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Design and Performance of Ethanol Purification Tests Using Poly Ether Sulfone Composite Membrane in The Membrane Pervaporation Process

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Abstract. The pervaporation process in the context of ethanol purification is a promising and rapidly developing technology. This is supported by various advantages possessed by pervaporation i.e. do not need additional chemicals, the process is simpler, and does not cause environmental pollution. The purpose of this research is to test the performance of pervaporation design and determine the optimal conditions in the ethanol purification process using a poly-ether-sulfone composite membrane with a coating of 3% alginate & 3% chitosan. This study used two treatments. The first treatment is the temperature of pre-heating in the feed section consisting of 63.64, 67.83, and 72.01 °C. The second treatment is a downstream pressure consisting of 41.325, 46.325, and 51.325 abs kPa. The process was carried out for 40 minutes with a batch system and ethanol feed concentration of 68.05% (w/t). The results showed the highest ethanol concentration was obtained at operating conditions pre heating feed 72.01 °C and downstream pressure of 51.325 abs kPa with ethanol concentration of 82.84% (w/t). The best conditions will be controlled using a non-coating composite membrane and the increase in ethanol concentration in the permeate section is 76.34% (w/t).

Keywords: Ethanol, flux, poly ether sulfone membrane, pervaporation.

1. Introduction

Energy is one of the main issues and is a very important topic in the sustainability of life in many countries. However, the world's fossil energy reserves are depleting which results in an energy crisis and has an impact on the disruption of world economic growth, besides that the use of fossil energy also causes greenhouse gas emissions which result in increased acidity of the waters and leads to environmental damage [1]. Therefore, new alternative energy is needed which is able to suffice or at least can save energy use from fossil fuels [2].

Ethanol is an alternative energy source that has the potential to be used as a substitute for fossil fuels. Before it can be used as fuel, the process of separation and purification of ethanol is one of the important steps that must be passed, because fermented ethanol only has a purity of less than 10% [3][4], while it can be used as a mixture of fossil fuels if the ethanol has a certain level of purity, which is equal to 95-96% whereas if ethanol is used as fuel it must have a purity of 99.5% [5]. The technology that has been widely used at this stage is conventional distillation, but the maximum yield of ethanol produced is only



95% due to the formation of an azeotropic mixture between ethanol and water [6][7]. Several methods have been proposed to separate the azeotropic ethanol-water mixture to obtain ethanol with a purity close to 100%. Extraction distillation and absorbent distillation is a well-known method, both techniques have been shown to be able to separate azeotropic mixtures, but the process is less competitive because it is very complex and requires the addition of chemicals [8]. One of the separation processes that are in great demand by industry as an alternative to distillation is the separation using membrane technology, better known as pervaporation. Pervaporation is a separation process using membranes with a pressure differential push force to solve ethanol purification problems. The ability of pervaporation to separate azeotropic mixtures with a simple process and without the need for chemical additives [9][10] makes this technology applicable for ethanol dehydration. For many cases, this technology provides better economic benefits, especially because the costs of installation and operation are low and do not cause environmental pollution [11]. Pervaporation can be applied in the process of solvent dehydration and separation of organic mixtures and this process is a combination of membrane permeation and evaporation processes, so research on pervaporation for ethanol purification into renewable energy is very interesting to observe. The purpose of this research is to test the performance of pervaporation design and determine the optimal conditions in the ethanol purification process using a poly-ether-sulfone (PES) composite membrane with a coating of 3% alginate and 3% chitosan.

2. Materials and Methods

The materials used in this study include: ethanol pro analysis, PES alginate-chitosan composite membrane with a concentration of 3% each, PES granule as composite membrane material, alginate and chitosan powder as the main raw material for coating, N-Methyl-Pyrolidone as main solvent of composite membrane, aquadest as ethanol solvent, and water as cooling material.

The equipment used in this study includes: pervaporation equipment, where the device consists of two erlenmeyer (Pyrex) with a volume of 500 mL as a feed tank and permeate tank, water bath used as a heater, vacuum pump (Rocker 300), condenser, circulatory pumps, connecting hoses, membrane spacer, and membrane modules.

The research method includes two treatments. The first treatment is the downstream pressure consisting of three variations i.e. 41.325, 46.325, and 51.325 abs kPa on the permeate side, while the second treatment is the pre heating temperature in the feed section which also consists of three variations i.e. 63.64, 67.83, and 72.01 °C. The selection of downstream pressure variations and pre heating temperatures is the result of preliminary research and has been adjusted for membrane use in the study. The design of the pervaporation tool set used in the experiment can be seen in Figure 1. The process begins by preparing 4 L of water as a hot intermediate medium in the water bath to heat the sample to the feed tank. Water is inserted and the temperature in the water bath is adjusted according to the desired temperature. Next, the membrane that will be used as an ethanol purification media is weighed first. The initial mass using an analytical balance is then assembled in a membrane module with membrane spacer. The indicator that there is no leakage in the circuit is when the manometer on the vacuum pump has exceeded 65 kPa (pressure gauge). Next, a sample of 200 mL was poured into the feed tank and closed tightly and then inserted into the water bath. Samples that have been entered into the feed tank. After the manometer on the vacuum pump reaches the expected pressure, the stopwatch is turned on for 40 minutes. The process runs in batches (continue) for 40 minutes.

The membrane used in this study was a PES composite membrane coated using 3% alginate and 3% chitosan in flat sheet form. PES used has a 3 mm size specification in the form of granules dissolved into NMP (N-Methyl-Pyrolidone). This membrane is used as a support part, then the support membrane is coated using alginate-chitosan solution with the concentration of each solvent of 3% so that the final composite membrane results Poly ether sulfone alginate-chitosan with a mean thickness specification of 1.62-2.34 mm and average pore size of SEM results are 4.33-6.73 μm . The membrane module specifications used are made of acrylic 170x90x40 mm with a wall thickness of 10 mm so that the membrane module has an active area of 150x70 mm. The membrane module is equipped with one input

to drain the feed to the membrane and two outputs to drain the permeate to the downstream and to retentate.

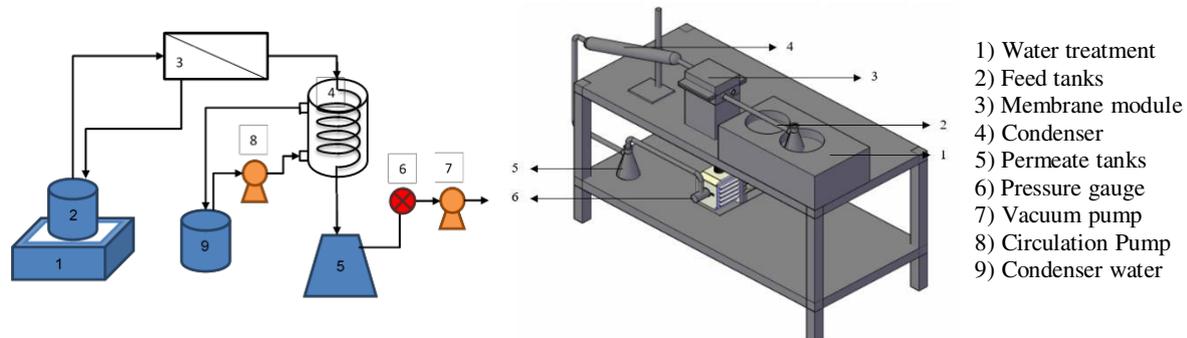


Figure 1. Pervaporation structural design.

3. Results and Discussion

All results in this study are presented in Table 1. Based on these results, the highest purity of 82.84% (w/t) was obtained from variations in heating operating conditions with a pre heating temperature of 72.01 °C with a pressure on the downstream regulated at 51.325 abs kPa. All of the data comes from 3 replications which have been calculated averaged. The results showed that the value of ethanol concentration in the permeate section fluctuated. Whereas when pre-heating at 72.01 °C by reducing the pressure to 46.325 and 41.325 abs kPa, the ethanol purity on the permeate side has decreased, this is because the free volume on the membrane is enlarged along with the reduced pressure on the downstream causing many water molecules which escaped and resulted in a decrease in the purity of the ethanol produced [12]. In accordance with the statement of Franken [13] who said that the temperature in the pervaporation process used to enlarge the driving force must be between 70-100 °C for the feed area and the temperature on the permeate should be in the range of 20-50 °C so that the best performance in the pervaporation process can be achieved.

Tabel 1. Average data results

Feed	Average Data									
	T	P	Permeate		Retentate		Feed		Fluks	Swelling
			% (w/w)	Vol (mL)	% (w/w)	Vol (mL)	% (w/w)	Vol (mL)		
63.64		51.32	80.11	14.66	72.15	25.66	58.78	131.33	0.598	0.229
		46.32	80.87	12.00	70.43	28.66	58.35	123.33	0.597	0.229
		41.32	77.81	15.66	64.02	15.00	56.17	124.66	0.601	0.310
68.05	67.83	51.32	78.40	21.00	68.52	13.33	59.41	126.66	0.601	0.391
		46.32	77.40	25.66	66.91	25.33	55.30	106.00	0.602	0.200
		41.32	79.22	34.00	61.88	24.00	56.48	108.00	0.599	0.163
72.01	72.01	51.32	82.83	40.33	63.04	21.00	51.75	102.66	0.593	0.446
		46.32	79.21	44.66	59.50	18.33	54.18	98.00	0.599	0.198
		41.32	78.82	56.33	58.65	24.66	47.97	86.00	0.600	0.132

In contrast to the volume of permeate produced, where the average volume has a tendency to increase volume along with the increase in pre heating temperature and decrease in downstream pressure. Based on the analysis of variance, the value of the treatment factor at pressure (P) is quite significant with the calculated F value of 25.939 greater compared to the F table which is equal to 3.63 for 5% and at 6.23 for the standard deviation of 1%. From this experiment, it can be concluded that the temperature variations in the feed section and the pressure on the downstream part will be continuous in providing free volume so that it will be easier to pass fluid on the membrane and produce an increasing volume.

The increase in volume in the permeate section also tends to be inversely proportional to the purity of ethanol produced because the more yield volume produced, the purity will decrease due to decreased selectivity on the membrane [13].

From the results of the study it can be seen that there is a downward trend in ethanol levels along with the increase in the temperature of pre-heating in the feed and the low pressure on the downstream side. This can occur due to the enlargement of free volume in the pores of the membrane used so that water molecules will more easily escape and result in a decrease in ethanol levels. At the same time the downstream pressure and heating feed temperature are varied, the higher the pre-heating temperature in the feed will reduce ethanol levels. Then when the pre heating feed temperature as a fixed variable while the downstream pressure is varied, it can be seen also a decrease in ethanol purity. So it can be concluded that downstream pressure variations and the pre-heating temperature of the feed will have an effect on decreasing ethanol levels on retentive.

Decrease in ethanol levels in feeds after heating due to ethanol has a density that is lighter than water so that it has the ability to evaporate faster, while water remains behind, so it will affect the amount or concentration of ethanol contained in the feed and cause ethanol levels in the feed reduced. On average, the increase in temperature and pressure will affect the decrease in ethanol levels in the feed. The greater the temperature and pressure used in the system will affect the volume decrease in the feed after the process takes place. This is inversely proportional to the amount of permeate produced, the greater the temperature and pressure used, the greater the volume of the permeate. This can occur due to the widening of the free volume on the membrane which causes fluid to pass through the membrane easily to the permeate tank which causes the volume in the feed tank to decrease due to fluid transfer to the permeate tank.

The flux value presented in Table 1 can be seen that along with the increase in the temperature of pre-heating in the feed section and the reduction of absolute pressure in the downstream section it will result in increasing flux produced in the permeate section. Based on variance analysis, it can be seen that the value of the treatment factor at pressure (P) is quite significant, with the calculated F value of 26.768 greater than F table which is equal to 3.63 for 5% and for 6.23 for the standard deviation of 1%. From the results of the LSD test, the pressure variations given in the downstream section have a significant effect on the addition of flux to the permeate section. This is in accordance with the free volume theory that the higher the pre-heating temperature and the decrease in downstream pressure will result in opening the pores of the membrane which results in the availability of a larger free volume so as to facilitate a substance to pass through.

From the results of this study at an operating pressure of 51.325 abs kPa, there is an increasing trend, this occurs because the membrane absorbs water with an increasing quantity. In this case it can be associated with the highest permeate side purity i.e. at operating conditions 72.01 °C / 51.325 abs kPa with the highest degree of swelling. Membrane coating in the form of alginate and chitosan has hydrophilic properties (tendency to absorb water) when the highest swelling degree, meaning that a lot of water is trapped in the membrane so that the refining process is more optimal.

The selectivity value will rise in proportion to the increase in operating temperature and then will decrease again. The higher the feed temperature that passes through the membrane, it will cause flux on the permeate side to increase, this will ultimately affect the physical properties of the membrane material to be plastic, and will cause a decrease in the level of selectivity, so that many of the participating water molecules are filtered and cause a decrease concentration on permeate [14]. From the results of the study periodic decline in selectivity values at operating temperatures of 63.64, 67.83, and 72.01 °C. Franken [13] in his research, stated that the increase in flux along with the increase in the temperature of the heating operation in the feed section, and the flux value will be inversely proportional to the selectivity of the membrane used.

Ethanol forms an azeotropic system with water at the ethanol concentration of 95.5% (w/t). At this concentration the ethanol-water mixture cannot be separated using usual distillation process. Therefore, in the pervaporation process after obtaining optimum conditions on variations in pre heating temperature and downstream pressure, a performance test of the PES composite membrane was carried out with

alginate-chitosan coating. In this experiment, pervaporation of an ethanol water mixture with ethanol concentration of 85.5490% (w/t) was compared. This comparison was carried out to examine the performance of PES composite membranes with alginate-chitosan coating in the separation of ethanol-water azeotropic mixtures as shown in Figure 2. From the results, the pervaporation refining process was using ethanol concentrations in the feeds of 85.55% (w/t), operating conditions temperature of 72.01 °C, and downstream pressure at 51.325 abs kPa. The results of ethanol purity at the permeate section were 92.5219% (w/t), there was an increase in ethanol levels in the permeate section of 6.9729% (w/t).

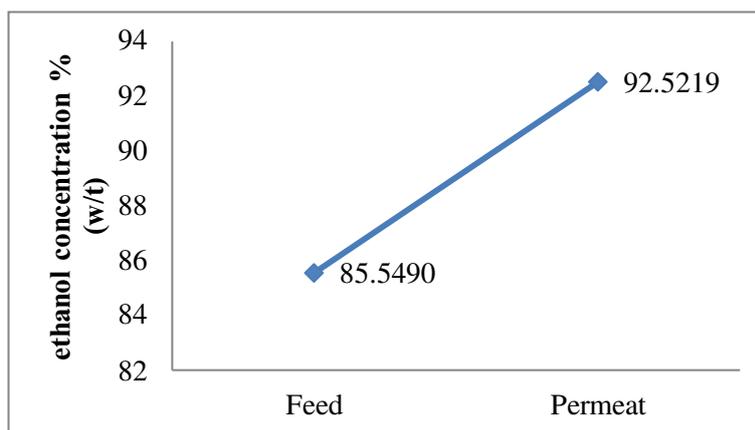


Figure 2. Separation of azeotropic water-ethanol.

4. Conclusion

The best condition is at pre heating temperature of 72.01 °C and downstream pressure of 41.325 abs kPa which produces ethanol purity of 82.84% (w/t) of feed concentration of 68.05% (w/t). The lowest increase in ethanol concentration occurred at conditions of pre heating temperature of 67.83 °C and downstream pressure of 46.325 abs kPa which produced ethanol purity at the permeate portion of 77.41% (w/t). The highest flux value was obtained when the pre heating operating conditions were 72.01 °C and the downstream pressure was 41.325 abs kPa. Where this is in accordance with the hypothesis that the higher the pre heating temperature used and the lower the vacuum pressure, the more flux produced will be inversely proportional to the value of membrane selectivity. Performance test of poly ether sulfone composite membrane with 3% alginate and 3% chitosan coating on the separation of azeotropic ethanol-water mixture was at the best condition with ethanol concentration in the feed at 85.55% (w/t) and the ethanol purity increased in the permeate section at 92.52% (w/t). The higher the temperature and operating pressure on the pervaporation will be directly proportional to the volume on the permeate produced, but this will be inversely proportional to the volume of residual feed and membrane selectivity.

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