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ENHANCED REMOVAL OF MULTI-METAL ION BY GRAPHENE OXIDE POLYETHERSULFONE NANOCOMPOSITE ADSORPTIVE MEMBRANE

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ABSTRACT: Water pollution due to natural and human activities is a critical issue of global concern. Moreover, rapid population growth that led to urbanization, industrialization and intensification of agricultural activities has rendered to the accumulation of pollutants in water and wastewater. Heavy metals were identified as one of the inorganic pollutants found in significant amounts in industrial effluent discharges. Therefore, this study aims to evaluate the efficiency of graphene oxide polyethersulfone nanocomposite (GPN) adsorptive membrane in a synthetic multi-metal ion system using dead-end filtration process. GO powder was incorporated with a polymer solution to fabricate the GPN membrane. After that, simulated industrial wastewater was prepared based on the real waste content including lead (Pb), iron (Fe), copper (Cu) and aluminum (Al) at pH 3.4±0.2. Then, the dead-end filtration was conducted using the prepared simulated wastewater at 3 bar of transmembrane pressure. The final concentration of the metals in the permeate demonstrated that the removal of multi-metal ions was in the descending order of Al > Pb >Cu > Fe and Pb > Al > Cu > Fe for Type 1 and 2, respectively. For Type 1 simulated wastewater (mining industry), the removal percentages were about 62, 40, 61 and 81% for Pb, Fe, Cu and Al, respectively. Meanwhile, for Type 2 simulated wastewater (semiconductor industry), the removal percentages were 93, 43, 63 and 67% for Pb, Fe, Cu and Al, respectively. The results exhibited that metal ions compete for these interactions and behave differently when they come into contact with the GPN membrane. The final concentration of the metals in the permeate demonstrated an enhanced removal of multi-metal ions. Therefore, it is suggested that this GPN membrane is suitable for industrial use and could be verified with real wastewater effluent in the future.

KEYWORDS: *Modified graphene oxide, Adsorptive membrane and Multi-metal ion removal.*

1. INTRODUCTION

Heavy metal contamination in water is a significant environmental issue that poses serious threats to human health and the ecosystem. While these metals have various industrial and commercial applications, their improper disposal and release into the environment can lead to water pollution. Once heavy metals enter water systems, they can persist for extended periods and accumulate in the environment. As a result, they are often found in elevated concentrations in rivers, lakes, and groundwater. These elevated levels can have harmful effects on aquatic life, plants, and humans [1,2]. To date, the incorporation of nanomaterials in membrane fabrication has been proven to improve membrane performance and fouling resistance as well

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as provide innovative systems for novel adsorptive/filtration membranes for improving wastewater treatment [3]. Graphene oxide (GO), a highly oxidized form of the graphene sheet, has been investigated due to easy dispersion in an aqueous solution as these oxygen moieties make GO hydrophilic [4-8]. This study explores the application of an effective membrane utilizing GO as the main nanomaterial of a nanocomposite adsorptive membrane for heavy metals removal known as graphene oxide polyethersulfone nanocomposite (GPN) membrane. In general, industrial effluent contains a variety of pollutants which is more than one of heavy metals. As a result, incorporating other heavy metals would serve as simulated real-world wastewater, allowing the hindrance of other metal ions to be determined utilizing the GPN membrane. However, only a few studies of the GPN have been tested on multi-metal ion rejection and no study has evaluated the simulated wastewater based on the wastewater effluent from mining/semiconductor industries. Therefore, the aim of this study was to investigate and validate the applicability of GPN for enhanced removal of multi metal ions and elucidation of its mechanism. This research envisages that heavy metals containing wastewater effluents that are currently discharged into the water environment could be efficiently treated and subsequently control the water pollution issues by integrating nanomaterials and adsorptive membrane systems. an attempt to make formatting easy, you can use the style menu just under the standard menu.

2. MATERIALS AND METHOD

All reagents used for the experiment were analytical grade. Lead nitrate (PbNO3: 99%), and iron sulfate (FeSO4: 98%) were purchased from R&M Chemicals. Copper sulfate (CuSO4: 98%) was supplied from Ajax Finechem and aluminum sulfate (Al2(SO4)3: 98%) was purchased from HmbG Chemicals. The synthesized graphene oxide according to Hummers' method and the fabricated GPN membrane as described elsewhere [9], were used throughout the experiment utilizing graphite powder (particle size < 50 μm) which was supplied by R&M Chemicals, Canada.

2.1. Simulated wastewater

In this study, the simulated industrial wastewater was prepared based on the real waste content as reported previously [10,11] and tested for the applicability of the GPN. The lead (Pb) ion concentration was mixed with other metals including iron (Fe), copper (Cu) and aluminum (Al) and the pH was 3.4±0.2, where the pH of the solution remains similar to that of the acidic real wastewater. The components of simulated wastewater from industrial wastewater were prepared according to previous findings from mining (Pb-100; Fe-30; Cu-70; Al-200 mg/L) and semiconductor (Pb-1.5; Fe-3.5; Cu-2; Al-3 mg/L) industries pertaining to the multi-metal ions system. Finally, the final concentration of the multi-metal elements was determined using ICPMS and the removal efficiency was observed.

2.2 Dead-end filtration

The dead-end filtration experiments were conducted on GPN membrane using simulated wastewater containing multi-metal ions as inlet feed. The filtration experiment was conducted according to [9] with slight modification using a hand-made dead-end membrane filtration set-up with constant transmembrane pressure (TMP) and a variation of filtration time. The compaction step was carried out for 15 min at a pressure of 3 bars to obtain a steadier solution flux and to remove residual chemicals. The permeate flux was calculated using the Equation(1).

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$$J = \frac{\Delta V}{A \Delta t} \tag{1}$$

where J is the measured permeate flux (L/m^2 h), ΔV is the permeate cumulative volume (L), A is the effective membrane surface area (m^2), and Δt is the filtration duration (h).

3. RESULTS AND DISCUSSION

3.1 Multi-metal ions rejection

The dead-end filtration experiments were conducted on GPN membrane using multi metal ions as inlet feed. The removal percentage of multi metal ions were obtained using ICPMS and analyzed. Type 1 simulated wastewater is based on the mining industry with high concentration of heavy metals content, while the Type 2 of simulated wastewater is based on the semiconductor industry which contains low concentration of heavy metals. The permeation flux of both sets is shown in Fig. 1 below. The fluxes values are 412 L/m^{2}h and 240 L/m^{2}h for low and high concentration of simulated wastewater, respectively. From the results, the flux is decreased with increasing concentration. This is suggested that membrane resistance increases linearly with increasing initial feed concentration [12]. However, it can be clearly seen that both attained steady flux in 30 minutes.

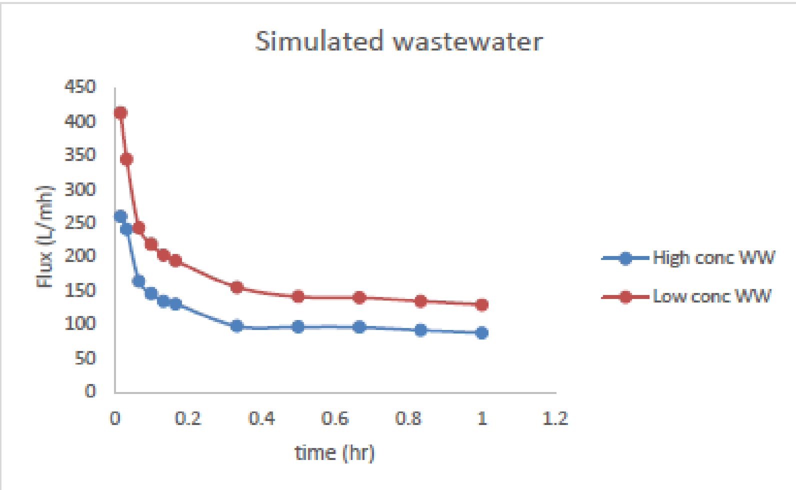

Fig. 1. Flux vs time plot for simulated wastewaters at 3 bar of TMP and acidic conditions (pH 3.2±0.2).

The percentages removal for heavy metals were shown in Figure 2. The multi metals ion rejection was determined using ICPMS analysis. The final concentration of the metals in the permeate demonstrated that the removal of multi-metal ions was in the descending order of Al > Pb >Cu > Fe and Pb > Al > Cu > Fe for Type 1 and 2, respectively. For Type 1 simulated wastewater (mining industry), the removal percentages were about 62, 40, 61 and 81% for Pb, Fe, Cu and Al, respectively. Meanwhile, for Type 2 simulated wastewater (semiconductor industry), the removal percentages were 93, 43, 63 and 67% for Pb, Fe, Cu and Al, correspondingly. From the results, it is revealed the rejection percentage for heavy metals did not follow an orderly trend, but it appeared to follow these manners which removal of Pb and Al was higher than the others for semiconductor and mining industry, respectively.

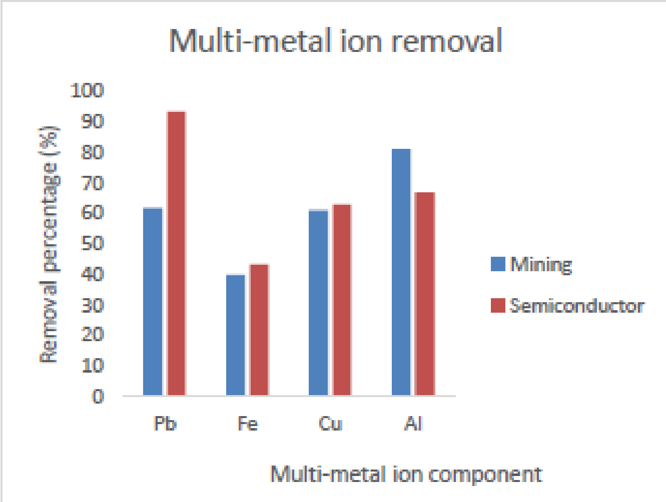

Fig. 2. Removal percentage of multi metal ions component through GPN membrane at 3 bar of TMP and acidic condition (pH 3.2±0.2)

The removal of metal ions from aqueous solutions is attributed to two types of interactions: electrostatic and Van der Waals interactions [5]. Metal ions compete for these interactions and behave differently when they come into contact with the GPN membrane. It is suggested that the metal ions can be coordinated due to the presence of OH and COOH groups in GO and the electrostatic interaction between the functional group in GPN membrane and metal ions are responsible for this selective permeation [5,13]. In addition, metal ions are removed with a three-step mechanism involving the external diffusion of metal ions, diffusion into the nanocomposite and adsorption in the nanocomposite as well as a synergetic effect [5,14].

4. CONCLUSION

The validation of the GPN applicability towards the simulated wastewater containing multi-metal ions is well achieved where the GPN membrane can be utilized to remove multi-metal ions from the wastewater. The final concentration of the metals in the permeate demonstrated an enhanced removal of multi-metal ions. Therefore, it is suggested that this GPN membrane is suitable for industrial use and could be verified with real wastewater effluent in the future.

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