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Fouling control in ultrafiltration of bovine serum albumin and milk by the use of permanent magnetic field

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ABSTRACT

Membrane separation processes are widely employed for protein concentration in the food industry. The major drawback is permeability reduction caused by concentration polarization and fouling. The present work evaluated the influence of a permanent magnetic field applied to the ultrafiltration process (UF) of protein solutions, as an alternative to improve the permeation performance and the permeability recovery. Permeation tests of bovine serum albumin (BSA) and milk as the feed protein solutions through a 50 kDa hydrophilic polyethersulfone (PES) membrane were carried out in a tangential flow module. The feed pH was varied (4.0, 6.5 and 8.0) and ionic strength was modified by sodium chloride (NaCl). Permanent magnets were placed so as to obtain a maximum 0.7 T magnetic field perpendicular to the membrane surface. The magnetic induction effect (MI) on the feed solutions was also studied by submitting the feed to the magnetic field for 2 h before permeation run. The presence of magnetic field and the MI effect of the solution were effective in increasing both the permeate flux and the recovery of hydraulic permeability. The magnetic field application in the UF of protein solutions has proven to be an attractive alternative for improving process performance.

1. Introduction

The separation and purification of bioproducts such as proteins, protein hydrolysates, polysaccharides, vitamins and amino acids are important steps in the food industry due to the large number of applications. Processes as precipitation, crystallization and centrifugation may not result in a good selectivity, while more selective methods like electrophoresis and chromatography separation are often costly (Saxena et al., 2009).

Membrane separation processes (MSP) have found many applications in food industry, particularly in the concentration, recovery and fractionation of proteins and protein hydrolysates (Sotoft et al., 2015). MSP have numerous advantages over other methods such as cost minimization, operational flexibility and scaling up, high throughput of products and while maintaining product purity under ambient conditions, and a great cost benefit regarding energy consumption (Luján-Facundo et al., 2015).

However, a major drawback of this technology is the flux reduction during permeation due to concentration polarization and membrane fouling, which may occur by solute deposition on membrane's surface, forming a gel layer or by solute adsorption inside the pore structure of the membrane, often irreversible (Saxena et al., 2009).

Some chemical and physical strategies can be used to reduce these undesirable effects. Despite being an effective cleaning method, the use of chemicals in membrane processes may damage both the membrane and the final product (Zhang and Ma, 1999). Other forms of chemical approach to minimize fouling involve chemical modification of the membrane surface, e.g. nanoparticle coating (Moghimifar et al., 2014; Razmjou et al., 2011), surface modification by copolymerization (Li et al., 2014; Yu et al., 2011) and plasma polymerization (Zou et al., 2011). The use of physical processes toward fouling reduction are the most attractive because they do not alter the molecular structures involved in the process, and consist in clean, non-intrusive technology, since no chemical agents are used. The physical strategies include the use of

Abbreviations: UF, ultrafiltration; BSA, bovine serum albumin; PES, polyethersulfone; MI, magnetic induction; MSP, membrane separation processes; WO/MF, without the presence of the magnetic field; W/MF, with the presence of the magnetic field; MMCO, molar mass cut off; WO/NaCl, without sodium chloride; W/NaCl, with sodium chloride; IEP, isoelectric point; PC, physical cleaning; CC, chemical cleaning.

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turbulence generating devices, sonication, centrifuge and use of electric and magnetic fields (Vardanega et al., 2013).

The use of magnetic field has been proposed for minimization of scaling in heat exchangers caused by scaling (Lipus et al., 2011; Shahryari and Pakshir, 2008). Then, some studies focused on its use to control of scaling in nanofiltration and reverse osmosis membranes (Al-Qahtani, 1996; Li et al., 2007; Long et al., 2005; Vedavyasan, 2001). Although not widespread yet, the use of magnetic field in some cases attracts attention due to some advantages such as low cost and low power consumption, simple operation and low environmental impact (Gabrielli et al., 2001; Vedavyasan, 2001; Wang et al., 1997). Studies show that the magnetic field influences the layer of hydrated ions in the solution and causes changes in the hydrating water structure around the ions, changing the water conductivity (Holysz et al., 2007; Szcześ et al., 2011). The magnetic field weakens the hydrogen bonds due to the competition between the different hydrogen bond networks (intra- and intermolecular) forming smaller clusters with greater bond strength (Toledo et al., 2008). It is also reported that the magnetic field causes the decrease of the surface tension and viscosity of water, elevates the wettability of polymeric surfaces (observed by contact angle); and increases the refractive index and dielectric constant of water. The authors suggest that the externally applied magnetic fields cause displacements and polarization of molecules and atoms, and result in changes of dipole moment in the transition and vibrational states of molecules (Amiri and Dadkhah, 2006; Pang and Deng, 2008). In saline solutions, the magnetic field changes the way how the salt nucleation and growth of salt crystals occurs. Several studies reported that the effects of magnetic field are mainly in the acceleration and in the increase of precipitate amount (Alimi et al., 2009). For instance, the magnetic field application in calcium carbonate solutions increases the salt formation in the aragonite form, less stable and more soluble in water in comparison with calcite form, decreasing the deposition (scaling) in tube and heat exchangers (Tai et al., 2014; Chang and Tai, 2010). Some authors report that the application of the magnetic field promoted an increase in potassium chloride ions transport through a cellulose membrane (Ohata et al., 2004); increased flux and reduced the calcium carbonate deposition on nanofiltration membranes (Li et al., 2007; Long et al., 2005) and promoted a greater recovery of permeate initial flux after physical and chemical cleaning procedures in new and used ultrafiltration membranes (Vardanega et al., 2013). A patented device used for brackish water desalination is also reported (Ballester and Garrido, 2012).

In this context, this work assessed the application of static permanent magnetic field on the ultrafiltration of protein solutions in a tangential module as a possible alternative to improve process performance, by reducing membrane fouling and consequent increase in permeate flux. So far, no further studies on the effect of magnetic fields on protein UF can be found elsewhere, apart from the preliminary study of the group (Vardanega et al., 2013).

2. Materials and methods

2.1. Experimental apparatus

The UF unit was operated in tangential flow and consists of a feed tank, one positive displacement pump (Micropump, cat. 75211-15, Cole-Parmer, USA), a pressure gauge (Fiedler Ltda, Brazil), a backpressure valve (SS4BK, Swagelok, USA) and a flow meter (Blaster Controles Ltda, Brazil). The UF module had dimensions of $100 \times 65 \times 10$ mm, made of polyoxymethylene, with effective filtration area of 0.0029 m^2, onto where the magnets are positioned. The solution leaving the module is divided in two streams, the permeate and the retentate. Fig. 1 shows a schematic diagram of experimental apparatus used in this study, and a side view of tangential ultrafiltration module in the presence of magnets.

2.1.1. Magnetic field

The magnetic field was generated by the presence of two neodymium–iron–boron ($Nd_2Fe_{14}B$) permanent magnets with dimensions of $50 \times 50 \times 25$ mm, positioned perpendicularly to the UF module as shown in Fig. 1(b). The magnetic field intensity was measured with a magnetic field transducer (model TMAG-1T, Globalmag Ltda, Brazil). The flux density at the central point of the module was 0.7 T.

2.2. Membranes

A poly(ethersulfone) membrane (Microdyn-Nadir GmbH, Germany) with molar mass cut off (MMCO) of 50 kDa was used in all experiments. The membranes were previously treated with 99% ethanol (Vetec Ltda, Brazil) for 30 min and then rinsed thoroughly with ultrapure water. A new sample of membrane was used for each set of experimental runs.

2.3. Assay and reagent solutions

The feed solution consisted of bovine serum albumin (BSA, Sigma–Aldrich #A2153, average molecular mass 66 kDa and isoelectric point (IEP) 4.7) at a concentration of 2.5 g L^{-1}. Homogenized standardized pasteurized milk was also tested for validation purposes. The pH chosen to BSA solutions were 8.0 (higher than IEP) and 4.0 (lower than IEP) and pH 6.5 for milk (pH commonly found *in natura*). Sodium chloride (NaCl) was added to the feed solutions at a concentration of 0.5 M for testing the process performance in the presence of an inorganic salt.

The cleaning protocol consisted of sequential rinses with solutions of hydrochloric acid (HCl, at pH 4.0), sodium hydroxide (NaOH, at pH 10) and phosphate buffer (pH 7.0). Ultrapure water was used for preparing all solutions and reagents.

2.4. Experimental procedure

The experimental procedure was carried out in three different ways. The standard procedure (control) was performed without the presence of the magnet (WO/MF). The second set of assays was carried out in the presence of magnet during ultrafiltration of the protein solution (W/MF). Finally, magnetic induction effect (MI) assays were performed. The latter consisted in the protein solution circulation through the magnetic field for 2 h before the permeation process. All assays were carried out in duplicates at flow rate 0.3 L min^{-1} (0.055 m s^{-1}), while experimental errors were lower than 5%. Control runs were carried out to account for changes in the protein solution due to circulation of protein solution through the system. The membrane hydraulic permeability was determined after compaction and after physical and chemical cleaning procedures, at a flow rate of 0.3 L min^{-1}, varying the pressure from 2.0 to 0.5 bar at 25 °C.

UF of protein solutions were carried out for 120 min at constant pressure of 2 bar, feed flow rate of 0.3 L min^{-1} and 25 °C, with pH ranging according to the solution used. The assays were performed in batch mode, with full recycle of retentate and permeate, thus keeping the feed concentration constant. The return of the retentate and permeate streams caused a constant mixing in the feed tank. No precipitation of solids was observed in any of the experimental assays. Permeate flux was determined at each 15 min for BSA solutions and at each 20 min for milk solutions.