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Vibratory Membrane Separation for Wastewater Treatment

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Abstract. To meet the requirements defined by environmental protection regulations effective wastewater treatment is required to process effluents before discharging them into sewers or living waters. While membrane separation offers a quite advantageous method to reduce the organic load of wastewaters, membrane fouling is still limiting its application in wastewater treatment.

In this study, the possibility of membrane fouling reduction by increased shear rates on the surface of the membrane was investigated. 7 and 10 kDa *MWCO* ultrafiltration and 240 Da nanofiltration membranes were studied, with the use of a laboratory mode Vibratory Shear Enhanced Processing. This work mostly focused on studying the effects of module vibration and recirculation feed flow rate on permeate flux, specific energy demand and membrane rejections. Using the same operation parameters, vibration and non-vibration mode experiments were carried out with high and low recirculation flow rate to have a deeper understanding of the shear rate effects. It can be concluded that higher shear rate had a positive effect on the process: increased shear rate resulted in higher flux, higher overall rejection values, as well as a significantly decreased specific energy demand. By calculating and comparing the shear rates in experiments with different operating parameters, both vibration and non-vibration mode, both low and high recirculation flow rate, we have reached the conclusion that vibration causes a significantly higher shear rate increase than setting the recirculation flow rate high.

Keywords: vibratory membrane separation, dairy wastewater, ultrafiltration, nanofiltration, fouling, shear rate, vibration

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1. Introduction

The European Union is constantly making serious efforts to address environmental issues, mainly by restricting the protection regulations. Food industry – including dairy industry – uses a huge amount of water for its processes, resulting in vast amounts of effluents. Generally, these effluents can be characterized by high organic load, going along with high chemical oxygen demand (*COD*) and biological oxygen demand (*BOD*). Regulations concerning wastewater disposal require the effective decrease of these pollutants until they meet certain criteria (Rezvantalab et al., 2015). In addition to the conventional wastewater treatment technologies, membrane separation is a good means to reduce both organic and inorganic load of dairy effluents (Luján-Facundo et al., 2017; Frappart et al., 2008). An important advantage of membrane separation is the low amount of chemicals required by the process, while the technology can run on mild operation parameters, and it is easily combinable with other technologies (Molina et al., 2008; Limsawat et al., 2010). Unfortunately, membrane separation has some drawbacks. Both the efficacy and the feasibility of the technology is limited by membrane fouling caused by pore blocking or concentration polarization, leading to flux decline (Bian et al., 2000; Takács et al., 2006). Numerous researches have been addressing this issue, by increasing the shear rates present on the surface of the membrane. In some studies, researchers have managed to increase shear rates by using a static promoter (Koris et al., 2011; Schroen et al., 2017). Others were experimenting with different mechanical methods, to increase shear rates for example by rotating or vibrating the membrane module (Zhenzhou et al., 2016; Goh et al., 2018). Jianquan et al. claimed that by increasing shear rates, one can reduce pore blocking, thus increase flux (Jianquan et al., 2012). In this study, the feasibility of Vibratory Shear Enhanced Processing *(VSEP)* was investigated in dairy wastewater treatment, by processing model dairy effluent with a laboratory mode *VSEP*, equipped with ultrafiltration (*UF*) and nanofiltration (*NF*) membranes. Shear rates caused by recirculation flow rate (*RFR*) and vibration were calculated and compared. The impact of shear rates (in both low and high *RFR*, and in both vibrated and non-vibrated modes) on flux, specific energy demand and rejection values were analyzed and compared.

2. Materials and methods

2.1. Model dairy wastewater

Model dairy wastewater was used as feed in the experiments, which was prepared from distilled water and contained skim milk powder (MilkQuick, Hungary) in a concentration of 5 g/dm^3 and CL 80 anionic detergent (HungaroChemicals, Nagycserkesz) in a concentration of 0.5 g/dm^3. Characteristics of this dairy wastewater were measured at 50°C and are shown in *Table 1.*

Table 1. Model dairy wastewater characteristics at 50°C

Cemical oxygen demand	Electric conductiv ity	ph	Turbidity	Denisity	Protein	Dry matter	Lactose	Viscosity
[mgL^{-1}]	[μScm^{-1}]	[-]	[NTU]	[kgm^{-3}]	[g/g]	[g/g]	[g/g]	[mPas]
5000	1300	7.25	330	953.9	0.32	0.102	0.233	0.37

2.2. Analytical methods

Chemical oxygen demands of the samples were determined with ET 108 digester and a PC CheckIt photometer (Lovibond, Germany). The digestion was done at 150 °C for 2 hours, as the European protocol requires. Turbidity was measured with a HACH 2100AN turbidimeter (Hach, Germany). Density measurements were done with a Mettler Toledo 30PX Densito (Mettler-Toledo, Switzerland) portable density meter. Lactose, protein, and dry matter content of the samples were analyzed with a Bentley 150 infrared milk analyzer (Bentley Instruments, USA). Electric conductivity and pH were determined with a BVBA C5010 multimeter (Consort, Belgium). Viscosity was given by an A&D vibro viscometer SV10 (A&D, Japan). All the analytic measurements were done at least three times, and the results were averaged.

2.3. Membranes and VSEP operating parameters

VSEP Series *LP* filtration apparatus (New Logic Research Inc., USA) equipped with an *L* (Laboratory) module was used. Inside the module, a single circular membrane was inserted, with an effective membrane area of 503 cm^2, inner radius (R_1) of 4.7 cm and outer radius (R_2) of 13.5 cm. Two polyethersulfone (*PES*) *UF* membranes were used, one with a molecular weight cut off (*MWCO*) of 10 kDa (PES-10 SYN, New Logic Research Inc., USA) and one with 7 kDa (PES-5/Tyvek, New Logic Research Inc., USA). Furthermore, a thin film composite *NF* membrane with a *MWCO* of 240 Da (*NF-TFC,* New Logic Research Inc., USA) was also tested. Membranes were kept under distilled water for at least 24 hours prior to separation experiments, which were conducted at 50°C. Transmembrane pressure (*TMP*) was set to 0.8 MPa during *UF* and 3 MPa during *NF*. A high recirculation flow rate of 16 dm^3/min and a low, 4 dm^3/min *RFR*s were applied in different experiments. During experiments in vibration mode, the amplitude was set on 2.54 cm (1 inch) by increasing the frequency. Before starting the separation experiments, flux was measured with distilled water, and after the separation experiment was finished, water flux was measured again and compared in order to determine the flux decrease rate. 10 L of model dairy wastewater was used as feed, and was processed until 2 L of retentate was left, resulting in a volume reduction ratio (*VRR*) of 4, though the dead volume of approximately 1.5 L of the apparatus needs to be considered, as well as the evaporation which is not negligible in longer experiments.

2.4. Calculated parameters

The flux decrease rate, *FDR* [%] was calculated by Eq. 1:

$$\mathrm{FDR} = \left(1 - \frac{J_{WA}}{J_{WB}}\right) 100 \tag{1}$$

where J_{WA} [$m^3m^{-2}s^{-1}$] is the water flux measured – after the separation experiment – on the used, fouled membrane and J_{WB} [$m^3m^{-2}s^{-1}$] is the water flux measured – before the separation experiment – on the unused, clean membrane. The specific energy demands in non-vibration mode, SED_{NV} [kWh m^{-3}] and in vibration mode, SED_V [kWh m^{-3}] were defined using the following equations:

$$\mathrm{SED_{NV}} = \frac{\eta_{fp} \times P_{fp}}{J \times A_{membrane}} \tag{2}$$

$$\mathrm{SED}_V = \frac{\eta_{fp} \cdot P_{fp} + \eta_V \cdot\ P_V}{J \times \cdot A_{membrane}} \tag{3}$$

In non-vibration mode the shear rate, γ [s^{-1}] was determined with the following Eq. 4 (Delaunay et al., 2008). The maximal shear rate, $\gamma_{w\max}$ [s^{-1}]

and the mean shear rate γ_w were defined using the Eq. 5 and Eq. 6 (Al-Akoum et al., 2002):

$$\gamma \frac{4}{\frac{1}{2}h} V_{\max} \tag{4}$$

$$\gamma_{\mathrm{w\,max}} = 2^{\frac{1}{2}} A(\pi f)^{\frac{3}{2}} \mathrm{v}^{-\frac{1}{2}} \tag{5}$$

$$\gamma_{\mathrm{w}} = \frac{2^{\frac{3}{2}}(R_2^3 - R_1^3)}{3\pi R_2\,(R_2^2 - R_1^2)}\ \gamma_{\mathrm{w\,max}} \tag{6}$$

where h is the height of the fluid inside the module [m], $v_{\max}$ is the maximal flow velocity inside the module [m s^{-1}], A is the amplitude [m] and f is the frequency [Hz] of the vibration, ν is the dynamic viscosity of the feed [m^2 s^{-1}]. R_2 is the outer radius of the membrane [m], R_1 is the inner radius of the membrane [m].

3. Results and Discussion

3.1. Calculation of shear rates

By using Eq. 4, 5 and 6 shear rates were calculated in both non-vibration (*NV*) and vibration (*V*) mode. Also, Reynolds number (*Re*) was calculated in *NV* mode. *Re* number turned out to be 8494 if the *RFR* was set to low, and 33975 if it was set to high, meaning that the flow characteristic is laminar in both cases, as the lower limit of transitional flow characteristic is *Re* = 30000, as far as liquid films are concerned. The shear rate we have calculated for *NV* mode was 521 with low, and 2085 with high *RFR* setting. In *V* mode, when the amplitude was set to 2.5 cm (1 inch), the mean shear rate turned out to be 121908, while the maximum shear rate was quite high: 129692.

3.2. Effect of vibration and RFR on flux, specific energy demand and rejections

Fluxes of nanofiltration in both *V* and *NV* mode, with the *RFR* set on both high and low are shown in *Fig. 1*. On the one hand, the highest fluxes can be achieved in vibration mode, but *RFR* has almost no impact on flux in *V* mode. On the other hand, the effect of *RFR* is quite remarkable in *NV* mode, four times higher *RFR* results in two times higher flux values. Similar experiments were conducted with ultrafiltration, and the results showed a very similar tendency.

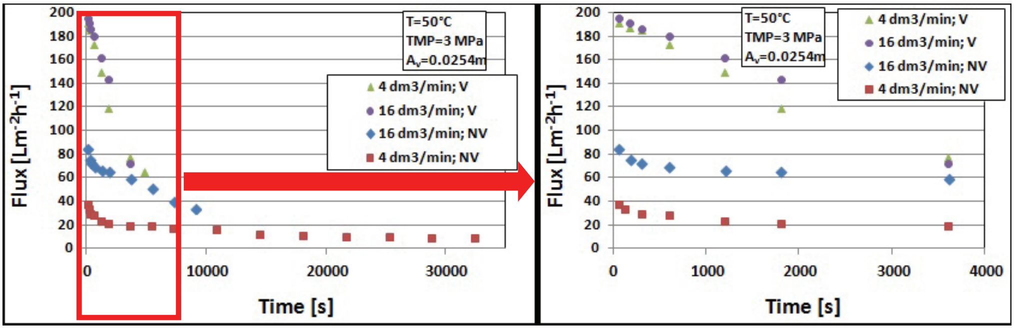

Fig. 1. The impact of vibration and recirculation flow rate on nanofiltration flux values

Specific energy demands of the previously discussed experiments were calculated (Eq. 2 and 3) and are shown in *Fig. 2*. The conclusion we have reached concerning specific energy demand is the following: In *NV* mode with low *RFR*, the process has significantly higher specific energy demands, so we can claim that the effect of *RFR* is more significant in *NV* mode. Clearly, in *V* mode, *RFR* does not seem to have remarkable influence on energy demands at all. It can be concluded that the vibration had a quite positive effect on the specific energy demands.

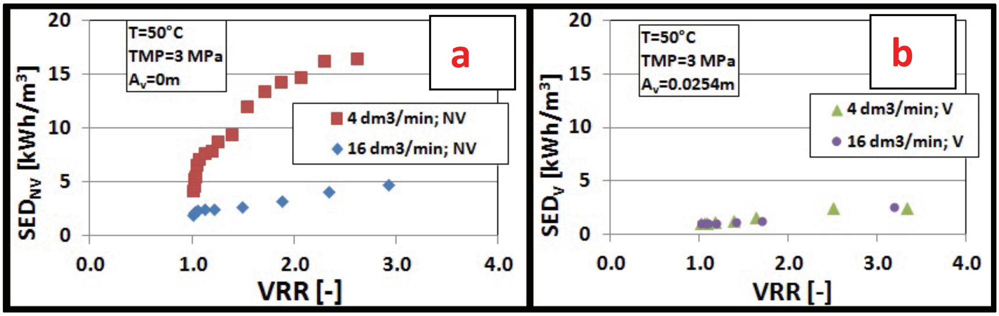

Fig. 2. Specific energy demands (*SED*) in low and high recirculation flow rate experiments: (a) non-vibration mode (SED_{NV}), (b) vibration mode (SED_V)

Chemical oxygen demand (*COD*) rejections of both *UF* and *NF* are shown in *Fig. 3*. By comparing the *COD* rejection values calculated in both *V* and *NV* mode, and also at high and low *RFR*, there is no significant difference. Though this study focuses on the effects of vibration and *RFR*, it is necessary to keep in mind that the purpose of membrane separation in wastewater treatment is to decrease the pollutants, to meet certain criteria. In Hungary, at present the 28/2004 KvVM regulation defines that wastewaters discharged into sewers may have a maximum of 1000 mg L^{-1} *COD*, and wastewaters discharged into living waters may not have a *COD* value higher than 50–100 mg L^{-1} (varies by region) (www.kvvm.com, 2016). In our study, processing the model dairy wastewater (*COD* = 5000 mg L^{-1}) with *UF* resulted in a permeate with a *COD* of ~2000 mg L^{-1}, which is significantly higher than the criteria one has to meet to discharge the effluent into sewer. On the other hand, processing it with *NF* resulted in a permeate with a *COD* lower than 50 mg L^{-1}, which allows the effluent to be discharged into living waters.

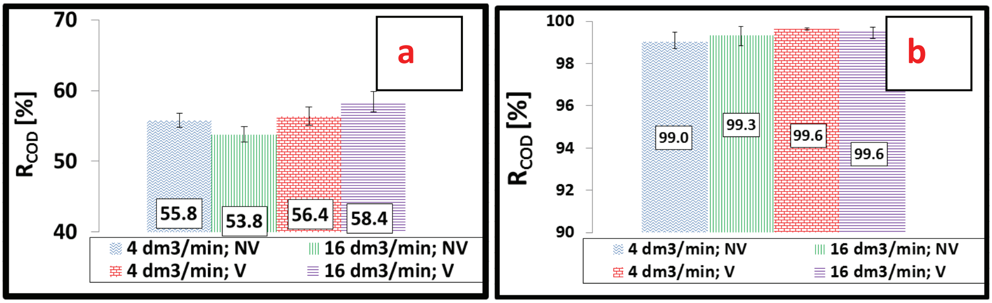

Fig. 3. Chemical oxygen demands rejections of (a) 10 kDa UF membrane and (b) 240 Da NF membrane

Flux decreasing rates (*FDR*) were calculated by Eq. 1. Regarding both *UF* and *NF*, increased *RFR* does not have a significant effect on *FDR*, neither in *V* mode nor in *NV* mode. However, using vibration causes a slight decrease in *FDR* values, especially in case of *NF*.

3.3. Experiments aiming to increase rejection values

In order to find a way to meet the 1000 mg L^{-1} *COD* threshold criteria of the sewer discharge, another *UF* membrane with a lower, 7 kDa *MWCO* was tested. As discussed before, we concluded that the vibration does have a positive overall effect on the process, and the high *RFR* has some (minor) advantages compared to the low one, in *V* mode, thus, we have decided to run the following experiments in *V* mode, with the *RFR* set to 16 dm^3/min.

Separation tests with the 7 kDa *UF* membrane were run by the same parameters as the 10 kDa *UF* tests. *COD*, electric conductivity and turbidity rejections of the two *UF* and one *NF* membranes were measured and are shown in *Fig. 4a*. Comparing the 10 kDa and the 7 kDa *UF* rejections shows a significant increase in *COD* rejections. There are certain components in milk that have molecular weight (*MW*) between 10 kDa and 7 kDa – for example glycomacropeptides, with a *MW* of 8 kDa (Berry et al., 2014), which means that only the 7 kDa *UF* membrane rejects them. It explains the difference between the *COD* rejections of the two *UF* membranes. It was observed that, since our feed model dairy wastewater had a *COD* of 5000 mg L^{-1} and the *COD* rejection turned out to be higher than 80% with the 7 kDa *UF* membrane, this experiment resulted in a permeate with a *COD* lower than 1000 mg L^{-1} – meaning that we have managed to meet the requirements for wastewater discharge into sewers. *Fig. 4a* also shows a significant, 20% increase in electric conductivity rejections, when the 10 kDa and 7 kDa *UF* values are compared. Turbidity rejections were above 99% in every case. Protein and lactose rejections of the two *UF* membranes are shown in *Fig. 4b*. As we could assume based on the *COD* rejection values, the 7 kDa *UF* membrane had higher, almost 100% protein rejection values. It is important to note that the 7 kDa *UF* lactose rejections are twice as high as the 10 kDa *UF* rejections.

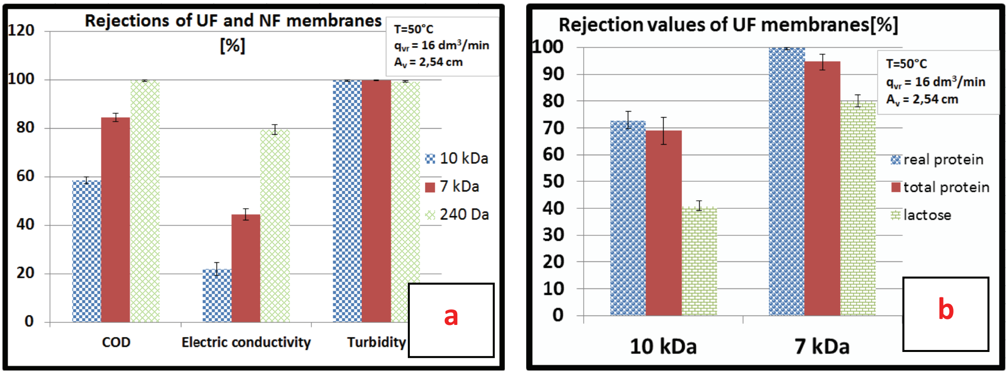

Fig. 4. Rejection values measured calculated for different *UF* and *NF* membranes:
(a) COD, EC and turbidity rejections of UF and NF membranes,
(b) protein and lactose rejections of UF membranes

As previously stated, experiments with both *UF* membranes were carried out with the same parameters. Even if the measured permeate fluxes were similar, the values of specific energy demands showed a noteworthy difference: during the experiment with the 10 kDa *UF* membrane specific energy demand measured at the 30-minute mark was 1.66 kWh/m^3, while this value at the same 30-minute mark turned out to be 2.21–1.66 kWh/m^3 when the 7 kDa *UF* membrane was used, so for the better rejection values we have to invest more energy.

4. Conclusion

In this study, *UF* and *NF* of the tested dairy model wastewater were investigated. We can conclude that higher shear rate has a positive effect on the process in almost every regard. Increased shear rate resulted in higher flux, higher overall rejection values, as well as a significantly decreased specific energy demand. Furthermore, flux decrease rates became lower. By calculating and comparing the shear rates in experiments with different operating parameters (both vibration and non-vibration mode, low and high recirculation flow rate) we have reached the conclusion that vibration causes a significantly higher shear rate increase than setting the *RFR* high. This explains another observation we have made, namely, that in non-vibration mode the *RFR* played a quite important role, the higher *RFR* resulted in a remarkable positive change in every regard. In vibration mode, the *RFR* had negligible impact on most investigated aspects, the results were practically the same whether the *RFR* was set high or low. Also, regarding that the requirements defined by the regulations should be met, we have compared permeate *COD* values with the limits set by the law, and concluded that permeate quality of the NF and 7 kDa *UF* could reach the threshold *COD* limit, while 10 kDa could not.

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