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Development of Chitosan-based forward Osmosis Membranes for Emergency Drinking Water Supply

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Abstract - Development of forward osmosis (FO) membranes became one of the alternative methods for drinking water supply in an emergency. In this research, drinking water bags based on FO membrane have been developed using chitosan as the basic material of the membrane. The chitosan membrane used for the manufacture of drinking water bags has a thickness of 0.043 mm, 30.3% porosity, tensile strength 28.83 kgf / mm2, swelling degree 43.5% and elongation of 7.16%. Drinking water bags are made with a combination of Polypropylene Plastic (PP) and aluminum foil with FO membrane inside. This drinking water bag can be applied for brackish water purification to be energy drinking water as one of the solutions for the drinking water supply in emergencies. FO process testing is done by using a variation of sugar solution as the draw solution that are glucose, fructose, sucrose, and mixture. The concentration of applied draw solution is 1, 2 and 3M within 1-hour treatment. The highest water flux was obtained in 3M sucrose solution with a flux value of 5.25 L/m^2hour. The results of drinking water quality parameters analysis in the form of pH, Total Dissolved Solids (TDS), salinity, conductivity, heavy metals, and *Escherichia coli* (*E. coli*) contents showed that the FO water product meets drinking water quality standards based on Indonesian government regulation of drinking water quality. FO-membrane-based drinking water bags are potentially used as an alternative solution for the energy drinking water supply in an emergency.

Keywords: forwards osmosis membrane, drinking water, chitosan membrane, emergency, water bag

Introduction

Water is a natural resource that is needed for various needs of human life, both domestic, agricultural, and industrial (Ardiansyah and Kusumo, 2013). Access to clean water today, especially for drinking water is still very limited, even though surface water available in nature is quite abundant. The availability of water on the earth more than 97% is sea water that cannot be consumed directly by humans, as many as 2% is stored as glaciers and water vapor, which also cannot be used directly. Water that is truly available for human needs is only 0.62%, including water found in lakes, rivers, and groundwater. Regarding quality, water available for human consumption is only 0.003% (Budiyono and Sumardino, 2013). This condition is due to a large number of polluted water sources that are not suitable for consumption, while the need for water continues to increase to meet human consumption globally (Su et al., 2012). The unavailability of drinking water can cause dehydration, and metabolic processes are disrupted, attacked by various diseases and even lead to death. This condition is makes drinking water a critical basic need and must be available even in emergencies (Haryoto and Hidayat, 2007).

Indonesia is one country that has great potential for natural disasters. Geologically, Indonesia is located between two sets of world mountain ranges, the Pacific and Mediterranean circuits, while geographically, Indonesia is crossed between Asia and Australia and between the Pacific Ocean and the Indian Ocean. This condition has caused Indonesia to be vulnerable to various natural disasters, such as earthquakes, tsunamis, volcanic eruptions, floods, landslides, and various other types of natural disasters (Haryoto and Hidayat, 2007).

The most significant impact of the disaster is a creation of emergency conditions that threaten and disrupt the lives and livelihoods of the community, one of which is the availability and quality of clean water which is decreasing, especially drinking water. This situation is due to contamination or damage to the infrastructure of drinking water sources as well as water delivery that is hampered due to disruption of the transportation system which is usually used to supply drinking water. The handling that has been done so far is by sending clean water tank trucks, but it is often constrained due to the limited number and access, especially to reach isolated areas (Steele and Clarke, 2008). For this reason, it is necessary to develop an alternative method that is easier, cheaper, affordable and appropriate as a solution for drinking water supply, especially to respond to emergencies that are very likely to occur, for example in disaster areas, water crisis areas, sea trips, backpackers and conditions — other emergencies. One of them is by procuring drinking water bags based on membrane technology through *Forward Osmosis* (FO) process (Cormick et al., 2008).

The FO process is an alternative method that is being developed to improve the efficiency of water purification processes. FO method is a purification process where water from a solution with low concentration will move to a solution with a high concentration through a semipermeable membrane due to the osmosis gradient (Cath *et al.*, 2006). The advantage of this method is that it does not require external pressure (hydraulic), the possibility of fouling on membranes is small, low possibility of contamination and does not require electricity in its application so that it is simpler, cheaper and practical to use (Cormick et al., 2008; Shaffer *et.al*, 2015). FO membranes can be made from natural polymer materials or synthetic polymers. One of the natural polymers that can be used is chitosan (Saiful et al., 2015). Chitosan is a natural biopolymer derived from chitin, a polysaccharide, which has the second largest availability after cellulose, so the use of chitosan as a membrane offers high economic value and is more environmentally friendly (Mak and Sun, 2008). In this study the development of chitosan membranes that have been made previously into drinking water bags has been carried out. The FO process is carried out by using a variety of sugar solutions as draw solution, in the form of glucose, sucrose, fructose, and mixtures, so that later water will be obtained with a certain amount of sugar which is expected to be energy drinking water in an emergency. The bag of drinking water produced has been tested as a medium for purifying surface water in the form of brackish water.

Material and Method

Materials

All chemical were purchased from suppliers company for pro analysis. The chemicals used are chitosan, sodium hydroxide, acetic acid, dimethylformamide, distilled water, glucose, fructose and sucrose and Polypropylene (PP) plastic and aluminum foil plastic. The equipment used in this study were a magnetic stirrer, oven, beaker, measuring cup, Erlenmeyer, ceramic, double tape, glue, scissors, sealing machine, pH meter CT-6022, Salinity Meter SA287, Digital Electrical Conductivity Meter WTW LF320, TDS meter, SEM TESCAN VEGA3 LMU, and AAS Shimadzu 5960A.

Membrane preparation

Three grams of chitosan dissolved in 100 mL of 1% (v/v) acetic acid solution, then formamide is added with a concentration of 10% (v / v) as an additive. Then the mixture is stirred using a magnetic stirrer for ± 24 hours at room temperature; a chitosan polymer solution is obtained. Next, the polymer solution is casted as a thin film on a ceramic plate with thickness 0.45 mm. The solvent is allowed to evaporate using an oven at a temperature of ± 30 °C until the membrane dries and detaches from the ceramic. After that the membrane was washed with 1% NaOH (b/v), rinsed with distilled water until neutral and dried at room temperature. The chitosan membrane obtained was then characterized including measurement of thickness, swelling degree, tensile strength, porosity and membrane morphology using SEM.

Drinking water bags preparation

Drinking water bags are made using Polypropylene (PP) and aluminum foil plastics. The two plastics are cut in a rectangular shape with the same size, then between the two are placed the chitosan membrane that has been made beforehand to form 3 layers. Next, the three plastics are joined together through lamination on all the edges of the plastic using a sealing machine to form a bag of drinking water and installed a lid on the front and back side.

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Draw solution preparation

The draw solution used in the study was a sugar solution in the form of glucose, fructose, sucrose and its mixture with various concentrations of 1,2 and 3M dissolved using distilled water in a 100 mL volumetric flask to the boundary markers. Then it was put into a bag of drinking water and calculated the water flux obtained after the FO process.

Use of drinking water bags

The use of drinking water bags was carried out by filling 100 mL of with draw solution in the form of glucose, sucrose, fructose, and its mixture into a bag of drinking water through the front lid and then filled 200 mL of feed solution in the form of brackish water taken from the Kr. Lamnyong River, Limpok, Aceh Besar. This draw solution will attract the water molecules (solvents) present in the feed solution through a semipermeable membrane made from chitosan which is used as a barrier so that that water will move from the feed solution to the draw solution. The FO process is carried out for 1 hour (Reller, 2011). The FO process illustration that occurs is shown in Figure 1.

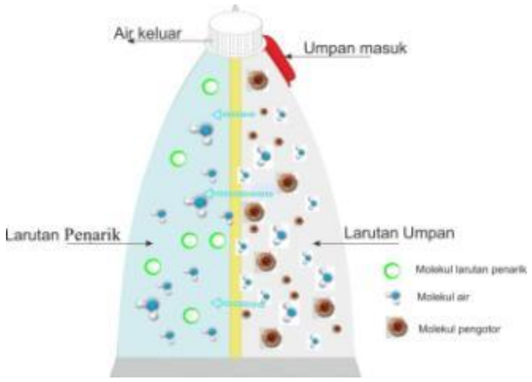


Figure 1. *Forward osmosis process illustration*

Water quality testing of FO products was carried out by measuring several water quality parameters, like pH, TDS, conductivity, salinity, metal content and E.coli bacteria.

Results and discussion

Chitosan membrane

The process of preparation of FO membrane was carried out by dissolving chitosan in 1% (v/v) acetic acid and adding 10% DMF. Then stirred for ± 24 hours at room temperature for homogeneous polymer solution. The use of acetic acid as a chitosan solvent is due to the soluble nature of chitosan in organic acids such as acetic acid, formic acid and citric acid (Mekawati et al. 2000). The addition of DMF can increase porosity because DMF is a porogen additive compound, which can create greater space between polymers chains, causing more membrane pores to form and can facilitate water to move through the membrane (Saiful et al., 2017). The resulting polymer solution is then casted on a ceramic plate that has been set to thickness using tape and dried in an oven at a temperature of ± 30 °C. The selection of ceramics as a membrane printing medium because ceramics do not interact with the polymer solution so that the membrane is easily released after drying. The dried membrane was then washed with 1% (w/v) NaOH solution to neutralize acetic acid through an acid-base neutralizing reaction and rinsed using distilled water to neutral, then dried at room temperature.

Membrane characterization

The chitosan membrane is reported to have transparent, thin and strong properties with a tight and porous structure. Membrane characterization in this study included thickness, porosity, tensile strength, swelling degree, percent elongation, and morphological appearance observed using Scanning Electron Microscope (SEM). The resulting membrane has a thickness of 0.043 mm, porosity 30.3%, swelling degree 43.5 %, tensile strength 28.83 kgf / mm2 and percent elongation of 7.16%. Where as from SEM data shows the membrane has an asymmetry structure, where the structure and pore size of the membrane is not uniform. Characterization results of SEM 10% (v/v) DMF chitosan membrane on the upper, lower surface and cross section with 2500x magnification can be seen in Figure 2.

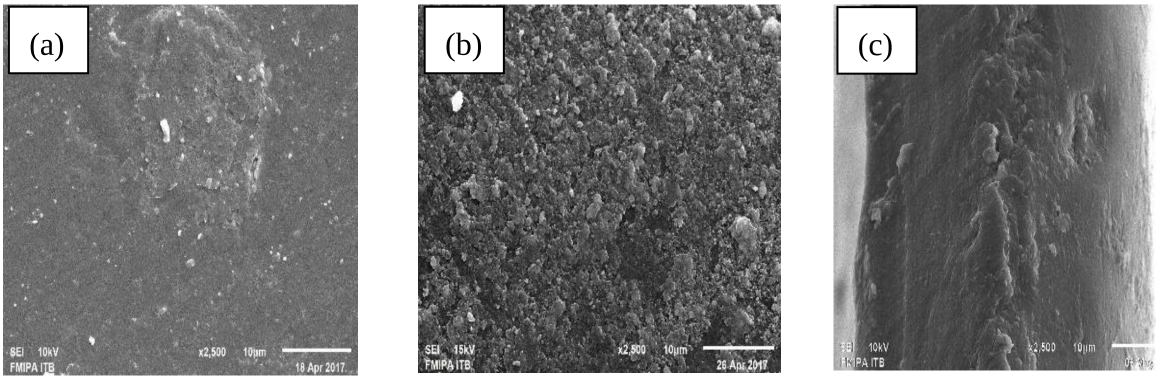


Figure 2. Membrane morphology characterized using SEM with 2500x magnification, (a) the upper surface, (b) the lower surface and (c) the cross section

In addition, the Figure 2 shows that there are differences in structure between the upper and lower surfaces. It is seen that the upper surface is tighter and flatter, while the lower surface is rougher or rarer with 2500x magnification. In the cross section of the membrane with a 2500x magnification, it is seen that the membrane has a well-connected structure and is free of macrovoid. In this study, the desired membrane for FO testing application is a tight and porous membrane (Shakeri *et al*., 2017).

Use of drinking water bags

The drinking water bag produced has a total volume capacity of ± 400 mL, which consists of two sides with a volume of ± 200 mL each, which is limited by a membrane made from chitosan. The front part is used to insert the draw solution in the form of glucose, fructose, sucrose and/or mixture whereas the back part is used for feed solutions, brackish water. The use of drinking water bags for the FO process was carried out by inserting 200 mL of brackish water as a feed solution through the back cover of a bag of drinking water. Then added 100 mL of with draw solution in the form of sucrose, fructose, glucose or mixtures thereof with variations in concentrations of 1, 2 and 3 M respectively from the lid of the front of the bag and left for 1 hour for the FO process.

The FO process was observed for 1 hour with an effective surface area of membrane contact is 41.25 cm2, and the amount of water flux produced was calculated for each draw solution used. When observing the FO process is carried out constant, which is 1 hour because within 1 hour the volume of permeate produced is close to the maximum value and almost meets the bag capacity used. Besides, based on previous research, the first hour of the FO process is the optimum time with the highest flux value, while after that the flux starts to decrease to a constant. The amount of flux produced for each draw solution is shown in Figure 3.

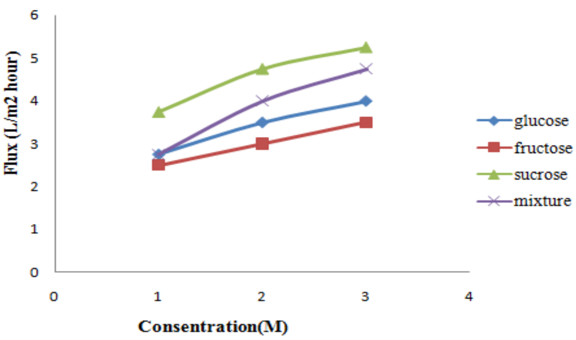


Figure 3. The value of flux for 1 hour FO process

Based on Figure 3 shown that the flux is directly proportional to the concentration, where the higher concentration of the draw solution used was obtained a higher value of the flux. This value is consistent with the theory that the osmosis process is a process that works based on differences concentration in solution, where a solution with a low concentration will lead to a solution with a higher concentration through a semipermeable membrane. The higher gradient or the difference in the concentration of bait and permeate, the higher the resulting osmotic pressure, so that the osmosis process will get longer and produce high flux values (Han, 2015). The maximum flux for each type of draw solution is obtained at 3M concentration, where the resulting flux is higher than using 1 or 2 M draw solutions.

The variation of the draw solution used also affects the water flux produced. In this study, the variation of draw solution used was glucose, fructose, sucrose and a mixture of the three with a concentration variation of 1, 2 and 3 M respectively at the same time and surface area of the touch. Maximum flux is produced from 3M sucrose solution with a flux value of 5.25 L/m²hour. With the same concentration, the flux values for glucose, fructose, and mixtures were 4 L/m²hour, 3.5 L/m²hour and 4.75L/m²hour respectively. Thus the sequence of water flux produced following the sequence of with draw solutions used is sucrose> mixture> glucose> fructose. Sucrose produces a relatively higher flux because it has a higher osmotic pressure and solubility than glucose, fructose or mixture.

Water quality testing for FO products

Analysis of pH, salinity, conductivity, and TDS

Samples or feed solutions used are brackish water taken on the Kr. Lamnyong borders the village of Limpok, Aceh Besar. The characteristics of the sample as brackish water were indicated by salinity values of 9.4 ppt, pH 8.77, the conductivity of 4490 mg/L and TDS 4530 mg/L. These characteristics are following the characteristics of brackish water which is a mixture of fresh water and seawater with salinity between 5-30 ppt (Efendi, 2003). The water from the FO product that was measured was the permeate produced from glucose, fructose, sucrose and/or the third mixture with a concentration variation of 1, 2 and 3M, respectively. The results of the pH analysis of the water obtained are shown in Figure 4.

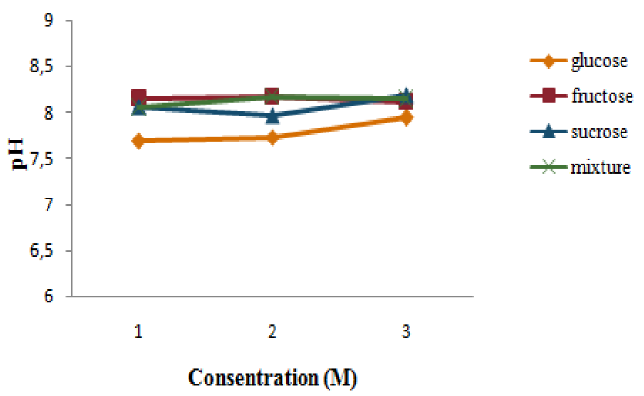
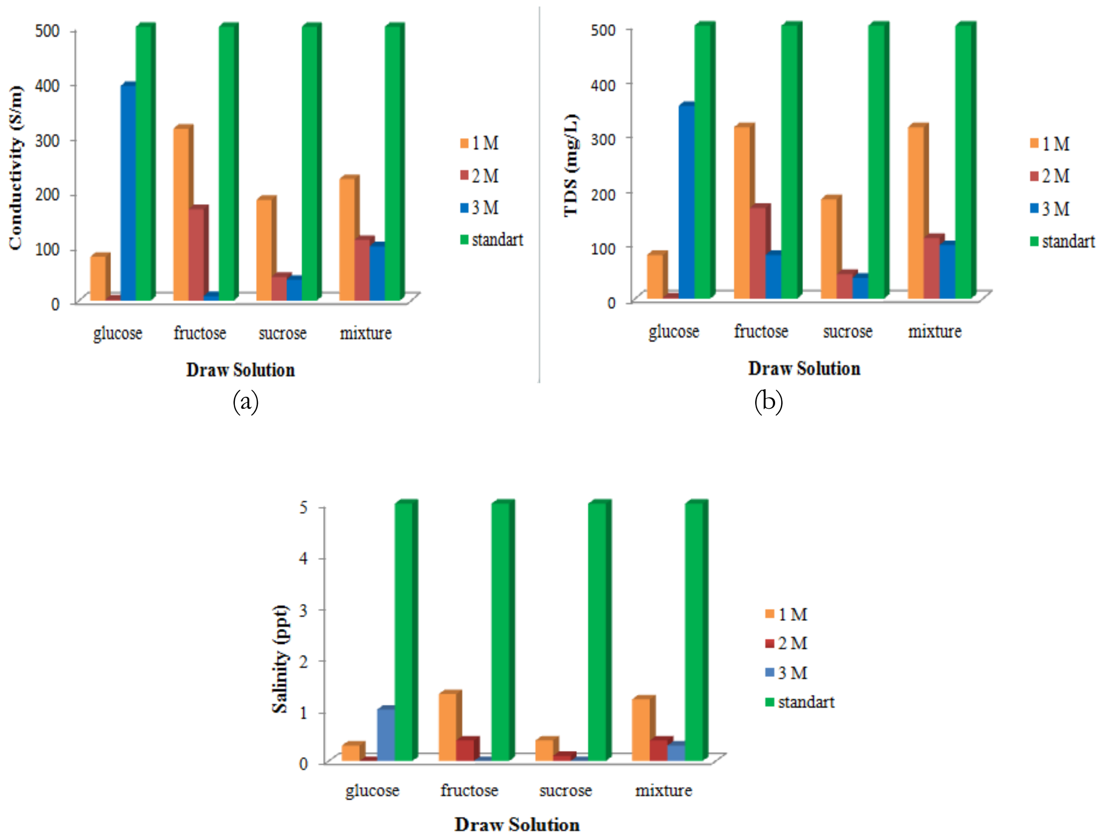


Figure 4. The result of FO product pH analysis

From Figure 4 can be seen the results of the pH analysis of the water produced by FO, where the pH of the draw solution before the FO process for glucose, fructose, sucrose, and the mixture solution was 7.54, 8.01, 7.96 and 8.02, respectively. After FO process for 1 hour the pH of glucose solution fluctuated to 7.69, 7.73 and 7.94 for each concentration of 1, 2 and 3 M. Likewise with fructose solution after FO process, the pH of each concentration became 8,15; 8,17 and 8,11. For sucrose solution, pH became 8.06, 7.97 and 8.19. While the pH of a mixture of glucose, fructose and sucrose for concentrations of 1, 2 and 3M after the FO process was 8.05, 8.15 and 8.14. The results of this pH analysis indicate that the FO process does not result in a significant pH change to the draw solution. The pH of the water produced by the FO process obtained ranged from 7.69 to 8.17. This value is still in the range of pH requirements for drinking water according to PERMENKES No: 492/Menkes/Per/ IV/ 2010, the pH allowed for drinking water is 6.5 - 8.5. For the results of the analysis, salinity, conductivity, and TDS from the FO product water are shown in Figure 5.

Figure 5 shows that the results of FO product water quality testing for each measurement parameter: TDS, conductivity and salinity are still below the general tolerance limit or applicable drinking water quality standards. This result shows that FO-based membranes are capable of filtering dissolved substances or species in the feed solution so that they do not move towards the draw solution, while those that are attracted and capable of passing through the semipermeable membrane are only water. Then the water salinity parameters for FO products also showed good results, where the salinity values obtained ranged from 0-1.3 ppt. This salt content indicates that the water produced is still classified as fresh water and below the maximum salinity limit for commonly used water is 5 ppt. This result proves that the FO membrane can block salt from passing through the membrane so that salt from brackish water does not move through the chitosan membrane during the FO process. Relatively high salinity was obtained from 3 M glucose, 1 M fructose and 1 M mixture with a value of 1.0, 1.3 and 1.2 ppt, respectively. A small leak on the membrane causes the small amount of salt that is displaced because it does not have a pore distribution of membranes that is not homogeneous but does not affect the salinity of the resulting water past the quality standard.



(a) (b) (c)

Figure 5. Quality FO water product analysis, (a) Conductivity
(a) Total Dissolve Solid (TDS), (c) Salinity

TDS values for FO water products ranged from 2-353 mg/L from initial TDS from 1.3 to 11.6 mg/L while for conductivity ranged from 1.92 to 392 S/m from initial conductivity 1.6 to 12.1 S/m. This increase is caused by more water moving from the feed solution (brackish water) to the draw solution. The moving water carries a solute with a particle size smaller than the membrane pores so that it can enter and cause the TDS and conductivity of the draw solution to increase. However, the overall TDS and conductivity values of all draw solutions after the FO process analyzed were still allowed because the value was below 500 mg/L. This result is following PERMENKES: 492/Menkes/Per/IV/2010 that is the maximum allowable TDS standard is 500 mg/L.

Testing of metal content

Analysis of the content of heavy metals in feed water and FO product water was measured using Atomic Absorption Spectrophotometer (AAS). Where the types of metals measured are Hg, As, Mn, Zn, Cr, Fe, Cu, and Cd. The measurement of these metals is because all of these metals are chemical parameters for the types of metals required in drinking water according to the applicable quality standards according to PERMENKES: 492/MENKES/PER/IV/2010. The results of the metal analysis are shown in Table 3.1.

The test results of heavy metals from the sample are brackish water indicating that the sample or brackish water used does not contain metals with high concentrations, where the concentration of small metals is below the detection limit of the tool. So that, the water from the FO product did not show any excess metal content. Based on Table 1. It is seen that all types of metals measured are still below the drinking water quality standard. However, based on previous research reported chitosan-based FO membrane can hold or filter metal to not pass through the membrane so that the metal does not move towards the FO product (Saiful *et.al*, 2018).

Table 1. Metal analysis result

No	Parameter	Unit	Analysis result					Standard
			Sample	Sucrose	Fructose	Glucose	mixture	
1	Mercury (Hg)	mg/L	<0,001	<0,001	<0,001	<0,001	<0,001	0,001
2	Arsenic (As)	mg/L	<0,003	<0,003	<0,003	<0,003	<0,003	0,01
3	Mangan (Mn)	mg/L	<0,002	<0,002	<0,002	<0,002	<0,002	0,4
4	Zinc (Zn)	mg/L	<0,01	<0,01	<0,01	<0,01	<0,01	3
5	Copper (Cu)	mg/L	<0,002	<0,0026	<0,0052	<0,0026	<0,00884	2
6	Chromium Cr)	mg/L	<0,002	<0,002	<0,002	<0,002	<0,002	0,05
7	Ferro (Fe)	mg/L	<0,009	<0,009	<0,009	<0,009	<0,009	0,2
8	Cadmium(Cd)	mg/L	<0,002	<0,002	<0,002	<0,002	<0,002	0,003

Bacterial analysis

Analysis of the presence of bacterial content in FO water product was carried out using the Most Probable Number (MPN) method. The samples analyzed consisted of brackish water and FO water product diluted 1, 2 and 3x dilution respectively. The analysis was carried out by estimation tests and assertion tests. Estimation test is done by incubating water samples that have been inserted into a test tube containing Lactose Broth (LB) and Durham tubes for 24 hours. The positive presence of *E.coli* is indicated by the turbid color of the solution and the appearance of fermented gas which pushes the Durham tube upwards. The results of the estimation test showed positive brackish water samples containing *E.coli* while the FO water was negative. For quantitative assertion, the amount of *E.coli* in brackish water was tested by using EC (Escherichia coli) medium. Positive is that *E.coli* is marked with a metallic green color and the gas produced. The *E.coli* amount is then determined using the MPN table attached in the appendix. The results of E. coli analysis are shown in Table 2. Where in the brackish water sample contains 23/100 mL, while the negative FO water contains *E.coli* (0/100mL). This result shows that the FO membrane can filter *E.coli* from the sample because the membrane pore size is smaller than the size of the bacteria so that the FO product is free of *E.coli*.

Table 2. E.coli analysis result

No	Parameter	Unit	Analysis result	Standard
1	Sample (Brackish water)	MPN/100 mL	23	0
2	FO water product	MPN/100 mL	0	0

Conclusion

Chitosan-based FO membranes combined with polypropylene plastic and aluminum foil can be developed into bags of brackish water filter into energy-ready drinking water ready to drink. Drinking water bags that have been made have maximum water flux using 3M sucrose draw solution. The drinking water bag has a water flux of 5.25 L /m^2j in its application to brackish water filtration. Drinking water products from the FO process meet drinking water quality parameters concerning pH, TDS, salinity, conductivity, metals and the contents of E. coli and coliform bacteria following the regulations of the Minister of Health Regulation No. 492 / MENKES / Per / IV / 2010. FO membrane-based drinking water bags can be used as an alternative solution for energy drinking water supply in emergencies.

Acknowledgments

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