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Research Article

Reducing Silica Levels in WTP PLTGU X Wastewater using Rice Husk Filter Membranes

Penurunan Kadar Silika pada Air Limbah WTP PLTGU X Menggunakan Membran Filter Sekam Padi

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Abstract

During the water demineralization process, silica content is increased due to the failure of the chemical reagent. This research aimed to determine the effectiveness of silica reduction in the Water Treatment Plant (WTP) of PLTGU X by observing the permeate flux values relative to time and operating pressure variables, using ceramic membrane made from rice husk. To achieve the objective, ceramic membrane was made from rice husk additives, with a pore diameter of 365 nm and a surface area of 25 cm². The results showed that the composition ratio of clay, rice husk, and iron powder was 82.5%, 15%, and 2.5%, respectively. Furthermore, ceramic membrane with rice husk additives successfully reduced silica content from 1250 ppb to 890 ppb at a pressure of 1.5 bar and 90 minutes of operation and from 1250 ppb to 710 ppb at 2 bar and 90 minutes of operation. This suggested that wastewater could be processed again in the demineralization plant to produce demineralized water. The best membrane performance in the filtration process was achieved at 90 minutes with a pressure of 2 bar, which successfully reduced silica content by 43.2%, with a permeate flux of 3.44 L/m².
Keywords: filter; membrane; rice husk; silica

Abstrak

Tujuan penelitian ini yaitu untuk mengetahui efektifitas penurunan silika pada air limbah PLTGU X dilihat dari nilai fluks permeat terhadap variabel waktu dan tekanan operasi dengan menggunakan membran keramik dari sekam padi. Metode penelitian yang digunakan yaitu dengan membuat membran keramik berbahan aditif sekam padi dengan diameter pori 365 nm dan luas permukaan 25 cm^2 dengan perbandingan komposisi tanah liat, sekam padi dan serbuk besi terdiri dari; 82.5%; 15%; 2.5%. Membran keramik dengan aditif sekam padi berhasil menurunkan kadar silika dari 1250 ppb menjadi 890 ppb pada tekanan 1,5 bar dengan waktu 90 menit dan dari 1250 ppb menjadi 710 ppb pada tekanan 2 bar dengan waktu 90 menit dan air limbah tersebut dapat diproses kembali di demin plant untuk menjadi air demineralisasi. Kinerja membran terbaik dalam proses filtrasi yaitu pada waktu 90 menit dengan tekanan 2 bar yang dapat menurunkan kadar silika 43,2 % dengan fluks permeat sebesar 3,44 L/m^2.
Kata Kunci: *filter; membran; sekam padi; silika*

1. Introduction

The working principle of membrane is to act as a thin and highly selective barrier that separates two or more components of a fluid flow. In this context, the flow in membrane occurs due to the driving force, which can be the convection or diffusion of each molecule [1][2]. This has led to a significant increase in the use of porous ceramic membrane, suggesting potential opportunity for natural materials [3]. Ceramic membrane is usually made from clay, which has a high porosity level with a composition of hydrous aluminum silicate ($Al_2O_3.2SiO_2.3H_2O$) and metal oxides such as Fe_2O_3, MgO, and K_2O, as well as ions that can be exchanged with other ions [4]. In general, membrane technology is used to separate solutes from solvents, for example, separating silica content in water.

Water containing high levels of silica is considered unfit for use in the PLTGU (Gas and Steam Power Plant) process. This is due to the production of silica crust that causes blockages in the steam pipe to the turbine. Therefore, silica content in boiler water should be controlled by using filter membrane.

Generally, synthetic membrane is used in industries such as polyamide, polysulfone, and polycarbonate [5][6]. However, there is natural membrane found in living cells, such as cellulose and its derivatives, including cellulose acetate. Previous research has proven that cellulose acetate membrane from nutmeg seed shells using the phase inversion method produces characteristic tests that can be applied further [7]. This derivative is obtained from Nata de Coco with the addition of polyethylene glycol additives, which meet the requirements as ultrafiltration membrane. [8] There is also a derivative from banana stems and water hyacinth that can be used in the purification of drilled well water [9].

As an effective method, PLTGU X currently uses silica filter membrane made from empty oil palm bunch waste. According to Bramanta, oil palm fiber and shell ash have many minerals, one of which is silica at 27.5% and 36.1% [10]. The research by Laelasari stated that rice husk ash had silica content of approximately 16.98% [11]. Silica ash can be produced from the controlled burning of rice husk at high temperatures above 650°C. The weight of rice husk is approximately 20% of the weight of the grain, with 15% as ash. With a high silica content, it shows that the performance of membrane made from rice husk additives is quite good [12].

The water filtration process usually uses zeolite and activated carbon compounds. Silica (SiO_4) and Alumina (AlO_4) are the main minerals in the zeolite compounds that have cavities. After modifying by adding clay, white Portland cement, and PVA, membrane has advantages in the filtration process because of high selectivity to Mn ions [13]. Additionally, natural zeolite is also easy to obtain with selective filtration capabilities and molecular-sized pores [14]. In this context, rice husk is very likely to be used as an alternative material for processing waste from various industries because of silica content [15].

Based on the background, this research aimed to determine the effectiveness of silica reduction in PLTGU X wastewater from the permeate flux value against time and operating pressure variables, using ceramic membrane from rice husk. To achieve the objective, zeolite was combined with rice husk containing silica to improve the performance of ceramic membrane in removing silica levels in PLTGU X wastewater. The effectiveness of this method was determined by knowing the reduction in silica levels using a filter membrane from rice husk in PLTGU X wastewater to be reprocessed into deionized water at the PLTGU X. The results were expected to maximize the use of abundant rice husk waste, specifically in agricultural areas such as South Sumatra.

2. Research Methods

2.1. Tools and Materials

The materials used in this research consisted of rice husk, clay, zeolite, iron powder, ammonium molybdate solution, oxalic acid solution, hydrochloric acid solution, ascorbic acid solution, and distilled water. Meanwhile, the instrumentation tools included Scanning Electron Microscopy (SEM), FTIR Spectrometer, and UV-Vis Spectrophotometry. The sample used was PLTGU X wastewater.

2.2. Data Collection Methods

The data obtained were sourced from literature related to the research conducted, both from the company and outside, such as books, journals, and websites. Besides the literature review, the data listed were also obtained directly from the PLTGU X Laboratory, discussion, and Q&A activities, whose sources were obtained from PLTGU X employees.

2.3. Making Ceramic Membrane

In this research, ceramic membrane made was cylindrical, including the printing and sintering processes. Membrane was made with two variations of the mixture composition where the first was from clay, rice husk additives, zeolite, and iron powder with a composition ratio of 82.5%, 7.5%, 7.5%, and 2.5%, respectively. The second was made from clay, zeolite, and iron powder with a composition ratio of 82.5%, 15%, and 2.5%, respectively, printed using a cylindrical mold made of stainless steel and compacted for 15 minutes. This treatment was necessary to ensure the pressure given could be evenly distributed on membrane and sintered at a temperature of 500^{o}C.

Ceramic membrane was used for wastewater treatment by varying the feed flow rate at a pressure difference (ΔP) of 1.5 and 2 bar. Observations were made at different operating times ranging from 15, 30, 45, 60, 75, and 90 minutes.

2.4. Silica Analysis

Silica analysis was carried out using UV-Vis Spectrophotometry. A total of 2 mL of 7.5% (w/v) ammonium molybdate solution and 1 mL of 1:1 HCl were added to 2 mL of sample and 50 mL of distilled water. Furthermore, 15 ppm of ortho-phosphate was added to the boiler feed. Ammonium molybdate in acidic conditions was reacted with silica and ortho-phosphate to produce heteropoly acid. The resulting molybdosilicic acid was then reduced with 1 mL of 10% (w/v) ascorbic acid to make a blue complex compound whose absorbance was measured at a wavelength of 660 nm. A 2 mL of 10% (w/v) oxalic acid solution was added to reduce molybdophosphoric acid without removing molybdosilicic acid. Moreover, it should be observed that reactive silica removed all silicic acid polymers ($SiO_2.H_2O$).

2.5. Permeate Flux Analysis

Permeate flux usually showed whether membrane was performing optimally. Additionally, membrane performance was determined based on the amount of permeability, selectivity to certain chemical compounds, and the rejection percentage of unwanted compounds in the feed. The permeate flux value was obtained by Equation 1.

$$Jv = \frac{V}{Axt} \quad (1)$$

Jv represented the permeate flux in $L/m^2 \cdot hour$; V portrayed the permeate volume in Liters; A showed membrane surface area in m^2, and t indicated the time in hours.

2.6. Ceramic Membrane Filtration Test

Wastewater was fed into a 500 L storage tank. This test used a 100 L sample of PLTGU X blowdown wastewater. With the help of a pressure pump, wastewater from the storage tank was flowed into housing-1 containing a sponge filter with a pore diameter of 0.5 µm. The pump pressure was set at 1.5 and 2 bar by regulating the feed flow rate using a feed flowmeter. Subsequently, water filtered with a 0.5µm pore diameter sponge filter was flowed into housing-2 and 3, each containing a sponge filter with a pore diameter of 0.1 µm and activated carbon.

As shown in Figure 1, wastewater passed through the sponge filter, and activated carbon was returned to housing 4, which contains rice husk ceramic membrane. Subsequently, wastewater that had passed through membrane process was collected in a container as permeate. Sampling was carried out every 15, 30, 45, 60, 75, and 90 minutes. Each sample was calculated for the volume of permeate produced and analyzed for silica content in the permeate. The testing process is shown in Figure 1.

3. Results and Discussion

3.1. Membrane Performance Analysis

3.1.1. Permeate Flux Analysis

Membrane performance is better when the permeability value and selectivity level of membrane are greater. Sylvani et al. stated that the number of membrane pores affected the permeate flux value and water permeability [16]. When water flows easily through membrane, the permeability value is higher. Meanwhile, thicker membrane makes it difficult for water to pass through, reducing the permeate flux value.

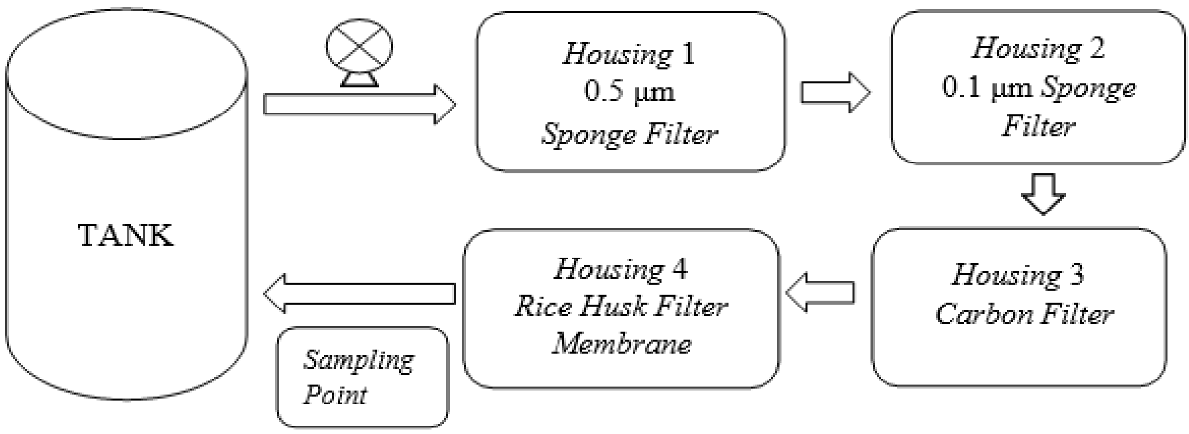

Figure 1. Membrane Filtration Testing Flow Chart

In this research, the best membrane permeate flux value for PLTGU X wastewater was obtained at a pressure of 2 bar with rice husk additives at the 15th minute of 18.08 L/m^2. Meanwhile, membrane without additives was obtained at the 15th minute with a pressure of 1.5 bar, which was only 15.68 L/m^2. This is due to the influence of pressure and time passed by membrane, which has not reached the saturation point. According to Diana et al., the lower permeate flux results are due to differences in the compounds in ceramic membrane, affecting the pores on membrane surface [17].

From Tables 2 and 3, longer time makes more impurities settle on membrane surface, which causes the permeate flux value to decrease. Meanwhile, the effect of pressure shows that the permeate flux increases with higher operating pressure. This occurs because of the influence of the main driving force of membrane operation. When pressure is applied, particles with a size smaller than the pores will easily pass through membrane pores. Meanwhile, larger particles, such as contaminants, will remain in it. As a result of the applied driving force, the pores can also enlarge, which causes membrane pores to become larger. Therefore, the solution feed rate is faster and passes through membrane.

3.1.2. Analysis of Silica in Liquid Waste of PLTGU X

Silica can cause fouling in boiler pipe parts when carried by water or hot steam at high temperature and pressure. When this happens, the boiler's heat transfer capacity will decrease, causing uneven heat transfer. Silica dissolved in hot steam will settle on the turbine fins at low temperatures. As shown in Table 1, it can be observed that the use of ceramic membrane made from rice husk additives is capable of reducing silica values. Based on the results of laboratory tests, the best reduction in silica levels was obtained in silica filter membrane made from rice husk additives, which were able to reduce by 43.2% from 1250 ppb to 710 ppb within 90 minutes at a pressure of 2 bar. Meanwhile, for membrane without rice husk additives, it was only able to reduce silica levels by 23.68% or 296 ppb.

Table 1. Silica Level Reduction Data

Time (Min)	1.5 Bar		2 Bar	
	With Rice Husk (ppb)	Without Rice Husk (ppb)	With Rice Husk (ppb)	Without Rice Husk (ppb)
0	1250	1250	1250	1250
15	1120	1241	1100	1220
30	1050	1233	950	1210
45	980	1190	895	1130
60	920	1150	800	1000
75	900	1170	760	993
90	890	1123	710	954

Table 2. Ceramic Membrane Permeate Flux Value at 1.5 Bar Pressure

Time (Min)	Permeate Volume		Permeate Flux	
	With Rice Husk (L)	Without Rice Husk (L)	With Rice Husk (L/m^2)	Without Rice Husk (L/m^2)
15	0.0105	0.0098	16,80	15,68
30	0.0109	0.0095	8,72	7,60
45	0.0109	0.0098	5,81	5,23
60	0.0105	0.0095	4,20	3,80
75	0.0109	0.0098	3,49	3,14
90	0.0105	0.0098	2,80	2,61

Table 3. Ceramic Membrane Permeate Flux Value at 2 Bar Pressure

Time (Min)	Permeate Volume		Permeate Flux	
	With Rice Husk (L)	Without Rice Husk (L)	With Rice Husk (L/m^2)	Without Rice Husk (L/m^2)
15	0.0113	0.0080	18,08	12,80
30	0.0105	0.0081	8,40	6,48
45	0.0125	0.0083	6,67	4,43
60	0.0136	0.0085	5,44	3,40
75	0.0129	0.0087	4,13	2,78
90	0.0129	0.0085	3,44	2,27

Based on the results, membrane without rice husk additives were observed to be less effective in reducing silica levels. From Table 1, there are non-linear results at minute 75 at conditions 1.5. This is because there is seepage in the column, which causes PLTGU X wastewater not to pass through membrane despite the effectiveness. The operating pressure conditions also affect silica levels, as 2 bar has greater effect than 1.5 bar. This is possibly due to the influence of pressure, which enlarges membrane pores, causing a high potential for impurity particles to escape from membrane surface.

The adsorption process using rice husk membrane containing silica content greatly affects the reduction of silica. During this process, the phenomenon of concentration polarization occurs due to blockage of membrane pores by silica. This causes silica concentration on membrane surface to be higher than concentration passing through membrane. The polarization occurs as a result of the diffusion of substances moving towards the surface or through membrane, which causes the accumulation of substances on the surface. In areas farther from membrane surface, the concentration of substances remains lower. Additionally, a dialysis process occurs, namely the transfer of silica molecules from the solution due to diffusion through membrane.

3.1.3. SEM Analysis on membrane Surface

Photomicrographs of ceramic membrane with a composition of clay, zeolite, and iron powder are shown in Figure 2. Meanwhile, ceramic membrane with a composition of clay, rice husk additives, zeolite, and iron powder is shown in Figure 3.

The morphology between ceramic membrane with and without rice husk additive shows significant difference. The use of rice husk additives produces membrane surface pores that are dense and evenly distributed. Although ceramic membrane with rice husk has a random and non-uniform pore structure and different grain boundary sizes and ratios, it is better than sample without additives. The more pores formed from the addition of rice husks, the more pollutant material or particles are expected to be blocked on the membrane surface.

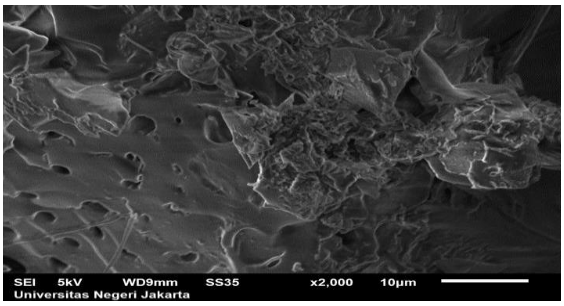

Figure 2. SEM Image of Ceramic Membrane without Rice Husk Additive at 2000x Magnification

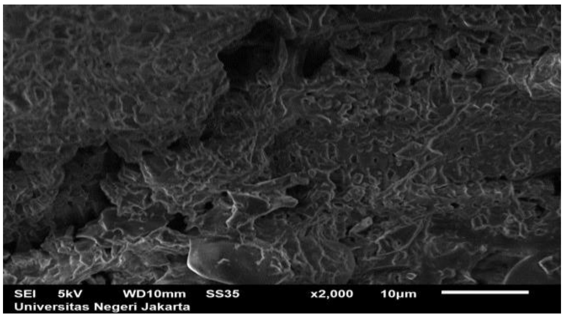

Figure 3. SEM Image of Ceramic Membrane with Rice Husk Additive at 2000x Magnification

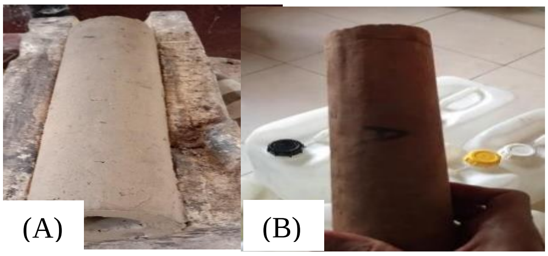

Figure 4. Ceramic Membrane (A) with Rice Husk Additives, (B) without Rice Husk Additive

Based on the ceramic membrane images in Figures 4, the addition of rice husk additives makes membrane color more inclined to that of rice husk. Meanwhile, membrane without rice husk tends to resemble clay. On both membrane, the distribution of iron powder can also be observed with visual black spots on membrane surface.

The FTIR results (Figure 5) of rice husk show that the ceramic membrane with rice husk additives contains H stretching vibrations in the form of -OH and NH in the wave range of 3417.67 with an intensity of 92.02%. This wide peak shows the presence of OH groups originating from carbohydrate compounds that are components of rice husk. The fingerprint area shows inorganic compounds -C-NO_2- in the wave range of 1352 with an intensity of 87.59% [18]. These organic compounds may be formed from membrane combustion process at high temperatures.

4. Conclusion

In conclusion, the effectiveness of reducing silica content in PLTGU X wastewater with silica filter membrane made from rice husk additives showed an optimal result at 90 minutes with a pressure of 2 bar. Silica content was reduced by 43.2% with a permeate flux of 3,44 L/m^2.

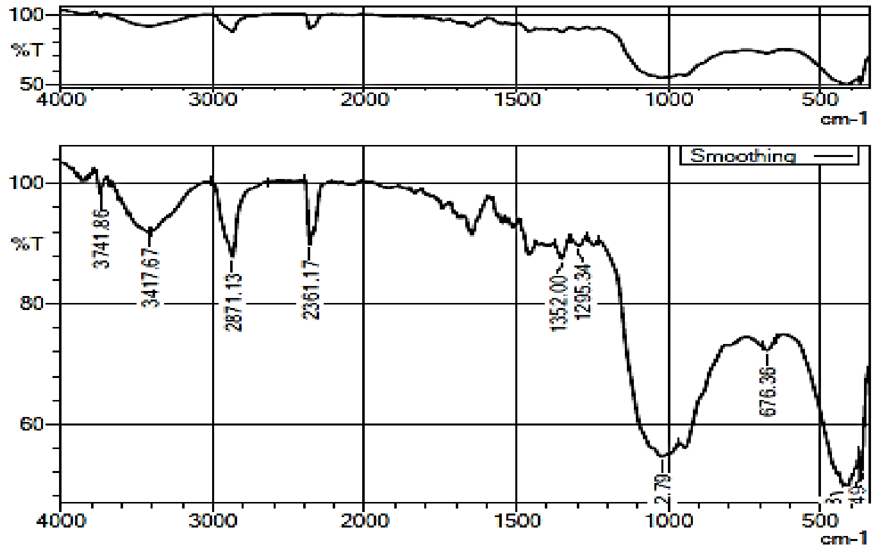

Figure 5. FTIR Analysis of Rice Husk

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