Contents lists available at ScienceDirect

Food and Bioproducts Processing

journal homepage: www.elsevier.com/locate/fbp

Integrated membrane process for purification and concentration of aqueous *Syzygium cumini* (L.) seed extract

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ARTICLE INFO

Article history:
Received 21 May 2015
Received in revised form 23 November 2015
Accepted 14 December 2015
Available online 23 December 2015

Keywords:
Syzygium cumini (L.)
Polyphenol
Membrane processes
Concentration polarization

ABSTRACT

The aim of the present study was to investigate the potentiality of an integrated membrane process for the purification and the concentration of phenolic compounds from aqueous jamun (*Syzygium cumini* L.) seed extract. The aqueous seed extract obtained at optimal condition (temperature: 49.2 °C, time: 89.4 min, and liquid to solid ratio: 51.6:1 mL/g) was submitted to cross flow ultrafiltration for initial clarification, followed by concentration using nanofiltration under batch concentration mode. A detailed parametric study was carried out to investigate the effect of various process parameters such as transmembrane pressure, cross-flow velocity (or stirrer speed) on the permeate flux and permeate quality. Using classical film theory, a steady state gel polarization model incorporating the effect of transmembrane pressure difference and viscosity variation was proposed for the prediction of permeate flux during cross flow ultrafiltration of aqueous seed extract. The predicted flux values were successfully compared with the experimental results. Experimental results showed that the operating conditions had significant effect on permeate flux, recovery of polyphenols, purity and antioxidant activity of phenolic extract. Ultrafiltration experiments at lower operating pressures (276 and 414 kPa) using 100 kDa membrane resulted in the recovery of 59–66.7% of total polyphenol content in the clarified extract with the purity of 49–58.3% starting from an extract purity of 39.2%. The clarified extract could be successfully concentrated about three times higher using 250 Da nanofiltration membrane at volume concentration ratio of three. The present study revealed that the UF/NF integrated membranes process was successful in clarifying and concentrating phenolic extract obtained from jamun seed with enhanced purity and antioxidant activity.

1. Introduction

Jamun (*Syzygium cumini* L.), is one of the nutritious fruit of the Myrtaceae family. Jamun fruit, native to India, is widely cultivated in the various parts of India (ICAR Report, 2014). It is also being grown in most of the tropical and subtropical regions of the world such as Thailand, Philippines, Madagascar, West Indies, California, and Algeria. The domestic and industrial use of these large quantities of jamun fruit, especially for the production of juice and wine, results in the accumulation of large amounts of seed as a by-product which account about 20% of the fruit weight. This represents a serious disposal problem from an economical and environmental point of view (Patil et al., 2012). Previous studies have reported that jamun seed extract is a rich source of phenolic compounds with antioxidant, anti-inflammatory capacity, such as ellagitannins, gallic acid, β-sitosterol and ellagic acid (Kaneria and Chandra, 2013; Swami et al., 2012). The

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http://dx.doi.org/10.1016/j.fbp.2015.12.005

Nomenclature

a_1, a_2, a_3	coefficients in Eq. (A1)
a, b	model parameter
C	concentration of solute, kg/m^3
C_o	solute concentration in feed, kg/m^3
C_b	solute concentration at bulk, kg/m^3
C_g	solute concentration in gel layer, kg/m^3
C_g^*	dimensionless solute concentration in gel layer defined as, $C_g^* = C_g/C_0$
C_p	solute concentration in permeate, kg/m^3
d_e	equivalent diameter, m
D	effective diffusivity of solute, m^2/s
k_0	mass transfer coefficient, m/s
k_1	modified mass transfer coefficient, m/s
L	channel length, m
N	number of experimental data
P_{ew}	dimensionless permeate flux defined as, $P_{ew} = V_w d_e/D$
Re	Reynolds number, dimensionless
R_g	gel layer resistance, m^{-1}
R_m	membrane resistance, m^{-1}
R_g^*	dimensionless gel layer resistance defined as, $R_g^* = R_g/R_m$
R_t	total resistance, m^{-1}
R_{rev}	reversible fouling resistance, m^{-1}
R_{irr}	irreversible fouling resistance, m^{-1}
R_{CP}	concentration polarization resistance, m^{-1}
R_{CP}^*	dimensionless concentration polarization resistance defined as, $R_{CP}^* = R_{CP}/R_m$
R_0	observed retention, dimensionless
Sc	Schmidt number, dimensionless
Sh	Sherwood number, dimensionless
Sh_m	modified Sherwood number, dimensionless
t_0	initial time, s
t_f	final time, s
V_P	volume of permeate, mL
V_R	volume of retentate, mL
V_w	permeate flux, $L/m^2\,h$
V_w^0	initial permeate flux, $L/m^2\,h$
V_w^f	final permeate flux, $L/m^2\,h$
$V_{w,ss}^{exp}$	experimental steady state permeate flux, $L/m^2\,h$
$V_{w,ss}^{cal}$	calculated steady state permeate flux, $L/m^2\,h$
ΔP	transmembrane pressure difference, kPa
μ_0	viscosity of bulk solution, Pa s
μ_m	viscosity at membrane surface, Pa s
δ_c	thickness of concentration boundary layer, m
β	parameter in Eq. (A7)

recovery of any target compound from food by-products can be accomplished with the so-called "5-Stages Universal Recovery Processing" including: (a) macroscopic pre-treatment, (b) separation of high-molecular from low-molecular compounds, (c) extraction, (d) purification/isolation and (e) encapsulation or product formation (Galanakis, 2012, 2014). In the recovery downstream processing, microfiltration and ultrafiltration are able to remove macro-molecules (i.e. proteins, pectin, etc.) while nanofiltration isolate, purify and concentrate target compounds prior their encapsulation with conventional spray or freeze drying (Galanakis, 2015). The energy efficient membrane technologies have been used in different sectors of food industry for several decades.

Nowadays, membrane technology has become one of the most important industrial separation technique and has been a topic of growing interest for purifying and concentrating bioactive phenolic compounds from extract of various plant sources, such as fruits, seed, peels, leaves, roots, and barks (Conidi et al., 2011, 2012; Chhaya et al., 2011; Yu et al., 2007; Prudêncioa et al., 2012) although some emerging technologies are explored recently in research level and in some cases applied in the food industry (Galanakis, 2013). During extraction of phenolic compounds, higher molecular weight polysaccharides (cellulose, pectins, etc.) are also co-extracted leading to decrease in purity of the phenolic extract. The aim of the membrane-based clarification of plant extract is the removal of polysaccharides and maximum permeation of phenolic and flavonoid compounds resulting to an increase in purity of the extract. Microfiltration (MF) or ultrafiltration (UF) represent well-established technologies in the clarification of plant extracts, while concentration by nanofiltration (NF) or reverse osmosis (RO) has been reported in several studies (Díaz-Reinoso et al., 2009; Murakami et al., 2011; Mello et al., 2010). Recently, the potential applications of integrated membrane process have been proposed for concentration of plant extracts (Cissé et al., 2011; Chhaya et al., 2012; Torun et al., 2014) as well as fruit juices (Cassano et al., 2003, 2006; Alvarez et al., 2000).

Very few studies with limited information are available on the antioxidant potential of the jamun seed (Swami et al., 2012). However, there are almost no scientific references dealing with the treatment of jamun seed extract by membrane processes. For this reason the present work is focused on the aqueous extract from jamun seed and in membrane processing for clarification/concentration of polyphenols. Although there are many advantages of membrane-based processes, a common phenomenon in this process is the decline in permeate flux with time. This occurs due to concentration polarization and membrane fouling. In the jamun seed extract clarification, the process is mainly limited by the accumulation of pectin, starch, hemicelluloses, cellulose, etc. over the membrane surface. Membrane fouling can be reduced by selecting a suitable membrane and set of operating conditions. Hence, the role of operating conditions is extremely important in membrane separation processes.

The aim of this work is to evaluate the potential of an integrated membrane process, by selecting suitable membrane and operating conditions, for purifying and concentrating phenolic compounds from jamun seed extract using water as a solvent. In particular, aqueous jamun seed extract is submitted to a preliminary purification process using UF membranes, followed by a concentration by nanofiltration. Furthermore, a suitable model has been formulated to predict steady state permeate flux during cross flow ultrafiltration of seed extract. Aqueous seed extract being a complex mixture of several solutes, the unknown transport properties like gel layer concentration, solute diffusivity etc. are difficult to estimate. However, these parameters are system specific and their estimation is warranted for efficient design and subsequent scaling. Moreover, during ultrafiltration there is a significant variation of solution viscosity and solute diffusivity within thin concentration boundary layer near the membrane surface due to sharp variation in the solute concentration. The ultrafiltration of seed extract is assumed to be a gel controlling and the proposed model is based on classical film theory. The