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USING OF DIRECT CONTACT MEMBRANE DISTILLATION FOR WASTEWATER TREATMENT OBTAINED AFTER WHEY PROCESSING

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ABSTRACT
This paper presents the results on the study of separation of model solutions of nanofiltration whey permeate by direct contact membrane distillation. The solute concentration varied from 0 to 450 g/L. The dependence of the change in water activity was determined for this range of concentrations. It would allow simulating the process of membrane distillation in future. There was a decrease in the selectivity of the membranes at a concentration of 300 g/dm^3 and above. It was most likely caused by the formation of a deposit layer on the membrane surface. A process flow diagram of the treatment of such wastewater was proposed. It consists of two stages: direct contact membrane distillation and electrodialysis. Using of the proposed technological scheme will allow us to reuse up to 92% of treated wastewater.

1. Introduction

The process of direct contact membrane distillation is based on the evaporation of the solvent through the hydrophobic porous membrane (Khayet and Matsuura, 2011), herewith "hot" and "cold" solutions are contacted from different sides of the membrane. Using of membrane distillation in water purification technologies makes it possible to obtain high-quality water, even if the initial solution contains components that are difficult to remove, such as arsenic, (Macedonio and Drioli, 2008), boron (Macedonio and Drioli, 2008, Hou et al., 2013), fluorine (Hou et al., 2010, Boubakri et al., 2014a), nitrate (Boubakri et al., 2014b), etc. An advantage of membrane distillation is also the opportunity to concentrate solutes to the limit of their solubility (Mariah et al., 2006, Hickenbottom and Cath, 2014). This is the reason why a number of technologies for processing of wastewater (Lu et al., 2014), groundwater (Hou et al., 2010) , seawater (Al-Obaidani, et al., 2008, Xu et al., 2006, Shirazi et al., 2014) etc. use membrane distillation.

The disadvantage of membrane distillation is the need to heat the feed solution to a temperature of 50-70° C. Therefore, to improve the economic indicators of the process, it is best to use in the presence of cheap sources of heat. One of these places can be a dairy plant with vacuum evaporation facilities where cooling water in condensers can be heated from 10-20° C to 45-55° C. This water can be reused after cooling in the cooling towers. If the wastewater is directed into the condensing apparatus instead of the pure water and is further concentrated (treated) by membrane distillation, it will be able to develop the high performance technology for utilization of excess heat and some quantity of wastewaters.

A promising target of such treatment can be nanofiltration whey permeate (Myronchuk et

al., 2013). Having the low temperature (8-20 ° C), it is not virtually treated and is discharged into drains as the wastewater (Kyrychuk et al., 2014).

Thus, about 65% of the water on the amount of the processed whey is lost. After the preliminary treatment this water can be reused in the dairy plant. Nanofiltration whey permeate contains about 4 g/L of solutes (about 2 g/L of lactose and about 2 g/L of salts) which after the appropriate separation and concentration, can be used for different technological processes (Zmievskii et al., 2014).

The aim of the present work was to study the process of direct contact membrane distillation during separation of model solutions of nanofiltration whey permeate. It will make possible to provide the dairy enterprises with an additional amount of technical water.

2. Materials and methods

The laboratory setup was composed of the membrane cell, two pumps and two heat-exchangers. The hydrophobic membrane MFFK-3 (ZAO STC "Vladipor", Russian Federation) with effective area of $4.8 \cdot 10^{-3}$ m^2 was settled in the cell horizontally making two chambers – the lower and the upper. The "hot" and "cold" solutions were directed into the lower and upper chamber respectively by the pump with circulation flow of 0.1 m/s. The height of the chambers was 2 mm. The turbulence promoters were installed inside the chambers. The temperature of the solutions was controlled by the mercurial thermometers with the accuracy of ±0.1 °C. The salts content was measured by a conductivity meter (HANNA Instruments DIST 1) with expansion bend. The water activity was determined using a portable device Aqualab (series 3, model TE, USA) with the accuracy of ±0.003. The water activity can also be calculated from the equation (1)

$$a_w = \frac{p}{p_0}, \qquad (1)$$

where p and p_0 are partial pressure of water vapour under the real solution and distillery water (Pa), respectively.

During filtration, permeate flow rate was measured by weighting the mass of coming out permeate. Thus the permeate flux J ($kg \cdot m^{-2} \cdot h^{-1}$) was calculated from the equation:

$$J = 3600 \frac{m}{S \cdot \tau}, \qquad (2)$$

where m is the mass of the permeate (kg), S is the membrane area (m^2), and τ is time (s).
The rejection R (%) was calculated as:

$$R = \left(1 - \frac{C_p}{C_r}\right) \cdot 100, \qquad (3)$$

where C_p and C_r are permeate and retentate concentrations (g/L), respectively.

Considering that permeate penetrating through the membrane comes to the upper chamber and is mixed with the cold solution (distillery water), the real value of the permeate concentration C_p was calculated as follows

$$C_p = \frac{\left(V_c + V_p\right) \cdot C_{ск} - V_c \cdot C_{сн}}{V_p} \qquad (4)$$

where V_p is the permeate volume (L), V_c is the volume of the cold solution (L), $C_{сн}$ and $C_{ск}$ are the solute concentrations in the cold solution before and after permeate samples were taken (g/L), respectively.

"Edible" lactose was used for preparation of model solutions of nanofiltration whey permeate, while all inorganic substances were of "chempure" qualification. The preparation of model solution involved the use of the following components: KCl (37.5%), NaCl (11.5%), $CaCl_2$ (1%), and lactose (50%).

3. Results and discussions

Pure water flux of MFFK-3 membrane was determined at the first stage of the study (Figure 1). The maximum temperature was 55 °C in the hot chamber. It is possible due to heating water to this temperature in the tube and shell condensers of vacuum evaporators.

From Figure 1, it can be seen that the pure water flux is higher at higher average temperatures when the difference between temperatures of "hot" and "cold" chambers is even. It can be explained by the nonlinear dependence of partial pressure of water vapor on temperature.

During filtration of model solutions of nanofiltration whey permeate, it was observed the drop of permeate flux in proportion to the increase of the solution concentration (Figure 2). To concentrate the solution up to the 450 g/L it is required to apply higher driving force. Thus, at the temperature difference of 15 °C (T_h=45 °C, T_c=30 °C) the solution was concentrated only up to 300 g/L, while at the temperature difference of 25 °C (T_h=55 °C, T_c=30 °C) it was concentrated up to 450 g/L. First of all the decrease of the permeate flux with the increase of solution concentration is probably associated with the reduction of the water activity. To confirm this supposition the water activity a_w was determined as the function of the solution concentration C (g/L) (Figure 3). The graph is nonlinear and can be described by a polynomial of the second degree to a high precision.

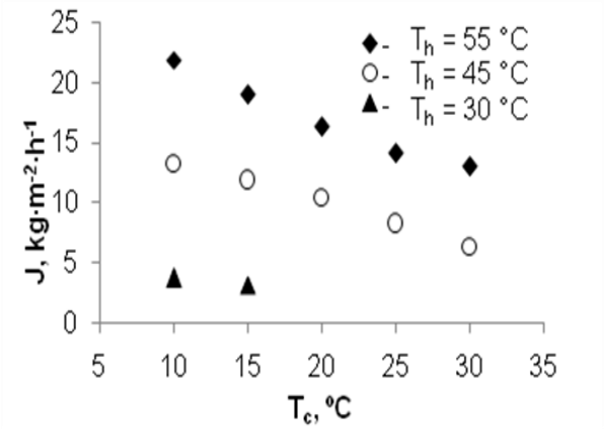

Figure 1. Pure water flux of MFFK-3 membrane vs. temperature in the "cold" chamber. T_h is temperature of the "hot" solution.

From figure 2, 3, it can be seen that at concentration of 300 g/L water activity decreased by just 11.1% while the permeate flux decreased from 35 to 77% as of initial value obtained during pyre water filtration. It indicates the need to consider the other factors such as membrane fouling, concentration polarization and heat polarization. It is obvious that the higher permeate flux is, the higher concentration and heat polarization are (Khayet and Matsuura, 2011). A brown deposit was observed on the membrane surface at the end of the run. For that reason the deep concentration is not always justified. For example, in paper (Hickenbottom and Cath, 2014) the deposition on membrane was observed at salt concentration of 250 g/L that resulted further in quality loss of permeate and decline of permeate flux. Authors (Hickenbottom and Cath, 2014) proposed several versions of reverse membrane distillation to reduce the membrane fouling. For the first one it was suggested to direct permeate into the concentration chamber and retentate into the chambers with permeate at regular intervals. The second one was the change in the solution temperature such as in the next chamber at regular times, i.e. the process was started inversely. Despite the positive effect of these measures it complicates the construction of the setup and increases time of its inefficient use. Thuswise, it is recommended to prevent scale or deposit formation on membrane surface. For this purpose the antiscalants may be used, as the special investigations showed (Sun *et al*., 2011).

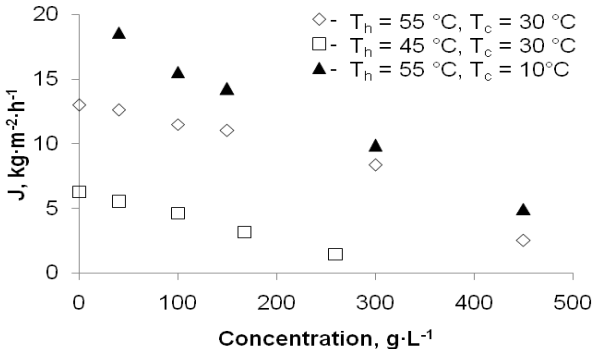

Figure 2. Permeate flux of MFFK-3 membrane vs. retentate concentration during filtration of nanofiltration whey permeate. T_h, T_c are

temperatures of the "hot" and "cold" solutions, respectively.

The water quality obtained after the wastewater treatment is of importance. Figure 5 shows the dependence of rejection and salt content in permeate on concentration of feed solution.

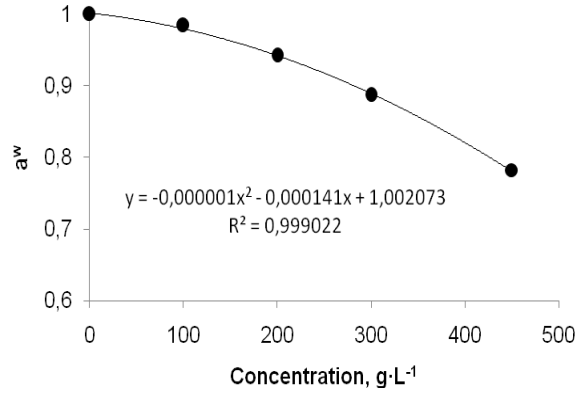

Figure 3. Water activity vs. solute concentration of the model solution of nanofiltration whey permeate.

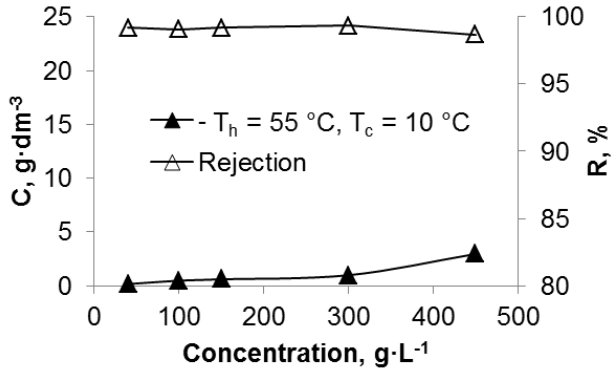

Figure 4.a.

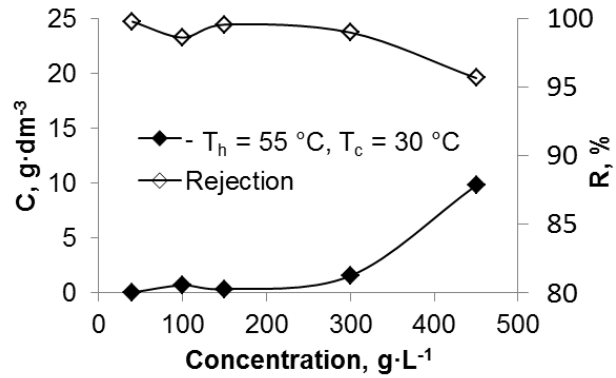

Figure 4.b.

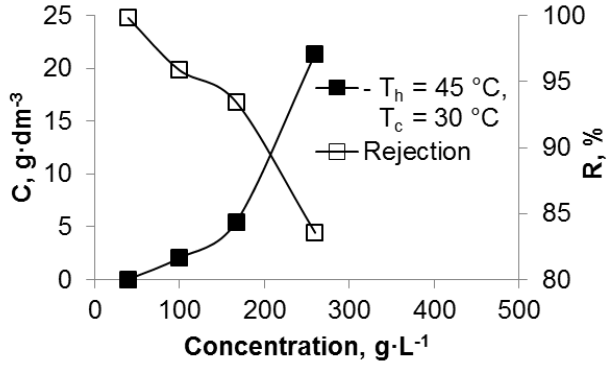

Figure 4. c.

Figure 4: a, b, c. Salt content in permeate and rejection of MFFK-3 membrane vs. solute concentration in the feed solution. T_h, T_c are temperatures of the "hot" and "cold" solutions, respectively.

The quality of the obtained water is essentially worsen during filtration of the solution with concentration higher than 300 g/L (Figure 4 a, b). It is probably associated with the scale formation on the membrane surface that resulted in the decrease of the selective masstransfer (Hickenbottom and Cath, 2014).

It is also can be seen from the figure 4 a-c, that the lower permeate flux is the more solutes permeate contains. It was supposed that MFFK-3 membranes have imperfections as hydrophilic pores, which are wetted by liquid. The diffusive or perhaps even the convective penetration of solutes is carried out through these pores. If we assume that these flows are constant and are functions only of the concentration difference, the quality of permeate will depend on the intensity of high selective mass transfer through the hydrophilic pores.

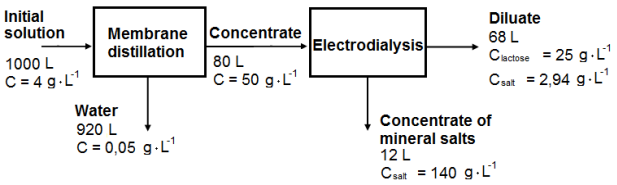

Figure 5. Process flow diagram of treatment of nanofiltration whey permeate, C – total solute concentration $C_{lactose}$, C_{salt} – lactose and salt concentration, respectively.

Summarizing the obtained results one must say that MFFK-3 membranes cannot be used for deep concentration of nanofiltration whey permeate. But they as well as process of membrane distillation can be used for pre-concentration of these wastewaters, e. g. up to 50 g/L. This solution can be directed further to electrodialysis (Zmievskii *et al*., 2014) to separate salts from lactose (Figure 5).

It will reduce the energy consumption at the stage of electrodialysis (Zmievskii *et al*., 2014) and will allow obtaining about 92 % of purified water.

Moreover 6.8% of lactose solution ($C_{lactose} \approx 25$ g/L) and 1.2% of salt concentrate containing mainly monovalent ions ($C_{salt} \approx 140$ g/L) on the amount of the treated wastewater are obtained after electodialysis. The possible reuse application of the received solutions after such two-stage treatment is lightened in paper (Zmievskii *et al*., 2014).

4.Conclusions

During filtration of model solutions of nanofiltration whey permeate, it was found that membrane distillation can concentrate the solutes from 4 to 450 g/L. However, the essential decrease of membrane rejection was observed at the concentration 300 g/L and higher. It is probably caused by deposit formation on the membrane surface.

The dependence of water activity on concentration in the range from 0 to 450 g/L was obtained for model solutions of nanofiltration whey permeate. It would allow simulating the process of membrane distillation in future.

The process flow diagram of two-stage treatment of nanofiltration whey permeate was proposed. It involves the use of direct contact membrane distillation for solutes concentration up to 50 g/L and electrodialysis for separation of salts from lactose. It allows obtaining approximately 92% of purified water on amount of treated wastewater.

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