Synthesis and Characterization of Montmorillonite Membrane for Nickel and Zink Metals Filtration in Electroplating Liquid Waste

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Article Info

Article history:

Received: May 2nd, 2020
Revised: May 26th, 2020
Accepted: June 12th, 2020

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ABSTRACT

The application of ceramic membrane technology has been applied in water treatment. The montmorillonite used was analyzed by XRD and showed typical montmorillonite peaks at 2θ 7.49°, 28.07°, 35.75 °, 40.73°, 49.14° dan 67.03°. Montmorillonite membrane was made by the process of dry pressing. PVA in montmorillonite is expected to function as a binder agent. The result variation on PVA composition 0%, 1%. 2%, 3% and 4%, show the flux value that increased with the adition of PVA composition. The montmorillonite membrane with a 4% PVA composition a result greatest flux and porosity values compared to other compositions. The resulting flux value is 1596.5679 L/m^2.h and porosity 20.9477%. SEM-EDX test result showed montmorillonite membrane is a porous membrane that is spread and evenly distributed but the resulting pore is not uniform. Montmorillonite membrane rejection of nickel metal was 55.5352% dan zink metal was 37.8614% with a 40-minute filtration process.

Keyword: : *montmorilonitte, membrane, electroplating*

1. INTRODUCTION

In line with the development of industry in Indonesia, there are many challenges that must be faced, one of which is about environmental pollution in the form of waste production. Industrial liquid waste can cause environmental pollution, especially in if discharged into water without the proper treatment. Industrial wastewater contains many polluting particles, such as metal ions and organic materials that have the potential to reduce water quality (Nurhasni, 2013). One of the industries that produce a lot of waste is the electroplating industry. The electroplatting industry is an industry that performs metal coating with the help of an electric current. The electroplating industry mainly produces liquid waste containing metal ions such as metal Cu^{2+}, Zn^{2+}, Cr^{2+}, Cd^{2+}, Ni^{2+}, and Pb^{2+} (Nurhasni, 2013). Waste containing metal ions can pollute the environment if discharged directly into water bodies without proper treatment.

Zn metal is actually not toxic, but in its ionic state, Zn has high toxicity (Zulfiah, 2017). The same is true for nickel metal, where the Nickel content is also not good for human health. Metal ions or its compounds that enter the body can interact with various physiological elements in the body so that they interfere with the body's metabolism (Said, 2010). Various attempts have been made to deal with industrial liquid waste such as adsorption, coagulation, filtration, and others. The filtration method is a method that is widely used in the separation method, especially in water treatment (Widyastuti, 2011). Filtration technique is a separation technique based on the fluid that is passed through a certain filter.

Filtration using membranes is one of the most widely used filtration methods. Membrane separation techniques can reduce energy use and obtain better results than other separation methods (Widyastuti, 2011). Membrane filtration technology continues to develop as an alternative treatment of water. Based on the particle size that can be rejected in the pressure difference-based membrane operation, the membrane can be divided into microfiltration, ultrafiltration, nanofiltration, andmembranes reverse osmosis (Li, 2006). Membrane technology works in 2 types of filtration, namely cross-flow filtration and dead-end filtration (Japaiko, 2015). The filtration process with the method dead-end filtration has advantages over the cross-flow filtration method, namely the dead-end filtration method can be used to separate contaminants that want to be separated with low concentrations (Japaiko, 2015).

Membrane materials are usually made from synthetic organic polymer materials (Wenten, et al., 2010). Apart from being made from organic materials, membranes can also be made from inorganic materials such as ceramics and metals. The use of ceramic membranes has several advantages over other separation methods, including it does not require changing the phase of the medium either physically, chemically or biologically. The process is fast, simple operation, thermal stability and pressure resistant. Besides that, it also has a long life, and has resistance to fouling and is easy to clean the membrane (Harabia, *et al*., 2012). One of the inorganic materials that can be used as a membrane material is clay. There are several types of clay including bentonite, montmorillonite, illete and others. The name montmorillonite comes from a type of plastic clay found in Montmorillonite, France in 1874 (Barleany, 2011). Montmorillonite has a high melting point, which is around 1330-1430^{o}C. In the high melting point, montmorillonite is very suitable as a material for the manufacture of ceramic membrane products. In addition, montmorillonite also has high expansion and shrinkage power when exposed to water, so this material is suitable for use as a membrane because it can absorb and fix metal ions and organic compounds.

The efficiency of a membrane process is determined by two parameters, namely flow rate and selectivity in passing through the membrane. Flow rate is usually expressed as flux. Flux is defined as the volume flowing through the membrane union of membrane area and time. The units of flux are expressed in $Lm^{-2}hours^{-1}$. The selectivity of a membrane to a mixture is usually expressed as a separation factor or retention factor (R). For solutions with low concentrations or dilute solutions containing a solvent (usually water) and a solute, the selectivity factor is usually expressed as retention (R) to the solute. Most or all of the solutes can be retained or retained by the membrane so that only water (the solvent) and a small portion of the solute can pass through the membrane (Kristianingrum, 2018). There are several methods for making membranes, namely dry pressing, plastic forming, casting techniques, hot pressing and hot isostatic pressing (Kopeliovich, 2013).

Dry pressing is the most common method of making membranes for forming components in ceramic membranes. Dry pessing is a relatively simple technology in the manufacture of ceramic membranes (Demirkol, 2019). The method is dry pressing very well used in the manufacture of ceramic membranes where the basic component of the membrane is dry powder. Dry pressing is the method most often used in the manufacture of ceramic membranes (Kocak and Karasu, 2018). Dry pressing can be defined as the simultaneous unaxial compaction and formation of a granular powder with a small amount of water and / or an organic binder during the compression process. The process is dry pressing carried out on a ceramic membrane body with a very low humidity or water content of <5%. The dry pressing method requires sufficient pressure to form a greenbody ceramic membrane. Dry pressing is one method that is widely used because of its relatively simple technology (Reed, 1976).

Based on the description above, it is necessary to have a study that can explain the characteristics of the membrane and the ability of the montmorillonite membrane which is made based on the dry pressing method in reducing the content of nickel and zink metals in electroplating industrial liquid waste Kotagede area of Yogyakarta. In this study, the filtration method was used dead-end filtration as a feed stream of industrial liquid waste.

2. RESEARCH METHOD

2.1. Membrane Preparation

About 0.2 grams of PVA were dissolved into water and 19.8 grams of montmorillonite was added. The mixture is heated until smooth., then dried and mashed with a 100-esh sieve. Afterwards, 3 grams of the mixture was taken and put into a ring-mold with a diameter of 4 cm and then pressed. The printed membrane was sintered at 900^{o}C for 3 hours.

2.2. Electroplatting Liquid Waste Filtration using Montmorillonite Membrane

Electroplating waste was analyzed by AAS to determine contaminant metal, which were nickel and zink. The waste that has been analyzed, was put into the dead-end filtration reactor. The filtrate was extracted every 10 minutes for 40 minutes of the filtration process. The time and volume of permeate passing through the membrane are recorded. Permeate was analyzed by AAS to determine the content of nickel metal ions and zink after the filtration process. The montmorillonite membrane was used in the dried conditions and analyzed by SEM-EDX.

Figure 1. Dead-end filtration reactor

3. RESULTS AND DISCUSSION

3.1. Initial Characterization of Montmorillonite

The main material in this study was montmorillonite. To confirm the main material used, montmorillonite was analyzed by XRD (X-Ray Duffraction). XRD is used in this analysis because the patterns resulting from the diffraction of each material will be different from one another, so that XRD can be used to identify and provide information about a crystal composed of molecular units. The diffractogram generated from the X-Ray Diffraction that can be seen in Figure 2.

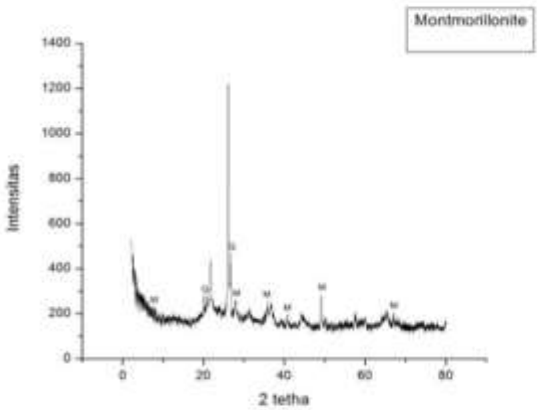

Figure 2. XRD diffractograms of montmorillonite

Based on the XRD difraktorgram analysis results, the typical peaks of montmorillonite contained in 2θ angle 7.49° 28.07°; 35.75°; 40.73°; 49.14° and 67.03°. The peaks resulting from the diffractogram analysis experienced a slight shift. In addition, the diffractogram montmorillonite also shows the peaks from other compounds, namely quartz which appears at an angle of 2θ 2.87° and 26.67°. This indicates that the montmorillonite used is impure or there are impurities

3.2. The effect of PVA concentration on the formation of the montmorillonite membrane

The montmorillonite membrane can be categorized as an inorganic membrane because it is synthesized from inorganic materials (Arief, 2013). This membrane was synthesized using themethod dry pressing which was then sintered at high temperatures. The sitering process is a process carried out by compacting powdered material by pressing (compaction) then heating it below its melting point (Yafie and Widyastuti, 2018). Grains ofparticles montmorillonite held together with a binder (a binding agent) is PVA (polyvinylalcohol). The choice of PVA as a binder is because PVA has an –OH group which can function to increase the adhesion betweenparticles montmorillonite (Prayitno, 2015). In addition, the use of PVA is expected to form pores in themembrane montmorillonite. The variation of the PVA concentration used was 0; 0.4; 0.8; 1,2; and 1.6% of the 20-gram mass of the mixture prepared. The use of PVA is limited to a concentration of 1.6% because with a large concentration of PVA it is difficult to dissolve in water and is too concentrated so that homogenization between montmorillonite powder and PVA solution is increasingly difficult.

Thegreen body membrane was printed using dry pressin techniques. In this technique, the mixture of membrane materials must be kept dry (Nugraha, 2018). Too much water content in the mixture may cause the mixture not to be printed or it will stick to the print. The pressure applied during theprocess dry pressing for all variations is kept constant. Too little pressure will cause the membrane to print poorly and become brittle. The mold used in theprocess is in the dry pressing form of a ring with a diameter of 4 cm. The green body that has formed issintered at 900°C for 3 hours. The results showed that as the PVA concentration increased, the shrinkage on the mass of themembrane montmorillonite was greater. The graph of the relationship between the concentration of PVA and burn loss is shown in Figure 3.

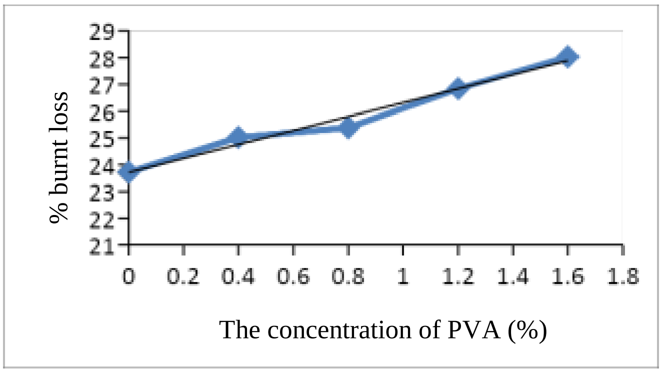

Figure 3. Graph of the relationship between PVA concentration and membrane mass

The membrane burnt loss occurs because there is evaporation from water and the PVA polymer used as a binder. The higher the concentration of PVA, the more PVA is lost and ultimately provide pore space for burn shrink. This is because PVA evaporates when the sintering is membrane montmorillonite carried out and forms pores in themembrane montmorillonite.

Density is a measurement of the mass per unit volume of an object. In this study, the density of the membrane was determined by the pycnometer method. The standard liquid used was kerosene. Kerocene was chosen because it is inert, does not react with membrane specimens and does not dissolve membrane specimens. In the CRC Handbook of Chemstry and Physics (1987), the density of kerosene at a temperature of 20°C and a pressure of 1 atm is 810 kg / m3. Kerosene density measurement results have shown the value of density of 789.2 kg / m3. The results of the kerosene density measurement are influenced by the room temperature and pressure when the measurement was made. The membrane density was measured by taking a few pieces of the membrane and then immersing it using kerosene for 2 minutes. Soaking for 2 minutes was carried out so that the kerosene really entered the pores of themembrane montmorillonite. The measurement results showed a decrease in the value of membrane density with increasing PVA concentration. This shows that increasing the concentration of PVA provides a lot of pore space in the membrane montmorillonite. The graph of the relationship between the PVA concentration and the density value shown in the Figure 4.

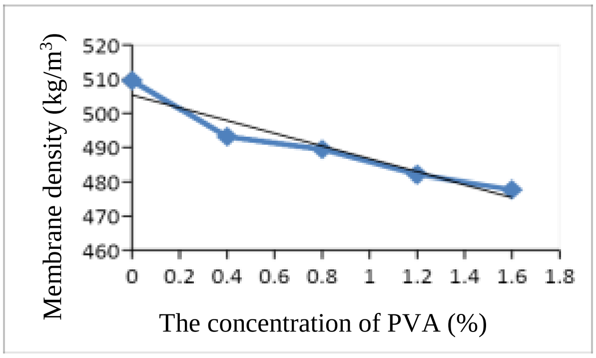

Figure 4. Graph of the relationship between PVA concentration and density of membranes

Flux is the amount of permeate volume that passes through the unit area of the membrane in a certain time. The permeability of the montmorillonite membrane is measured by passing the water feed to the membrane that has been installed in the dead-end reactor. Membrane flux measurements were carried out for 40 minutes. The results of the measurement ofmembrane permeability are montmroillonite shown in the graph of the relationship between the PVA concentration and themembrane flux value montmorillonite as follows.

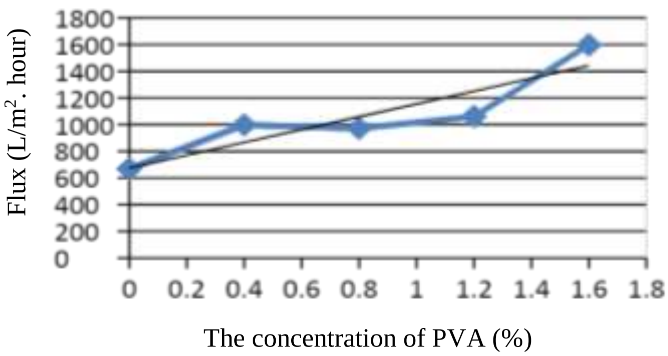

Figure 5. Graph of the relationship between PVA concentration and the value of membrane flux.

The results showed that the permeability value of the membrane increased with the increase in PVA concentration. The highest increase in the flux value of themembrane montmorillonite occurred at a concentration of PVA 1.6%, which was 1596.5679 L / m^{2}h. This shows that the increasing concentration of PVA, the more permeate that passes through the montmorillonite membrane. This is caused by the montmorillonite membrane has a lot of empty space or pores that the permeate can pass through. From the results of the measurement data, the membrane with a concentration of 1.6% PVA has more pores than the other concentrations. Therefore, amembrane montmorillonite with a concentration of 1.6% PVA was used in the filtration process of nickel and zink metal in liquid waste of electroplating. It is hoped that the membrane montmorillonite with a concentration of 1.6% PVA can filter nickel and zink metal. Furthermore, membranes montmorillonite with a concentration of 1.6% PVA were characterized by SEM-EDX before and after being used for filtration.

3.3. Filtration of nickel and zink metal in electroplating liquid waste using a montmorillonite membrane

The montmorillonite membrane which has been sinterized, continued to characterized by SEM analysis. SEM analysis was used to determine the morphological conditions of the montmorillonite membrane, the results are shown in Figure 6.

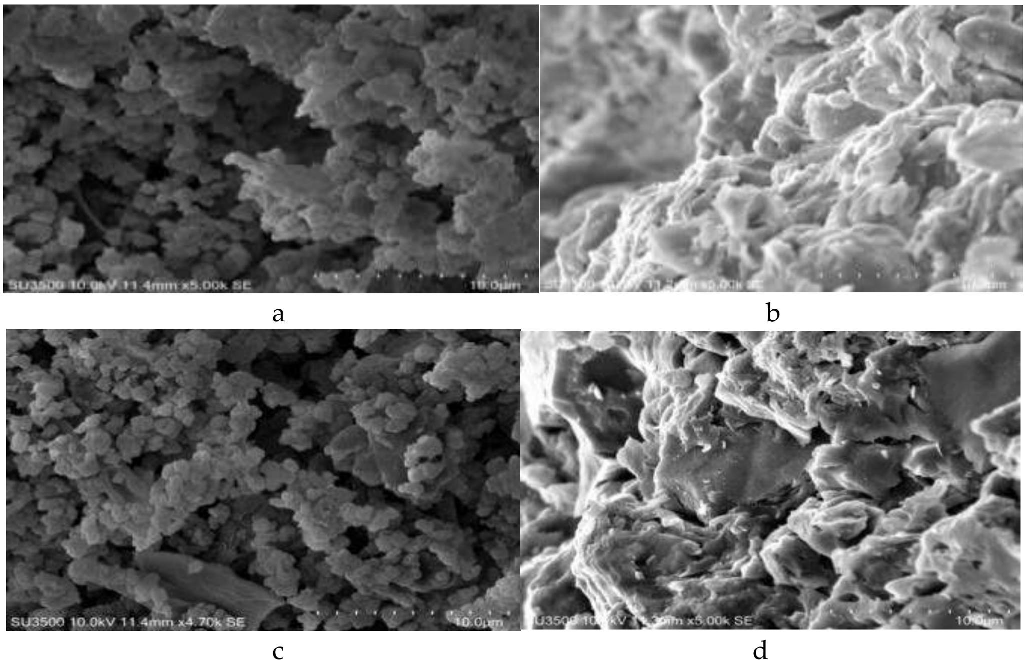

Figure 6. Morphological analysis of montmorillonite membrane, (a) surface before filtration and (b) cross section before filtration (c) surface after filtration (d) cross section after filtration

Based on Figure 6, particles of montmorillonite join each other. The montmorillonite membrane has many pores that are randomly scattered. The pores of the membrane are classified as micropores. Micropore membranes are membranes with pore sizes <2 nm (Arief, et al., 2013). This means that the montmorillonite membrane can repel particles larger than 2 nm in size. In Figure 6, the difference in the surface section of the montmorillonite membrane that has been used for the waste filtration on electroplating process. The pores on the montmorillonite membrane that have been used for filtration appear denser than the cross-section of the montmorillonite membrane before being used for filtration of electroplating waste. This means that themembrane montmorillonite can trap dissolved particles in the electroplating waste.

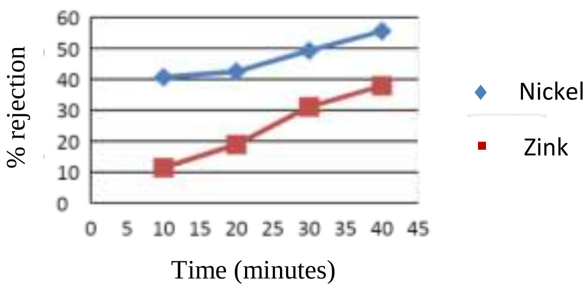

Figure 7. Graph of membrane rejection to nickel and zink metals

Permselectivity of a membrane is the ability of a membrane to hold or pass certain species. The parameter used to describe membrane selectivity is the rejection coefficient (Rahayu, 2017). The results showed that the selectivity of the membrane to nickel metal in electroplating liquid waste was 55.535% and zink metal were 37.8614%. The rejection of the membrane against the two metals was not large enough, this was because the metal ion size was smaller than the pore size of the montmorillonite membrane. Besides that, nickel metal rejection was higher than zink metal. This is possible because the content of nickel in electroplating liquid waste is more than the content of zink metal.

4. CONCLUSION

Based on the results of the research and data analysis can be concluded that the montmorillonite membrane synthesized by the method of dry pressing, with the concentration of PVA 1.6%, produces porous membrane. The characteristics of menvrane were (a) the value of flux 1596.5679 L / m^2h and (b) membrane density 477,707 kg / m^3. The rejection of membrane to nickel and zinc metals of electroplating liquid waste were 55.535% and 37.861% with an operating time of 40 minutes.

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