Periodica Polytechnica
Chemical Engineering

60(3), pp. 169-172, 2016
DOI: 10.3311/PPch.8539

RESEARCH ARTICLE

Insertion of Filtration Membranes into Electrodialysis Stack and Its Impact on Process Performance

Ondřej Dlask[1], Natália Václavíková[1*], Marek Doležel[1]

Received 02 September 2015; accepted after revision 30 October 2015

Abstract

The goal of this paper is to investigate the impact of the insertion of different filtration membranes into electrodialysis stack on the process performance and therefore, to gain a tool for choosing membranes suitable for certain applications. Six filtration membranes with different MWCO (molecular weight cut-off), matrix type and specific electric resistance were inserted into diluate chambers of laboratory electrodialysis stack and desalination of sodium chloride solution was conducted at three voltages (1, 2 and 3 V/cell). When determined the electrodialysis characteristics, it was found that the best results (expressed by mass fluxes) was obtained with filtration membranes with PES (polyethersulfone) matrix, whose specific resistance was the most similar to the one of ion exchange membranes. The pore size of filtration membranes did not affect the mass flux significantly except of the stack with nanofiltration membranes.

Keywords

electrodialysis, filtration membranes, mass flux, membrane matrix

1 Introduction

Electrophoresis is a purification process with very high resolution of separated compounds but it is limited in terms of production capacity. On the other hand, electro-membrane processes offer the possibility to achieve large production capacity with good separation efficiency, but they are limited by molecular weight of separated molecules which has to be up to 500 Da. Electrodialysis with filtration membranes (in some papers also known as electrophoretic membrane contactor) is an alternative method of separation and concentration of biomolecule mixtures into fractions and in some cases their simultaneous demineralization. It is a hybrid process that combines conventional electrodialysis with electrophoresis principles using filtration membranes. Ultrafiltration membranes serve as molecular barriers inserted into electrodialysis chambers, whilst the electrical potential difference remains the separation driving force (not pressure difference) [1, 2].

Therefore, separation of biomolecules in electrodialysis with filtration membranes is a consequence of either different electrophoretic mobilities of compounds or different size of their molecules or the combination of both. The process can be controlled in addition to typical electrodialysis parameters also by pH regulation that has an impact on the electrophoretic mobility and by filtration membrane properties (MWCO, type of matrix) [3]. According to requirements for product purity and composition, electrodialysis with filtration membranes can work either in separation or in elution mode [4]. Detailed description of mass transport in this process was studied by Galier and Roux-de Balmann in [5].

Electrodialysis with filtration membranes was successfully applied, e.g. for separation of chitosan oligomers [1, 6], cranberry juice enrichment by polyphenol antioxidants [7, 8], separation of whey proteins [3], separation of bioactive peptides from alfalfa protein hydrolysate [9, 10], soy protein hydrolysate [11] or snow crab protein hydrolysate [12, 13].

In this paper, the impact of filtration membrane insertion into electrodialysis stack on electrodialysis performance was examined in order to consider suitability of different filtration membranes for this application.

[1] MemBrain s.r.o., Pod Vinicí 87, 471 27 Stráž pod Ralskem, Czech Republic

[*] Corresponding author, email: natalia.vaclavikova@membrain.cz

2 Experimental

For electrodialysis experiments, laboratory-scale module EDR-Z (MemBrain s.r.o., Stráž pod Ralskem, Czech Republic) with five membrane pairs of heterogeneous ion-exchange membranes RALEX® (MEGA a.s., Stráž pod Ralskem, Czech Republic) was used. The spacer thickness was 1.6 mm and the effective area of one membrane was 64 cm^2. In order to examine electrodialysis process with filtration membranes, these membranes were inserted into diluate chambers (Fig. 1) with no specific separation function. The main purpose of such configuration was to observe how their insertion affects desalination ability of electrodialysis module.

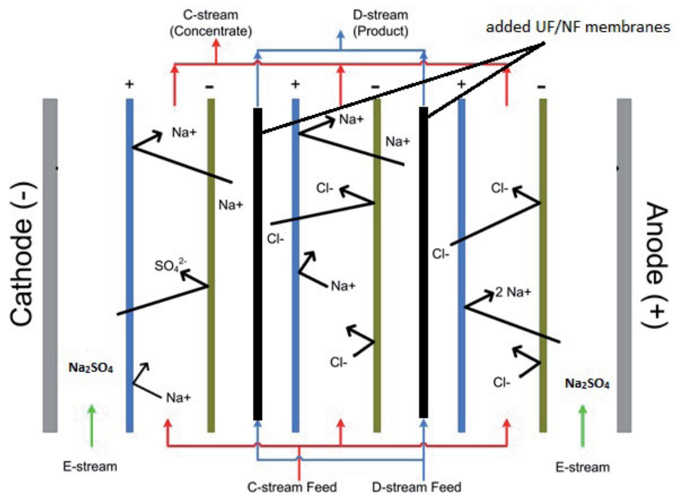

Fig. 1 Scheme of electrodialysis stack with filtration membranes inserted in diluate chambers (D – diluted stream, C – concentrated stream, E – electrode rinsing stream, UF/NF – ultrafiltration or nanofiltration)

Sodium chloride water solution (0.2 M, conductivity approx. 20.5 mS cm^{-1}) was used as a feed solution to be desalinated and sodium sulphate water solution (0.07 M) was used as electrode rinsing solution. Feed solution with initial volume of 1 L was circulated through the stack with linear flow velocity of 4.34 cm s^{-1}. Desalination experiments were performed at three different voltages – 5 V, 10 V and 15 V. Temperature of all solutions was kept in the range of 25 ± 2 °C. The end of each experiment was determined by the final conductivity in diluate that had to be less than 5 mS cm^{-1}. Apart from experiments with inserted filtration membranes, the same procedure was done for conventional electrodialysis stack with no filtration membranes. All experiments were made in triplicates.

Properties of filtration membranes (Synder® Filtration, Vacaville, CA, USA) used in experiments are summarized in Table 1. Specific resistance was measured in 0.5 M sodium chloride solution using direct current. Specific resistance of ion-exchange membranes RALEX® is 110 Ω cm and 115 Ω cm for cation and anion exchange membranes, respectively. Six different filtration membranes were chosen in order to cover relatively wide range of membrane MWCO – from nanofiltration membrane NFX with the smallest pores to ultrafiltration membrane BY with the largest pores.

Table 1 Summary of characteristic filtration membranes properties (PVDF is polyvinylidene fluoride, PA TFC is polyamide thin film composite)

Membrane brand name	Matrix [-]	MWCO [kDa]	Thickness [mm]	Specific resistance [Ω cm]
NFX	PA TFC	0.15 – 0.35	0.178	764.25
XT	PES	1	0.191	152.69
MT	PES	5	0.185	176.14
ST	PES	10	0.190	140.5
BN	PVDF	50	0.179	1711.06
BY	PVDF	100	0.191	512.00

3 Calculations

From recorded data, common characteristics of electrodialysis process were evaluated. Current efficiency η [%] was calculated using Eq. (1) and Eq. (2).

$$Q = \int_{t_0}^{t} I(t)\,dt \tag{1}$$

where Q [C] is the electric charge, I [A] the electric current, t [s] the time.

$$\eta = \frac{\upsilon_C z_C F \Delta n}{NQ} 100\% \tag{2}$$

where υ_C is the stoichiometric coefficient of cations in compound, z_C the valence of cations, F [C mol^{-1}] the Faraday constant, Δn [mol] the amount of substance (sodium chloride) transported from diluate to concentrate, N the number of membrane pairs in electrodialysis stack, Q [C] the electric charge.

Mass flux of sodium chloride J [kg m^{-2} h^{-1}] was calculated using Eq. (3) and specific energy consumption E [W h kg^{-1}] using Eq. (4):

$$J = \frac{\Delta n}{MNwL\Delta t} \tag{3}$$

where Δn [mol] is the amount of substance (sodium chloride) transported from diluate to concentrate, M [kg mol^{-1}] the molecular mass of compound, N the number of membrane pairs in electrodialysis stack, w [m] the effective width of membrane, L [m] the effective length of membrane, Δt [h] the duration of experiment.

$$E = \frac{UQM}{3600\Delta n} \tag{4}$$

where U [V] is the applied voltage, Q [C] the electric charge, M [kg mol^{-1}] the molecular mass of compound, Δn [mol] the amount of substance transported from diluate to concentrate.

4 Results and discussion

In Table 2, all results gained from experimental tests are presented. Since all filtration membranes had higher specific resistance than ion exchange membranes, it was supposed that electrodialysis performance get worse in the case of inserted membranes. Result approved this expectation as the insertion of filtration membrane into the electrodialysis stack always meant the decrease of its desalination capacity (herein expressed by mass flux). Therefore, to keep the same capacity as conventional electrodialysis stack had, it would be necessary either to work with higher energy consumption by the means of increased voltage or to enlarge effective membrane area. It is necessary to note that the increase of voltage in electrodialysis with filtration membranes can lead to changes in product composition so that in some cases it is not desirable [14, 15]. No dependence of desalination performance on MWCO of filtration membranes was observed.

Stacks with inserted filtration membranes with PES matrix achieved mass fluxes the most similar to these in conventional stack. Among them, the XT membrane had the highest mass flux. The lowest mass flux could be observed in the stack with NFX membrane although this membrane had not the highest specific resistance. The reason of this fact is probably in very small pores of this membrane; sodium and chloride ions with their solvation shells could be partially rejected by them. The stack with inserted BN membranes showed the second lowest mass flux. This is in accordance with expectations based on very high specific electric resistance of these membranes (almost 15-times higher than the resistance of ion exchange membranes).

In Figure 2, the dependence of mass flux on applied voltage for each examined stack is depicted. Except of BN and NFX membranes, in every stack the increase of mass flux with applied voltage is almost linear. In the case of NFX membrane, fluxes achieved at 5 V and 10 V did not differ significantly, but the voltage increase to 15 V caused more than doubled mass flux. Moreover, the slope of the dependence observed in the range of 10 – 15 V is very similar to curve slopes describing the other membranes. The reason is again probably the combination of high specific resistance and very small pores of NFX membrane. In the case of BN membranes, the increase of mass flux with the voltage is almost negligible. It can be explained by their specific resistance that is almost 15-times higher than the one of ion exchange membranes. Results of the module with BN membranes show that the behaviour of these membranes is close to membranes with infinite specific resistance (no change of mass flux with the increase of driving force).

Regarding the energy consumption, it was in most cases slightly higher in stacks with inserted filtration membranes that in that one with no filtration membrane; obviously because of lower current efficiency. This could be caused by external leakages that were observed in small amounts when working with stacks with filtration membranes. These membranes had different mechanical properties than ion exchange membranes and commonly used LDPE (low density polyethylene) spacers were not able to seal the stack in the satisfactory way. This fact has to be taken into account in the future research continuation and more suitable spacers have to be developed.

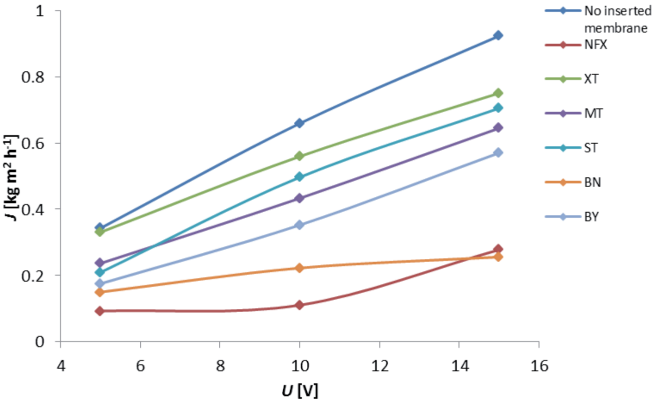

Fig. 2 Dependence of mass flux on applied voltage in stacks with different inserted membranes

Table 2 Desalination process characteristics achieved with different filtration membranes inserted in electrodialysis stack

Membrane brand name	Voltage [V]	Mass Flux [kg m^{-2} h^{-1}]	Current efficiency [%]	Energy consumption [W h kg^{-1}]
No inserted membrane	5	0.349	98.0	470.8
	10	0.659	88.7	1036.1
	15	0.923	86.2	1596.1
NFX	5	0.093	88.4	544.6
	10	0.111	71.5	1283.0
	15	0.278	76.6	1797.0
XT	5	0.331	88.4	536.5
	10	0.560	86.2	1064.8
	15	0.750	80.6	1708.9
MT	5	0.237	90.2	518.4
	10	0.433	89.1	1029.1
	15	0.645	87.8	1567.6
ST	5	0.211	78.7	566.6
	10	0.508	76.9	1194.9
	15	0.705	81.8	1697.2
BN	5	0.150	80.5	569.2
	10	0.223	78.2	1173.0
	15	0.256	74.9	1841.7
BY	5	0.176	84.7	545.2
	10	0.353	82.9	1106.1
	15	0.570	89.1	1545.2

Described results showed that the choice of filtration membrane for the process of electrodialysis with filtration membranes is not only dependent on MWCO. Very significant parameter is also electric resistance and matrix type of membranes. According to results described in this paper, PES filtration membranes are suitable for electromembrane processes as their specific resistance is similar to that one of ion exchange membranes and their use would not increase energetic demands of the process significantly.

5 Conclusions

Six different ultrafiltration and nanofiltration membranes were inserted into the diluate chambers of classical electrodialysis stack in order to investigate their impact on process performance. It was found out that filtration membranes with PES matrix are the most suitable for hybrid electrodialysis with filtration membranes as they did not decrease mass flux dramatically while energy demands remained almost the same as in electrodialysis stack with no inserted filtration membranes. The reason of this fact was the specific resistance of PES membranes, which was similar to that one of heterogeneous ion exchange membranes used in electrodialysis stack. MWCO of filtration membranes did not played the key role in the process, except of the case of NFX membrane with the smallest pores, where the performance was obviously affected by this parameter as well as by the PA TFC matrix of membrane and its high specific resistance. The results described in this paper will be used in future work to choose suitable filtration membrane for hybrid electrodialysis module for processing of real solutions, where the insertion of membranes could e.g. prevent fouling of ion exchange membranes or enable to separate mixtures of biomolecules. By choosing the membrane with appropriate matrix (and electric resistance) whole process can be performed in an energetically acceptable way.

Acknowledgement

The work was carried out within the framework of the project No. LO1418 “Progressive development of Membrane Innovation Centre” supported by the program NPU I Ministry of Education Youth and Sports of the Czech Republic, using the infrastructure Membrane Innovation Centre.

References

[1] Aider, M., Brunet, S., Bazinet, L. "Electroseparation of chitosan oligomers by electrodialysis with ultrafiltration membrane (EDUF) and impact on electrodialytic parameters." *Journal of Membrane Science.* 309(1-2), pp. 222-232. 2008. DOI: 10.1016/j.memsci.2007.10.029

[2] Galier, S., Roux-de Balmann, H. "Influence of electrostatic interactions in electrophoretic membrane contactors." *Desalination.* 149(1-3), pp. 351-356. 2002. DOI: 10.1016/S0011-9164(02)00818-4

[3] Galier, S., Roux-de Balmann, H. "The electrophoretic membrane contactor: A mass transfer-based methodology applied to the separation of whey proteins." *Separation and Purification Technology.* 77(2), pp. 237-244. 2011. DOI: 10.1016/j.seppur.2010.12.013

[4] Galier, S., Roux-de Balmann, H. "Study of biomolecules separation in an electrophoretic membrane contactor." *Journal of Membrane Science.* 241(1), pp. 79-87. 2004. DOI: 10.1016/j.memsci.2004.05.007

[5] Galier, S., Roux-de Balmann, H. "Study of the mass transfer phenomena involved in an electrophoretic membrane contactor." *Journal of Membrane Science.* 194(1), pp. 117-133. 2001. DOI: 10.1016/S0376-7388(01)00527-0

[6] Aider, M., Brunet, S., Bazinet, L. "Effect of solution flow velocity and electric field strength on chitosan oligomer electromigration kinetics and their separation in an electrodialysis with ultrafiltration membrane (EDUF) system." *Separation and Purification Technology.* 69(1), pp. 63-70. 2009. DOI: 10.1016/j.seppur.2009.06.020

[7] Bazinet, L., Brianceau, S., Dubé, P. Desjardins, Y. "Evolution of cranberry juice physico-chemical parameters during phenolic antioxidant enrichment by electrodialysis with filtration membrane." *Separation and Purification Technology.* 87, pp. 31-39. 2012. DOI: 10.1016/j.seppur.2011.11.017

[8] Husson, E., Araya-Farias, M., Desjardins, Y., Basinet, L. "Selective anthocyanins enrichment of cranberry juice by electrodialysis with ultrafiltration membranes stacked." *Innovative Food Science and Emerging Technologies.* 17, pp. 153-162. 2013. DOI: 10.1016/j.ifset.2012.09.011

[9] Firdaous, L., Dhulster, P., Amiot, J., Gaudreau, A., Lecouturier, D., Kapel, R., Lutin, F., Vézina, L.-P., Bazinet, L. "Concentration and selective separation of bioactive peptides from an alfalfa white protein hydrolysate by electrodialysis with ultrafiltration membranes." *Journal of Membrane Science.* 329(1-2), pp. 60-67. 2009. DOI: 10.1016/j.memsci.2008.12.012

[10] Firdaous, L., Dhulster, P., Amiot, J., Doyen, A., Lutin, F., Vézina, L.-P., Bazinet, L. "Investigation of the large-scale bioseparation of an antihypertensive peptide from alfalfa white protein hydrolysate by an electromembrane process." *Journal of Membrane Science.* 355(1-2), pp. 175-181. 2010. DOI: 10.1016/j.memsci.2010.03.018

[11] Roblet, C., Doyen, A., Amiot, J., Pilon, G., Marette, A., Bazinet, L. "Enhancement of glucose uptake in muscular cell by soybean charged peptides isolated by electrodialysis with ultrafiltration membranes (EDUF): Activation of the AMPK pathway." *Food Chemistry.* 147, pp. 124-130. 2014. DOI: 10.1016/j.foodchem.2013.09.108

[12] Doyen, A., Saucier, L., Beaulieu, L., Pouliot, Y., Bazinet, L. "Electroseparation of an antibacterial peptide fraction from snow crab by-products hydrolysate by electrodialysis with ultrafiltration membranes." *Food Chemistry.* 132(3), pp. 1177-1184. 2012. DOI: 10.1016/j.foodchem.2011.11.059

[13] Suwal, S., Roblet, C., Doyen, A., Amiot, J., Beaulieu, L., Legault, J., Bazinet, L. "Electrodialytic separation of peptides from snow crab by-product hydrolysate: Effect of cell configuration on peptide selectivity and local electric field." *Separation and Purification Technology.* 127, pp. 29-38. 2014. DOI: 10.1016/j.seppur.2014.02.018

[14] Doyen, A., Roblet, C., Beaulieu, L., Saucier, L., Pouliot, Y., Bazinet, L. "Impact of water splitting phenomenon during electrodialysis with ultrafiltration membranes on peptide selectivity and migration." *Journal of Membrane Science.* 428, pp. 349-356. 2013. DOI: 10.1016/j.memsci.2012.10.036

[15] Doyen, A., Roblet, C., L’Archeveque-Gaudet, A., Bazinet, L. "Mathematical sigmoid-model approach for the determination of limiting and over-limiting current density values." *Journal of Membrane Science.* 452, pp. 453-459. 2014. DOI: 10.1016/j.memsci.2013.10.069