Removing *Escherichia Coli* Bacteria in River Water using Ceramic Membrane from Mixed Clay and Fly Ash Material

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**Abstract.** The existence of water lately is quite alarming, especially in urban areas. The increasing of river water pollution through industrial and domestic waste, herbicide/pesticide and also the contaminant left-over cause the quality of river water getting worse beyond the allowance limit of standard for clean and drinking water. The most important criteria in examining clean and drinking is the ingredient test of microbiology with *Escherichia Coli* (*E. Coli*) bacteria, which is a pathogenic bacteria to human, as the indicator. This study aims to remove *E.Coli* bacteria in water river in Geudong, Lhokseumawe using membrane filter made of ceramic mixturing with clay and fly ash in various compositions; clay:fly ash (40%:60%), clay:fly ash (50%:50%) and clay:fly ash (60%:40%). The ceramic membrane was made with sintering method in 700°C temperature for 4 hours and the filtration test was done in varios of operating pressure; 0,25; 0,50; 0,75, 1,00 and 1,25 bar. The result of the study showed that ceramic membrane with the composition of clay-fly ash (40%:60%) was able to reject *E.Coli* up to 99,048% with operating pressure 0,25 bar. Using Scanning Electron Microscopy (SEM) test, it is known that the ceramic membrane with the composition of clay-fly ash (40%:60%) is included as microfiltration membrane with pore size 1.6 µm – 2 µm.

1. **Introduction**

The increasing of pollution through industrial and domestic waste cause the quality of river water worse beyond the allowance standard limit. One of the most important criterias to examine the quality of drinking water is based on the ingredient test of microbiology of *Escherichia coli* bacteria which is a pathogenic bacteria for human. Reviewing from bacteriologic requirements, adding more chlor in chlorination process, a conventional process in water treatment, will reduce the amount of *E.Coli* bacteria in water, but it is needed to be warried about the existence of organic materials with low molar mass, this chlor residue will bind and become a compound that will raise the risk to form THM (Trihalomethanes), that can cause cancer for human, during the chlorination process and this will make the water quality become worse. Several technologies of organic membrane for water treatment have been developed nowadays. *Clay* can be easily found in nature while *fly ash* which is the side waste of cement manufacture can be made as the mixture material for making ceramic membrane with low cost. Beside, the characteristic of *fly ash* and *clay* which have 1µm - 20µm microfiltration membrane can be used to remove bacteria in river water [2].

Some studies about *clay-fly ash* membrane for water treatment have been done previously. In 2013, Subreyer in his study stated that clay-fly ash membrane can remove the TDS level, Fe and Mn in water as required by drinking water standard No.492/Menkes/PER/IV/2010 [3]. Nurhayati, C and Susanto, T in 2015 analyzed the effect of fly ash-clay composition in peat water processing and they
concluded that the composition of 50%:50% fly ash-clay is capable to process peat water become clean water optimally [4]. The influence of sintering temperature in the anorganic membrane performance has also observed by Elfiana in 2017 which said that the anorganic membrane can reject turbidity of peat water up to 94, 17 % in temperature 700°C [5]. From those studies, the parameter of E.coli is not yet been analyzed as the requirement of drinking water as demanded by government regulation No.492/Menkes/PER/IV/2010 , for that reason, this research used the ceramic membrane filter with the mixture of clay-fly ash using various compositions ; 40%:60%, 50%:50% dan 60%:40% in sintering temperature 700°C and operating pressure 0.25; 0.50; 0.75, 1.00 and 1,25 bar to remove e.coli bacteria in water river.

2. Research method

2.1 Material and equipment
The material and equipment used in this research are; feed solution of Geudong river, clay, fly ash originated from side product of cement manufacture ,PT. SAI Banda Aceh, PVA (Polyvinyl Alcohol), aquades, and Brilliant Green Lactose Bile Broth (BGLBB), Housing membran, tubular mold for membrane with external diameter 6,5 cm, internal diameter 2,5 cm and height 25 cm), durham tube, analytical weigher, , furnace, autoclave, laminer flow, incubator, oven, and a set of membrane modul for flux and filtration test processes and SEM (Scanning Electron Microscope) merk JEOL JSM-651OLA which is used in analyzing membrane morphology structure.

2.2 The synthesis of clay:fly ash ceramic membrane
900 gram total amount of clay and fly ash mixture with various compositions : clay:fly ash (40%:60%), (50%:50%) and (60%:40%) was filled to beaker glass then Polyvinyl Alcohol (PVA) was added for about 36 gram. 700 ml water then aded to the mixture (clay, fly ash and PVA), little by little until it run out, at the same time the stirring process was done until the mixture became a paste of clay-fly ash-PVA. Then the paste was poured into the mold for form the membrane. The mold was then pressed by pressing tool slowly to keep it free of air. After 7 x 24 hours, the membrane was taken from the mold for then continued with the sintering cess in 700°C for 4 hours.

2.3 The removing process of E.Coli Bacteria using clay:fly ash ceramic membrane
The equipment was settled according to the research concept which along with cross flow filtration system (Fig 1). The equipment test was conducted by passing the pure water through the membrane to find out the possibility of leak and the equipment failure before the test based on the research variable. Before the filtration using MPN (Most Porbable Number) method, the amount of e.coli bacteria in water river was analyzed. Then the water was streamed through the hole of feed solution to the membrane surface with operating pressure variations ; 0,25; 0,5; 0,75; 1,0; 1,25 bar as water propulsive force through the membrane. The filtrate (permeate) passed the membrane was then binned and its volume was measured and then the rejection percentage of E.Coli of the filtrate was analyzed.

2.4 Permeability Test
The permeability of clay-fly ash ceramic membrane was tested by streamed the pure water (aquades) through the membrane modul by varying the operating pressure 0,25; 0,50; 0,75, 1,00 and 1,25 bar. The flux resulted of each operating pressure was calculated based on the following equation:

\[ J = \frac{V}{A \cdot t} \]

(5)

Where:
- \( J \) = Flux ( l/m².hr)
- \( V \) = permeat volume (ml)
- \( A \) = Area of membrane (m²)
- \( t \) = Time (hours)
2.5 Bacteria E. Coli analysis

E. Coli bacteria was analysed with MPN (Most Probable Number) method by filling 9 ml Brilliant Green Lactose Bile Broth (BGLBB) solution in to 9 (nine )test tubes that consist of three (3) series and each of them are consist of three (3) test tubes. The samples was then poured to the first series of tubes for 1 ml then sample was stirred until it is homogen. The solution of each tube from the first seri were then filled into each test tube of the second seri for 1 ml, stirred until it is homogen too. The 1 ml solution from each test tube from the third seri was separated and removed. Those nine test tubes then were incubated in temperature of 44 °C for 24 hours. After that, the E.Coli bacteria can be identified by looking at the gas formed in the durham tube and the result then was confirmed in MPN table.

2.6 Test of Membrane Characteristic

The ceramic membranes with each composition used to filtration process in removing E.Coli bacteria in water river that gives the best rejection will then be analyzed its morphology structure and the its pore size using SEM merk JEOL JSM-651OLA.

3. Result and Discussion

3.1 Membrane Permeability

The ceramic membrane permeability is the ability of the membrane to stream the water assisted with propulsive force; the operating pressure in ceramic membrane. Aquades streaming in the ceramic membrane is aimed to the ceramic pre-washing to remove the remaining dirt from the combustion process to prevent it mix with the permeat of the ceramic membrane [6]. From Figure 2, it can be seen that the flux value is increasing along with the increasing of pressure. The filtration process used aquades occurs constantly in long hours and the big volume due to the inexistence of particle that adhere or pile up on the wall or the pores of the ceramic membrane.
Figure 2. The aquades permeability to the pressure of ceramic membrane with composition clay:fly ash $M_1$ (40%:60%), $M_2$ (50%:50%), dan $M_3$ (60%:40%).

3.2 Flux of Clay-Fly Ash Ceramic Membrane

Figure 3 showed the raising of flux value which increases along with the increasing of the pressure, where the separation occurred because of the driving force caused by the different of input and output pressure of the membrane modul. The highest flux value was gained at the pressure of 1.25 bar for every ceramic membrane ($M_1$, $M_2$ and $M_3$). The increasing the the flux is caused by the increasing of the pressure which resulted to the increasing of the propulsive force given to the solution to pass the ceramic membrane so a big amount of feed solution can pass the ceramic membrane and this process will minimize the effect of concentration polarization.

Looking at the composition of $M_1$, $M_2$ dan $M_3$, it can be concluded that the ceramic membrane with $M_3$ composition gives the highest flux value compared to other compositions ($M_1$ and $M_2$), this is because the $M_3$ composition contains the biggest amount of clay. Clay can increase the number of pores and holes to support the performance of filtration process so many of feed solution can get in through the pores and pass the ceramic membrane. More clays will increase the physical characteristic of the membrane which are the strenghtness and unfragile. The high value of flux is also affected by the combustion which is aimed to open the pore of the ceramic membrane [7]. The addition of PVA is not only as the adhesive, but also is aimed to reduce the density and to enlarge the porosity of the ceramic membrane. This is due the the polymer characteristic of PVA which evaporate in high temperature. [8].

Figure 3. The effect of pressure to ceramic membrane flux with compositions clay:fly ash $M_1$ (40%:60%), $M_2$ (50%:50%), and $M_3$ (60%:40%) in river water.
3.3 E. Coli Rejection

E. Coli rejection is the ability of ceramic membrane to hold E. Coli which enter the feed solution. The rejection process is greatly influenced by the operating pressure and the composition of the membrane materials. It can be proven from the result of rejection measurement which show the higher pressure given, the lower rejection result gained as seen in Figure 4 where the highest percentage of E. Coli rejection is obtained in pressure of 0.25 bar; 99.048% and the lowering percentage of E. Coli rejection in pressure of 1.25 bar. This phenomenon is caused of bigger propulsive force which lead to the retention of some molecules in the membrane surface that pressed and get in to the membrane pores and become the permeat. The lowering percentage of E. Coli in pressure increasing is also caused by the ceramic membrane process which occurs generally flux phenomenon that inverse with selectivity or the rejection of ceramic membrane. The higher flux will decline the rejection of ceramic membrane and inversely. While the expected thing in ceramic membrane process is to higher the flux and its selectivity [9].

![Figure 4](image_url)

**Figure 4** The effect of pressure to E. Coli rejection in river water with membrane compositions: clay:fly ash M₁ (40%:60%), M₂ (50%:50%), and M₃ (60%:40%).

The parameter of E. Coli rejection is also influenced by the organizing components of the ceramic membrane itself, distribution structure and pore size of the membrane. Seen from the percentage value of E. Coli rejection in Figure 4 it show that the ceramic membrane with the mixture of clay-fly ash (40%:60%) with 0.25 bar pressure is able to reject E. Coli bacteria up to 99.048%. Fly ash can produce the pores in the ceramic membrane with 0.1 μm hence the membrane can hold bigger E. Coli bacteria with 0.7 μm size. Besides, the clay contained in the membrane can increase the number of pore and hold to support the filtration performance which makes more of feed solution can get in to the ceramic membrane pores and strean the membrane. This is also strengthened by adding PVA which functions as the binder to increase green strength of the ceramic for 4% of the total weight of the ceramic membrane, PVA has −OH group which function as the adhere among the ceramic particles. Clay also play the important role because it repaires the physical characteristic of the ceramic membrane to be more strong, crystalline and not easily broken up [4].

3.4 Morphology analysis of membrane

Analysis of membrane morphology is aimed to observe its characteristics covering the structure of the membrane and the transection to see the pores statistic which is the pore size of the membrane. The analysis uses SEM (Scanning Electron Microscope) by taking the ceramic membrane sample that produces the best percentage of E. Coli rejection and it is the ceramic membrane of M₁ with the composition clay:fly ash (40%:60%). The analysis is conducted before the filtration. The result of SEM analysis for the pore structure can be seen in the Figure 5.
Figure 5. The result of morphology analysis of M_1 clay:fly ash (40%:60%) membrane before filtration

The Figure 5 shows the pores of the membrane before the filtration, the condition of the pores are very small, it means that the M_1 membrane is very potential to be applied in water purification process. The size of pores area is between 2,011 µm^2 to 4,524 µm^2 hence the diameter of the M_1 membrane pores is 1.6 µm – 2 µm. This result shows that the ceramic membrane with clay:fly ash (40%:60%) is included as microfiltration membrane category and it is effective to reject E.Coli bacteria in river water.

Conclusion
Based on the research, it can be concluded that the highest flux resulted is 207,898 l/m².hour in 1.25 bar of pressure to the ceramic membrane with clay:fly ash (60%:40%). The highest percentage of E. Coli rejection is 99.048% in 0.25 bar of pressure for membrane with clay:fly ash (40%:60%). From the morphology analysis, the result shows that the ceramic membrane with clay:fly ash (40%:60%) has pores size 1.6 µm – 2 µm and it is included as microfiltration membrane.

References
[1] Wenten I G 2015 Teknologi Membran dalam Pengolahan Air dan Limbah Industri. Studi Kasus: Pemanfaatan Ultrafiltrasi untuk Pengolahan Air Tambak, https://www.researchgate.net/publication/281236127.
[2] Subriyer N, Rizka Wulandari and Jelita Intan 2011 Jurnal Teknik Kimia. 17 26-27
[3] Subriyer N 2013 Journal of Engineering Studies and Research.19 71-75
[4] Nurhayati, C and Susanto T 2015 Jurnal Dinamika Penelitian Industri. 26 95-105
[5] Elfiana, A Fuadi and S Diana 2018 Materials Science and Engineering. 345
[6] Apriyanti E and Wijayanto W 2017 Prosiding Seminar Nasional Kimia UNY. p 305-312
[7] Sandra K O, Budi A S and Susilo A B 2014 Prosiding Pertemuan Ilmiah XXVIII HFI Jateng & DIY Yogyakarta. p 392-395
[8] Nasir S, Hartaty A and Sulaiman D 2013 Jurnal Teknik Kimia. 19 7-13.
[9] Agmalini S, Lingga N N and Nasir S 2013 Jurnal Teknik Kimia. 19 59-68.