: unknown, unseen pollution threat. *Chem. Eng. News, 49 (3).* 29-30.

D.J. Miller, S. Kasemset, S., L. Wang, D.R. Paul, B.D. Freeman, (2014a). Constant flux cross flow filtration evaluation of surface-modified fouling-resistant membrane. *Journal of Membrane Science*. *452*. 171-183.

D.J. Miller, S. Kasemset, S., L. Wang, D.R. Paul, B.D. Freeman, (2014b). Comparison of membrane fouling at constant flux and constant transmembrane pressure condition. *Journal of Membrane Science*. *454.* 505-515.

Effluent Standards. (2017). Retrieved May 30, 2017, from Indah Water: https://www.iwk.com.my/do-you-know/effluent-standards.

J. Huang, F. Yuan, G. Zeng, X. Li, Y. Gu, L. Shi, W. Liu, Y. Shi (2016). Influence of pH on heavy metal speciation and removal from wastewater using micellar-enhanced ultrafiltration. *Chemosphere. 173.* 199-206.

N.A. Sulaiman, N.Z.K. Shaari, N.A. Rahman, (2016). Removal of Cu (II) and Fe II) Ions Through Thin Film Composite (TFC) with Hybrid Membrane, *Journal of Engineering Science and Technology Special Issues SOMCHE 2015*, 36-41.

S.K. Gunatilake (2015). Methods of removing heavy metals from industrial wastewater. *Journal of Multidisciplinary Engineering Science Studies*. 12-16.