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Membrane fouling mechanisms during ultrafiltration of skimmed coconut milk

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ABSTRACT

Ultrafiltration is a promising technique to produce value-added products from skimmed coconut milk. Unavoidably, membrane fouling always hinders the membrane performance. In this work, Hermia's models were used to investigate the fouling mechanisms. Effects of the molecular weight cut-off of the polysulfone membrane (10, 20 kDa), feed solution temperature (50, 55, 60 °C) and operating pressure (1.8, 2.0, 2.2, 2.4 bar) towards the membrane fouling were analysed. The results showed that the best fit ($R^2 \geqslant 0.98$) of the experimental data to all fitted fouling mechanisms (complete blocking, standard blocking, intermediate blocking and cake formation) occurred for experiments using a 20 kDa polysulfone and 60 °C feed temperature. All fouling mechanisms were present during the ultrafiltration but dominated by complete blocking, followed by standard, intermediate blocking and cake layer formation. The characteristics of the membrane and feed solution were found to be highly influential on the membrane fouling mechanisms in this study.

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1. Introduction

Coconut (*Cocos nucifera L.*) is an important palm species due to its high oil content (around 37–50% oil by weight of coconut kernel) (Santoso et al., 1996). Thus, virgin coconut oil (VCO) has become the major commercial coconut product and has gained in popularity in recent times (Marina et al., 2009). During the production of VCO, numerous by-products are discarded such as skimmed coconut milk (SCM) and soluble proteins. The abundance of these discarded by-products has contributed to environmental issues. These discarded coconut residuals still contain numerous nutritional compounds such as proteins, carbohydrates, sugars, vitamins and so forth. SCM contains high quality proteins (70% of the total coconut protein) whereas it possesses relatively well-balanced amino acid profiles (Naik et al., 2012). The dominant types of protein in SCM are globulin and albumins. The proteins in the SCM need to be processed using certain reliable and mild processes in order to make them suitable for use as functional foods, dietary supplements and formulated milk. However, the utilization of these by-products to produce value-added products has not yet been given much attention. This proteinaceous solution (SCM) has been chosen as the solution medium in this study. So far, a few attempts have been undertaken to extract the coconut proteins. (Chen and Diosady, 2003; Samson et al., 1971). However, such processes (enzymatic extraction and chemical extraction of coconut proteins) have been reported to be less productive in recovering proteins.

Ultrafiltration (UF) has been employed to concentrate and recover the proteins from skim milk (Makardij et al., 1999), whey proteins (Nigam et al., 2008), soy flour (Krishna Kumar et al., 2004) and even wastewaters (Wu et al., 2013, 2006, 2009). However, too little attention has so far been paid on coconut proteins. A UF process has been used in this study in order to obtain high quality protein concentrates from SCM. Membrane can be used to produce coconut protein concentrates that possess high functional and nutritional characteristics. The UF membrane performs based on a size-sieving mechanism. It has been postulated that the UF polymeric membrane manages to concentrate the coconut proteins while allowing the permeation of water and other smaller sized compounds. However, a complex protein mixture (such as SCM) can easily reduce the efficiency of the UF process due to the tendency of the membrane to foul. The permeate flux and selectivity of membrane are hindered due to the membrane fouling issue, especially when the involved subject is a complicated proteinaceous solution (Chan et al., 2002; Wu et al., 2007).

The characterization of membrane fouling mechanisms is highly important especially during the UF process. In order to

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Nomenclature

UF	ultrafiltration
MWCO	molecular weight cut-off (kDa)
PSF	polysulfone
VCO	virgin coconut oil
SCM	skimmed coconut milk
R^2	coefficient of determination
Jvt	volume flow at a particular time (m^3/m^2 h)
Jvo	initial volume flow rate (m^3/m^2 h)
εc	constant of the complete blocking model (s^{-1})
εs	constant of the standard blocking model (s$^{-1/2}$ m$^{-1/2}$)
εi	constant of the intermediate blocking model (m^{-1})
εcf	constant of the cake formation model (s m^{-2})
Kc	membrane surface area blocked per unit of permeate volume (m^{-1})
Ks	reduction in the cross-sectional area of the pores per unit of permeate volume (m^{-1})
Kck	area of cake formed per unit of permeate volume (m^{-1})
Ro	resistance of the fresh or clean membrane (m^{-1})
Rc	resistance of the cake layer formed (m^{-1})
Rr	$Rr = Rc/Ro$, ratio of resistance of the cake over the resistance of the clean membrane (dimensionless)
Vo	initial mean velocity of the filtrate through the membrane (m/s)
t	time (s)

produce higher permeate flux and greater solute rejections, a clear understanding on the membrane fouling mechanisms is essential. Besides, a loss in selectivity of the main products can occur during the occurrence of membrane fouling in UF process (Mohammad et al., 2012). Membrane fouling can either occur by the deposition of particles inside the membrane pores, on the membrane surface or a combination of both (Ho and Zydney, 2000; Kelly and Zydney, 1997; Maruyama et al., 2001). When the particles deposit on the membrane surface and cover up the membrane surface pores, the permeability of the membrane can be reduced as these particles will restrict the passage of the water molecules through the membrane surface pores. The extent of fouling strongly depends on the feed solution properties, membrane materials and operating conditions (Ramesh Babu and Gaikar, 2001). Many studies had attempted to improve the filtration processes of complex solutions by various methods such as varied experimental conditions, adjusted feed solution behaviour, modified operational design and the used of modified membrane (Boyd and Zydney, 1998; De Bruijn et al., 2005; Palacio et al., 2002; Rahimpour, 2011). However, the characterization and prediction of membrane fouling in typical condition remain as a challenge in the membrane technology.

In order to minimize membrane fouling during the ultrafiltration of SCM, various membrane fouling mechanisms need to be pre-evaluated. All the fouling mechanisms of the membrane can be described by blocking filtration laws. These blocking filtration laws consist of complete blocking, standard blocking, intermediate blocking and cake filtration mechanisms. This fouling model was firstly introduced by Hermia and named Hermia's model (Hermia, 1982). There have already been a few studies that analysed the fouling of a membrane using Hermia's model. Recently, researchers (Nourbakhsh et al., 2013) employed Hermia's model to describe fouling mechanisms during the clarification of red plum juice. They concluded that cake formation was the predominant fouling mechanism during juice clarification. However, other fouling mechanisms (complete, standard and intermediate pore blocking) were also involved when the filtration time was prolonged. In addition, Hermia's model has also been used in the UF of polysaccharide macromolecules (Sarkar, 2013). In this case, they found that a proposed flux model in which complete pore blocking predominated in the early stage of the UF process could satisfactorily describe the flux decline. This phenomenon has been frequently studied during past decade for the filtration of protein and polysaccharide solutions (Feng et al., 2009; Palacio et al., 2002). In addition, Hermia's model has also been used to determine nature of membrane fouling during the application of wastewater (Salahi et al., 2010), glycerine solution (Amin et al., 2010), colloidal suspensions (Wang and Tarabara, 2008), polyethylene glycol (Vela et al., 2008) and oil in water emulsions (Mohammadi et al., 2003).

To minimize fouling while using polymeric membranes, experimental parameters that influence reductions in flux need to be studied. Therefore, the fouling mechanisms involved in this study needed to be identified during ultrafiltration of SCM under different operating conditions including pressure (1.8, 2.0, 2.2, 2.4 bar), the membrane's MWCO (10 kDa, 20 kDa) and temperature (50 °C, 55 °C, 60 °C). Effects on membrane fouling due to the changes in operating conditions were also studied. The fouling mechanisms were analysed using Hermia's model and the predictions compared with the experimental data obtained.

1.1. Prevailing fouling mechanisms

Hermia (1982) derived a mathematical model (Eq. (1)) to describe permeate flux decline phenomena. The derivation of this theoretical model is typically based on the classic constant-pressure filtration process. The fouling mechanism can be identified using the so-called blocking filtration law or Hermia's model.

$$(d^2t/dV^2) = k(dt/dV)^n \tag{1}$$

The exponent n in Eq. (1) characterizes the type of filtration mechanism. In the following sessions, further description of each fouling mechanism will be given.

1.1.1. Complete pore blocking (n = 2)

Complete blocking normally occurs when the sizes of filtration solutes are greater than the pore openings in the membrane. The solutes will completely obstruct or seal the openings of the membrane pores without superposition of the solutes. The filtration resistance will increase as the number of unblocked membrane pores decreases (Hwang and Lin, 2002). As a result, the permeate flow rate will decrease exponentially with time.

The filtration volume flow can be related to the time using Eq. (2):

$$Jvt = Jvo[exp(-\varepsilon ct)] \tag{2}$$

where $\varepsilon c = KcVo$

Kc is the area of membrane surface blocked per unit of total volume permeated through the membrane and Vo is the mean initial velocity of the filtrate or initial volume flow per unit of porous membrane surface area.

Thus, the estimated evolution over time of the permeate volume flow is given by Eq. (3):

$$lnJvt = lnJvo - \varepsilon ct \tag{3}$$

1.1.2. Standard pore blocking (n = 1.5)

Standard blocking, also called internal pore blocking. This fouling mechanism occurs when small size solutes deposit or adsorb

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