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Effect of pulsed electric field and polarity reversal on peptide/amino acid migration, selectivity and fouling mitigation

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ABSTRACT

The deterioration of ion-exchange membrane (IEM) properties due to fouling and dissociation of water molecules at membrane-solution interface is considered to be an important limiting factor in electrodialysis (ED) separation processes such as ED with filtration membrane (EDFM). Non-stationary regimes such as pulsed electric field (PEF) is known to enhance ED efficiency by reducing fouling and water splitting, while polarity reversal (PR) of current with change in flow streams has been used as self-cleaning in place (CIP) tool in conventional ED processes. The present study aims to study, for the first time, the effect of PEF and PR (without changing the solution flow streams) on membrane characteristics and fouling behavior, energy consumption, and peptide migration rate as well as selectivity during peptide fractionation by EDFM. At the same time, the study was carried-out at two different constant voltages, 20 V (under-limiting current density) and 40 V (over-limiting current density). The peptide migration rate was unaffected for all types of electric field at 20 V while it was significantly lower with PR as compared to PEF and DC at 40 V. The selectivity of Arg and Lys was maximum in PEF mode at 20 V. A strong protective effect on membrane physicochemical properties was observed with PEF and PR regimes as compared to DC by reducing fouling and water dissociation at the membrane-solution interface, especially on AEM. Moreover, the relative amount of energy consumption was the lowest with PEF in relation to other two modes. Therefore, the present study demonstrated that PEF is energetically and technologically more feasible as compared to the conventional EDFM process with DC current.

1. Introduction

Some peptides isolated from food protein hydrolysates have demonstrated biological activities such as antimicrobial, anticancer, antidiabetic, anti-hypertensive and immunostimulatory. Consequently, these high-valued molecules are receiving considerable attention of scientific and industrial communities [1]. However, the separation and purification of bioactive peptides (BPs) from a complex mixture like protein hydrolysate represent a real challenge. The conventional pressure-driven membrane filtration processes have been widely used for peptide fractionation [2]. However, these techniques are most often criticized for their low selectivity for peptides having similar molecular sizes and their susceptibility to membrane fouling. Consequently, electrically driven membrane processes such as electromembrane filtration (EMF) [3] and more recently, electrodialysis with filtration membrane (EDFM) using ion-exchange membrane (IEM) and ultrafiltration membrane (UFM) were developed to improve the selective separation of electrically charged molecules such as proteins, peptides and amino acids [4]. Peptide fractionation and purification using EDFM process represents a very interesting perspective in functional food, nutraceutical and biopharmaceutical industries. However, to demonstrate the economic viability of EDFM, the problems associated to this specific process, such as membrane fouling and deterioration should be properly addressed. One of the major limiting factor during electromembrane process is the appearance of concentration polarization (CP) layer which subsequently promotes water dissociation at IEM-solution interface [5]. During electromembrane process especially at limiting current density (LCD), IEMs are found to be adversely affected by fouling by peptides and amino acids [6–11]. These phenomena could considerably reduce separation efficiency, productivity and selectivity, increase the energy consumption and cleaning frequency. Recently, effects of LCD and water splitting on peptide migration rate and selectively was demonstrated by Doyen et al.

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[12]. Furthermore, IEM, particularly anion-exchange membrane (AEM) containing quaternary ammonium as charged (functional) groups was found to be deteriorated during ED process at LCD due to chemical reaction with hydroxide ions (OH^-) generated by water dissociation [13,14]. However, it has been demonstrated by Doyen et al. [12] that working over LCD can improve the migration of some specific peptides. Moreover, a recent review by Nikonenko et al. [15] emphasized the effects of over LCD (OLCD) such as exalting effect of water dissociation products, current induced convection (electroconvection and gravitational convection) on mass transfer and their advantages in ED processes. Therefore, at this stage, it is a real challenge to define the compromised state between these two opposing effects of working in OLCD especially to find the ways to minimize CP and membrane fouling.

Different methods have been proposed to prevent the development of CP and fouling, and subsequently increase mass transfer. Some of the examples include applying non-stationary electric fields such as pulsed electric field (PEF) [16] and polarity reversal (PR) of current with change in solution flow streams [17–19]. The application of PEF has been found to intensify the ED process (demineralization) by preventing or delaying the development of CP layer and reducing the dissociation of water molecules [16]. Moreover, a significant decrease in organic as well as inorganic fouling on IEM in ED process with PEF mode as compared to conventional DC current has already been demonstrated. Indeed, Ruiz et al. [20] were the first to demonstrate that the use of PEF can prevent protein fouling on IEM as compared to DC regime. In the potentiostatic (constant voltage) mode, it is preferable to use pulse of shorter duration because the electrolyte concentration near the membrane decreases sharply as soon as current is applied to the system [16]. Furthermore, the duration of pause equivalent to 10–50% of applied pulse was considered to be sufficient to restore the state without CP. Similarly, a routine (2–4 times per hour) reversal of polarity of current has been claimed to mitigate the fouling and scaling on IEM [21,22]. The PR is also referred as an automatic self-cleaning-in-place (CIP) process. It should be noted that the application of PR of current (electrodes) also entailed reversal of solution flow streams with the help of valves which was not performed in the present study. To date, application of PR has been solely used in desalination process by ED, known as ED reversal (EDR) [23,24]. However, the effects of both PEF and PR techniques on peptide migration rate, selectivity, and membrane fouling and physicochemical properties have never been studied during EDFM.

In this context, the aim of the present study was to determine and compare the effect of PEF and PR with conventional EDFM with DC current on peptide migration rate, peptide selectivity and IEM fouling at under-limiting and over-limiting current densities. The peptide concentration as a function of time (number of charge transported) and total amino acid content in all peptide fractions recovered at the end of the treatment were determined, and the membranes used were characterized quantitatively (total nitrogen content, ATR-FTIR) as well as qualitatively (water content, electrical resistance and conductivity).

2. Materials and method

2.1. Materials and ED cell

2.1.1. Hydrolysate

A snow crab by-product hydrolysate (SCBH) was provided by the Québec fisheries and aquaculture innovation center (Merinov, Gaspé, QC, Canada). The protein hydrolysate (SCBH) was prepared following the protocol described by Beaulieu et al. [25]. Briefly, 100 kg of ground snow crab by-products was mixed with 100 L of distilled water and was hydrolyzed with an enzymatic blend, Protamex® (Novozymes, Bagsvaerd, Denmark; 1 g/kg of by-products) for one hour at pH 9 and 40 °C. Then, enzymes were inactivated by raising the temperature to 85 °C for 10 min. The SCBH fraction used in the present study was produced by pressure-driven filtration processes (ultrafiltration and nanofiltration). Moreover, The SCBH used in this work was the permeate of ultrafiltration (1 kDa) and the retentate of nanofiltration; therefore it contained mainly free amino acids and peptides of molecular weights ranging from 200 to 1000 Da [26]. The recovered SCBH fraction was then stored at −30 °C for EDUF treatment and further analyses. The total peptide/amino acids concentration in the SCBH was 140 g/L. The water and ash contents were 82 and 2.5%, respectively. As the initial electrical conductivity of SCBH solution was very high (16.65 mS/cm), it was demineralized up to 37% (10.50 mS/cm) and was stored at −30 °C. The demineralized SCBH was diluted with distilled water to a final peptide concentration of 2% (w/v) before ED with ultrafiltration membrane (EDUF) fractionation.

2.1.2. Chemicals

Hydrochloric acid and Potassium hydroxide solutions were purchased from Fisher Scientific (Montreal, QC, Canada). Sodium hydroxide and sodium sulfate were bought from Laboratoire MAT (Québec, QC, Canada) and potassium chloride was obtained from ACP Inc. (Montréal, QC, Canada).

2.1.3. Membranes

20 kDa ultrafiltration membranes (UFMs) made of polyethersulfone (PES) were purchased from Synder filtration (Vacaville, CA, USA). Neosepta AMX-SB, anion-exchange membrane (AEM) and Neosepta CMX-SB, cation-exchange membranes (CEM) were bought from Ameridia (New Jersey, USA).

2.1.4. Electrodialysis cell and configuration

A MP type electrodialysis (ED) cell manufactured by ElectroCell Systems AB Company (Täby, Sweden) with an effective surface area of 100 cm^2 was used in the present study. The EDUF cell configuration (Fig. 1) and concentrations of peptide and KCl were chosen according to the previous study which demonstrated higher degree of fouling on IEM and water dissociation [26]. The cell contained one AEM, one UFM with a MWCO of 20 kDa and two CEMs (CEM1 and CEM2). The CEMs placed near the anode and cathode were named CEM1 and CEM2, respectively. A dimensionally-stable electrode (DSA) and a 316 stainless steel electrode were used respectively as anode and cathode in the ED cell.

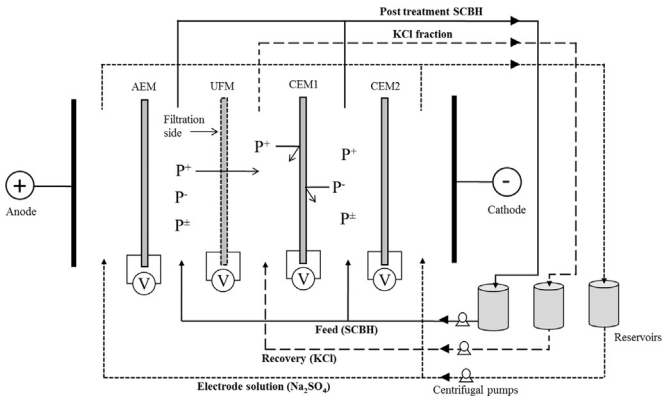

Fig. 1. Configuration of EDUF cell for the fractionation of SCBH. AEM: anion-exchange membrane, UFM: ultrafiltration membrane, CEM: cation-exchange membrane, P^+: cationic peptides, P^- anionic peptides and $P\pm$: neutral peptides and V: voltmeter connected to silver coated platinum electrode placed at the interface of membrane.