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Comparison of Sweepgas and Vacuum Membrane Distillation as In-Situ Separation of Ethanol from Aqueous Solutions

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The rising oil prices, as well as the desire to reduce the environmental impact of fossil fuels, increases the interest in fuels from renewable raw materials. Bioethanol production via starch fermentation is state of the art. Right now researchers focus on second generation biofuels from lignocellulose sources for example like wood and straw. Still investigations have to be done concerning fermentation yield and energy efficiency. Due to low product concentration in the fermentation broth latter downstream processing is quiet energy consuming. Many separation processes like Gas stripping, Liquid-Liquid Extraction, pervaporation and membrane distillation are investigated for an in-situ separation during continuous ethanol fermentation.
The aim of this work was to compare two membrane distillation configurations for ethanol separation from aqueous solutions. Two different process variations like vacuum membrane distillation (VMD) and sweepgas membrane distillation (SGMD) were examined. The influencing factors on the separation of the process such as feed temperature, feed concentration, permeate pressure at VMD and sweepgas flow at SGMD were investigated. The gained results were compared in order to find the best application and process conditions.
The initial feed concentration varied between 0.5 w% and 5 w% ethanol in the mixture and feed temperature was held at 20, 35 and 50 °C. During the VMD configuration the permeate pressure was kept at 25 and 50 mbar. Using the SGMD set up the sweepgas volume flow was varied between 360, 498, 900, 1,302 and 1,500 L/h. Membrane parameters like transmembrane fluxes and selectivity were calculated and discussed.
The results of the VMD configuration showed that transmembrane ethanol flux increased with rising feed temperature and lower vacuum pressure. A linear correlation between feed temperature and transmembrane ethanol flux was found. In comparison VMD showed much higher transmembrane fluxes than SGMD. This result corresponds with the theories, in which the driving force during VMD is much higher due to a larger partial pressure difference between feed and permeate side.
Comparison of selectivity results showed that VMD has better separation potential than SGMD. At a feed temperature of 50 °C selectivity was the same in both configurations. Feed concentration has a negligible influence on the selectivity in the investigated value range.
The result of this work showed that both membrane distillation configurations have the possibility to separate ethanol from aqueous solutions. VMD has a bigger separation potential due the higher driving force, but the applied vacuum makes permeate condensation more difficult. SGMD with selectivities of around 4 and low transmembrane fluxes shows advantages due to easier process configuration and lower energy consumption.

1. Introduction

Membrane Distillation (MD) for desalination is state of the art. Coupling membrane distillation with renewable energy sources like solar energy offer high potential for desalination in southern countries

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(Zaragosa et al., 2014). Further applications like separations of volatile organic compounds are still under research but offer high potential regarding energy savings (Chiam and Sarbatly, 2013). Especially in-situ separation during alcohol fermentation processes is investigated to overcome product inhibition and optimize downstreaming process (Gryta, 2001). Effective process configurations improve fermentation yields and raise alcohol concentration to an effective level for the needed downstream distillation.

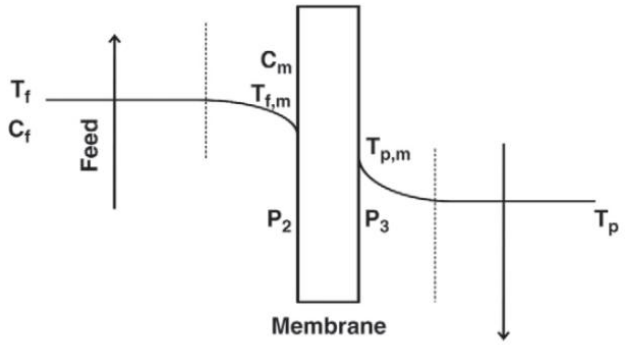

Figure 1: Driving force of the MD process due to concentration difference during SGMD and partial pressure difference during VMD (Alkhudhiri et al., 2012)

In MD the membrane enables an exchange surface for the mass transport from the liquid to the vapour respectively from the feed to the permeate side of the membrane. The membrane itself has no selective layer, however the pore size as well as the membrane thickness influences the process. In Figure 1 the scheme of the driving force is plotted. Temperature and concentration on the feed side as well as vacuum or concentration on the permeate side can influence the mass transport despite the membrane itself.
Masstransport in MD can be defined in different mechanisms, depending on collisions in between molecules or/and molecules and membrane (Alkhudhiri et al., 2012). Without considering detailed masstransport mechanisms, generally the dusty-gas model can be applied. According to Eq.1 the transmembrane flux J_i is proportional to the partial pressure difference Δp_i from feed side to the permeate side of the membrane. P_i is the MD coefficient also known as permeance. It has to be experimentally determined and is a function of process conditions, MD-configuration and Knudsen number Kn (El-Bourawi et al., 2006).
In this work the transmembrane flux is experimentally determined by the mass m_i, which passes the membrane during the experimental time t divided by the membrane area A.

$$J_i = P_i \cdot \Delta p_i = m_i / At \tag{1}$$

Next to transmembrane flux, selectivity is also used to describe membrane processes. It provides information about the separation rate of one component compared to the other component.
According to Eq.2 selectivity is defined as

$$\alpha = w_{i,p} w_{j,f} / w_{j,p} w_{i,f} \tag{2}$$

where $w_{i,j}$ is the weight fraction of component i,j respectively in the feed side f and the permeate side p of the membrane.
In this work experimental parameters were varied to investigate the influence of feed concentration, feed temperature as well as permeate conditions on transmembrane flux and selectivity.

2. Material and Methods

Model solutions were prepared with 96 % Merck ethanol and distilled water.
The setup in the laboratory contained a feed and a permeate cycle. As it can be seen in Figure 2 the feed cycle contains a solution tank on a balance to measure mass reduction. Manometer, flow meter and temperature before and after the membrane module, were installed to record all needed feed data. The feed tank was continuously heated up to overcome the heat loss due to evaporation in the module and to retain constant temperature. A capillary module with a pore size of 0.2 µm and a membrane area of 0.1 m² from the company MicrodynNadir© was used. Switching between SGMD and VMD setup requires a change on the permeate cycle; the feed cycle remained the same.

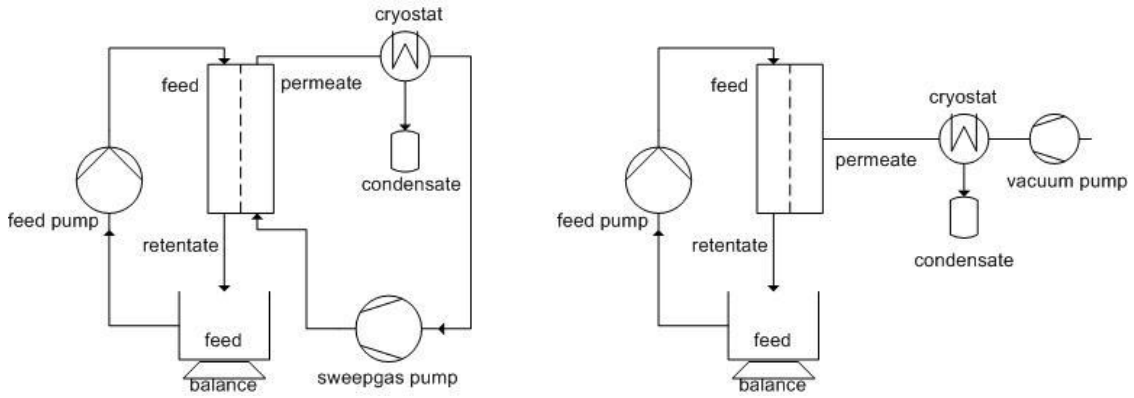

Figure 2: Flow sheet of sweepgas membrane distillation on the left and vacuum membrane distillation on the right

During SGMD run a membrane gas pump circulates the sweep gas flow from the module, to a condenser and back to the pump. By opening the valves in the bypass cycle low volume flows can be applied. VMD requires a vacuum pump to obtain the needed vacuum pressure.
Evaporated permeate condenses at 3 °C using both applications.
In Table 1 adjusted process parameters for both configurations are listed. Influence of temperature and feed concentration were investigated with both configurations. Using SGMD a sweep gas flow of 900 L/h was therefore fixed. Additionally during the investigation of the influence of the sweep gas volume flow constant temperature of 35 °C and initial concentration of 0.5 w% were fixed. In VMD applied vacuum pressure was changed from 25 to 50 mbar.

Table 1: Process parameters during SGMD and VMD

SGMD			VMD		
Temperature (°C)	Initial feed concentration (w%)	Volume flow sweepgas (L/h)	Temperature (°C)	Initial feed concentration (w%)	Pressure (mbar)
20	0.5	900	20	0.5	25
35	5		35	5	50
50			50		
35	0.5	360			
		498			
		900			
		1,302			
		1,500			
		1,740			

In VMD experiments were carried out for 1 h with sample taking every 15 min. It has to be noticed, that the experiment at 20 °C, 0.5 w% and 50 mbar configuration had a very low driving force. The transmembrane flux was too low to measure and is therefore not plotted in the diagrams in the results section. During SGMD configuration experimental time was set at 1.3 hours with sample taking every 20 min.

3. Results

Influences of feed temperature and feed concentration on transmembrane butanol flux and selectivity will be discussed in this chapter. Process specific investigations like sweep gas flow during SGMD or vacuum pressure in VMD configuration will be addressed as well.

3.1 SGMD

Increasing the sweepgas flow implicates higher driving force, due to low concentration on the permeate side of the membrane. As a result transmembrane ethanol flux increases with higher sweepgas volume flow as can be seen in Figure 3. Interestingly there is no effect on selectivity, which can be seen as constant over the whole investigated range at around 3.4.

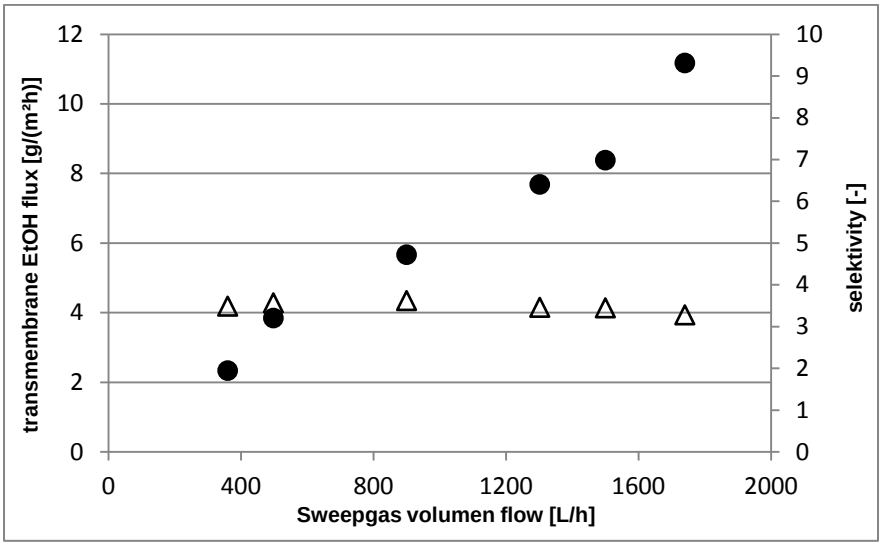

Figure 3: Influence of sweepgas volume flow on transmembrane ethanol flux and selectivity, a)transmembrane ethanol flux (circle), b) selectivity (triangle), process parameters: 35 °C feed temperature and 0.5 w% initial ethanol concentration in feed

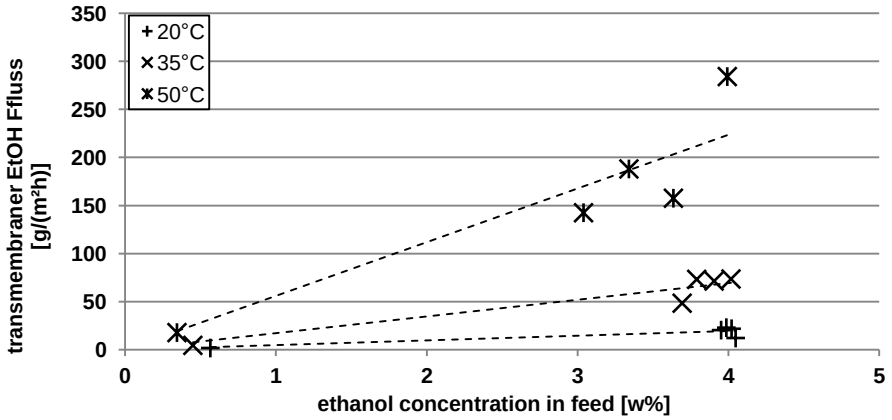

Figure 4: Influence of feed temperature and feed concentration on transmembrane ethanol flux in SGMD

In Figure 4 the influence of feed temperature and feed concentration on transmembrane butanol flux is plotted. Higher temperature results in higher transmembrane ethanol fluxes and increasing ethanol conentration in feed yields also in higher transmembrane ethanol fluxes. This influence raises with higher feed temperature, as can be seen when comparing the different gradients of the trendlines. According to the temperatures 20, 35 and 50 °C gradients change from 4.8 to 17.4 and 56. The highest transmembrane ethanol flux was reached at 50 °C and 4 w% at around 280 g/m²h.

3.2 VMD

As it can be seen in Figure 5 in VMD higher feed concentration results in higher transmembrane butanol fluxes. In the investigated concentration range a linear correlation was found. Similar to the SGMD results the influence of feed concentration raises with higher feed temperature. Applied vacuum pressure of 25 mbar resulted in higher transmembrane fluxes due to higher driving force. At 20 °C and 50 mbar transmembrane flux was negligible and is therefore excluded from the diagram. At 50 °C and 25 mbar the results are lower than at 50 mbar. Due to the very high transmembrane flux at the beginning the vacuum pump could not retain the vacuum, which stayed at around 60. Additionally the high flux deceased feed temperature, which yields in even lower transmembrane fluxes. If the heat and the vacuum could be retained constant over the whole experimental time transmembrane flux is supposed to be the highest in the diagram.

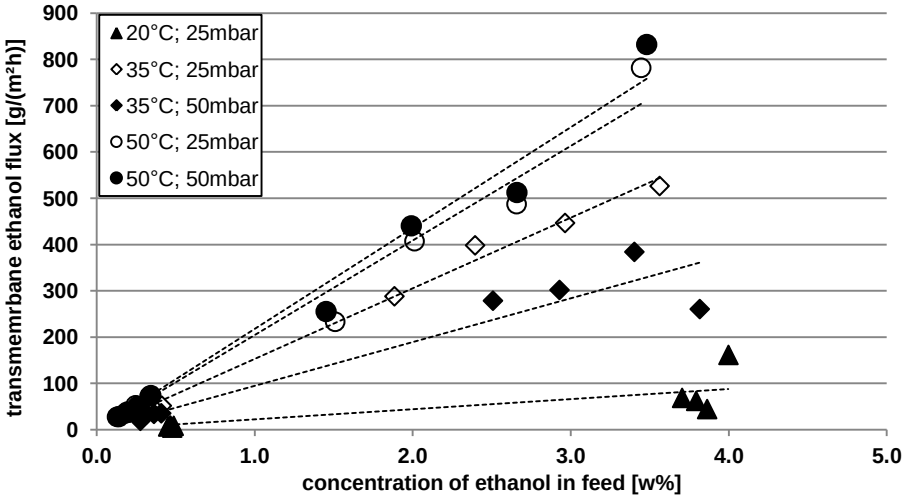

Figure 5: Influence of feed concentration, feed temperature and vacuum pressure on transmembrane ethanol flux in VMD a) 20 °C, 25 mbar; b) 35 °C, 25 mbar; c) 35 °C, 50 mbar; d) 50 °C, 25 mbar; e) 50 °C, 50 mbar

3.3 Comparison of process selectivity in VMD and SGMD

Next to transmembrane flux selectivity is a very decisive factor to describe the effectiveness of a membrane separation process.

In Figure 6 the comparison between VMD and SGMD selectivity is plotted.

In SGMD concentration influence can be neglected over the whole investigated temperature range regarding beam e and f, still selectivity at low concentration is slightly higher. However selectivity raises, when feed temperature increases.

In VMD the influence of feed concentration on selectivity is noticeable and varies the most at 35 °C and 25 mbar between 2.6 and 5. At 20 °C selectivity is around 5.5 when applying 25 mbar. Due to low driving force, when applying 50 mbar at 20 °C, results from this experiment are not significant. At temperatures of 35 and 50 °C applying 50 mbar results in highest process selectivities of 7 and 5.7.

However when feed temperature is at 50 °C selectivities are similar around 4.5 up to 5.5 in both configurations.

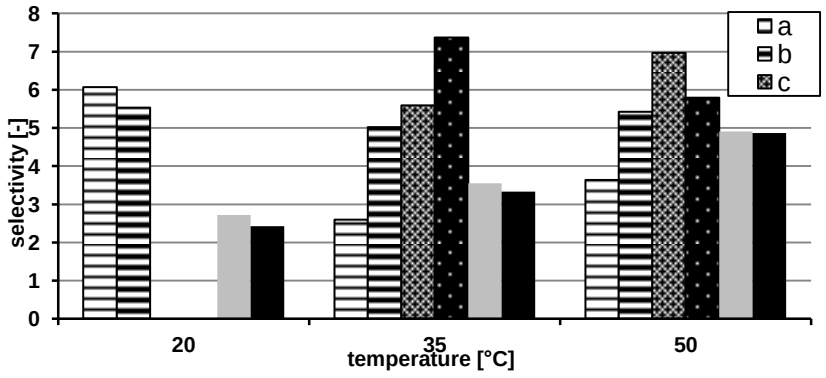

Figure 6: Comparison of VMD and SGMD regarding process selectivity a) 0.5 w%, 25 mbar; b) 5 w%, 25 mbar; c) 0.5 w%, 50 mbar; d) 5 w%, 50 mbar; e) SGMD 0.5 w%; f) SGMD 5 w%

4. Conclusion

In this work two very similar process configurations of membrane distillation, sweepgas and vacuum MD respectively, were investigated as separation technology for aqueous ethanol solutions. Therefore same process parameters were examined using both configurations.

The results of the VMD configuration showed that transmembrane ethanol flux increased with rising feed temperature and lower vacuum pressure. A linear correlation between feed concentration and transmembrane ethanol flux was found.
In comparison VMD showed much higher transmembrane fluxes than SGMD. This result corresponds with the theory, where the driving force during VMD is much higher due to a larger partial pressure difference between feed and permeate side. Comparison of selectivity results showed that VMD has better separation potential than SGMD. At a feed temperature of 50 °C selectivity was the same in both configurations.
The result of this work showed that both membrane distillation configurations have the possibility to separate ethanol from aqueous solutions. VMD has a bigger separation potential due to the higher driving force, but the applied vacuum makes permeate condensation more difficult. SGMD with selectivities between 2.5 and 5 and low transmembrane fluxes shows advantages due to easier process configuration and lower energy consumption.

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