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Separation and purification of phenolic compounds from pomegranate juice by ultrafiltration and nanofiltration membranes

Carmela Conidi, Alfredo Cassano*, Francesca Caiazzo, Enrico Drioli

Institute on Membrane Technology, National Research Council, ITM-CNR, Via P. Bucci, 17/C University of Calabria, I-87036 Rende, Cosenza, Italy

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ABSTRACT

Pomegranate juice is well recognized for its nutritional and health benefits due to the presence of phenolic compounds, including anthocyanins, ellagic acid, phytoestrogenic flavonoids and tannins. Therefore, the demand for the production of functional foods containing bioactive compounds isolated from the juice has remarkably increased in the last decade.

In this study ultrafiltration (UF) and nanofiltration (NF) flat-sheet membranes, with nominal molecular weight cut-off (MWCO) ranging from 1000 to 4000 Da, were tested to purify biologically active compounds from clarified pomegranate juice. The filtration process was evaluated in a crossflow pilot unit equipped with a Sepa CFII Membrane Cell System featuring an effective membrane area of 0.014 m^2.

A first screening was made in order to evaluate the performance of selected membranes in terms of productivity, fouling index and retention towards sugars, phenolic compounds and total antioxidant activity. Among these membranes the Desal GK membrane, with a MWCO of 2000 Da, displayed higher permeate fluxes, lower fouling index and a good separation efficiency of sugars from phenolic compounds in comparison with the other tested membranes. Therefore further experiments were addressed to evaluate the separation capability and the productivity of this membrane at different transmembrane pressure (TMP) values. Concentration/diafiltration experiments were also performed in order to obtain a retentate fraction enriched in phenolic compounds and a permeate stream mainly containing glucose and fructose.

According to the proposed process the yields of polyphenols and anthocyanins in the retentate stream were of the order of 84.8% and 90.7%, respectively. The diafiltration step allowed to obtain a recovery efficiency in the permeate side for glucose and fructose up to 90% and 93%, respectively.

1. Introduction

Pomegranate is one of the first five cultivated foods in the world widely grown in many countries including Iran, India, Turkey, Tunisia, Pakistan, China, USA and Spain. Popular in Eastern as well as Western parts of the world, pomegranate thrives well in regions with semi-arid and sub-tropical climatic conditions but is also naturally adapted in regions with cold winters and hot summers (Ozgen et al., 2008). The total world production is estimated currently at 2 million tons/year (Erkan, 2011). In recent years, the interest for pomegranate fruit and its derivatives has increased remarkably as evidenced by hundreds of publications on their chemical composition, potential uses and proven health-promoting effects (Gumienna et al., 2016; Jurenka, 2008; Lansky and Newman, 2007). Several studies have focused on the ability of different components of the fruit, including the juice, seed oil, peel, flower extracts or their derivatives to protect against several diseases such as cancer (Dai and Mumper, 2010), type 2 diabetes (Banihani et al., 2013), atherosclerosis (Al-Jarallah et al., 2013) and cardiovascular diseases (Aviram et al., 2008) providing the scientific basis for some use of pomegranate in traditional medicine. In addition, these products have been shown to posses antimicrobial, anti-hepatoxic and antiviral properties (Faria and Calhau, 2011). These health benefits have been attributed to the high antioxidant capacity that is strongly correlated with the high concentration and chemical composition of phenolic anthocyanins and hydrolysable tannins such as punicalagins, punicalin, peduncalagin, and ellagic acid (Vegara et al., 2013). Different studies have also shown that the antioxidant activity of pomegranate juice is much higher than most other fruit juices and beverages (Gil et al., 2000; Seeram et al.,

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* Corresponding author.
E-mail address: a.cassano@itm.cnr.it (A. Cassano).

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2008).

Given the wide spectrum of health promoting activities exerted by pomegranate and the enormous interest that bioactive compounds isolated from this fruit have raised in the scientific community, the interest of researchers has been addressed in recent years to the optimization of the extraction and purification procedures of these compounds for the development of functional foods meeting the consumer requirements.

Solvent organic extractions (SOEs) are the most commonly used procedures to extract bioactive compounds from pomegranate fruits (Sood and Gupta, 2015). It is generally known that the yield of chemical extraction depends on type of solvent (polarity), extraction time and temperature (Singh et al., 2014). However, solvents commonly employed such as methanol, ethanol, acetone and ethyl acetate are not always “food friendly” and not suitable or safe for their utilization in the food industry (Amyrgialaki et al., 2014). In addition, long extraction times and high temperatures may produce an oxidation of phenolics leading to a decreased yield of phenolics in the extracts. It has been also shown that high temperatures (>70 °C) cause a rapid degradation of anthocyanins (Havlíková and Miková, 1985). Alternative methods, such as microwave extraction (Zheng et al., 2011), ultrasound-assisted extractions (Tabaraki et al., 2012) and supercritical fluid extractions (He et al., 2012), have been also applied in the extraction of phenolic compounds from pomegranate peels and seeds. However, low extraction efficiency, partial oxidation and degradation of compounds of interest, high requirements of instrumentations and costs on industrial scale are typical drawbacks which often outweigh the technical benefits. Therefore, it is of critical importance to select efficient extraction procedures in order to maintain the stability of phenolic compounds.

In this contest, membrane separation processes (MSPs) represent a valid alternative to traditional technologies due to their low operating and maintenance costs, mild operating conditions of temperature and pressure, easy control and scale-up and highly selective separations. They do not require any extraction mass agents or chemical additives, avoiding product contaminations and preserving the biological activity of the compounds of interest (Drioli and Romano, 2001).

Pressure-driven membrane operations, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) are today well-established technologies in food and beverage industries for the treatment of several products and by-products (Daufin et al., 2001; Patsioura et al., 2011; Tylkowski and Tsibranska, 2015). Other membrane processes, such as osmotic distillation (OD), membrane distillation (MD) and pervaporation (PV) have been also investigated in recent years for selected applications in the same area. Moreover, the development of hybrid processes based on the combination of different membrane unit operations and conventional separation technologies offers new and much more opportunities in terms of competitiveness, improvement of quality, process or product novelty and environmental friendliness (Cassano, 2016; Conidi et al., 2014). The use of membrane technology in the treatment of pomegranate juice has been recently investigated. In particular, MF and UF processes have been studied to clarify pomegranate juice as alternative technologies to the traditional use of fining agents (gelatin, bentonite, diatomaceous earth, silica sol, etc.) and other techniques including centrifugation, decantation, depectinization and filtration (Baklouti et al., 2012; Cassano et al., 2015; Mirsaeedghazi et al., 2010a, 2010b); MD and OD have been evaluated for their potential in the concentration of the juice as alternative to the thermal evaporation (Cassano et al., 2011; Onsekizoglu, 2013).

In recent years UF and NF operations have gained a great interest for the separation and concentration of bioactive compounds from plant extracts and by-products of agro-food industries (Cassano et al., 2014; Cissè et al., 2011; Díaz-Reinoso et al., 2009; Galanakis et al., 2013; Giacobbo et al., 2013; Li and Chase, 2010; Mello et al., 2010; Murakami et al., 2013; Tsibranska and Tylkowski, 2013; Tylkowski et al., 2010). According to the so-called “5-Stages Universal Recovery Processing” approach, the production of target compounds from food wastes includes the following steps: (i) macroscopic pre-treatment, (ii) separation of macro-from micromolecules, (iii) extraction, (iv) purification and (v) product formation (Galanakis, 2012). UF and NF are considered key physicochemical and non-destructive techniques applied in the second, third and fourth step of the above downstream processing (Galanakis, 2015).

NF membranes have a nominal pore size in the range of 0.5–1 nm; the typical range of MWCO levels is between 200 and 1000 Da. UF involves the use of membranes with a MWCO in the range of 1–300 kDa and a pore size of about 0.01 μm (Baker, 2004). The separation capabilities of UF and NF membranes are mainly related to size exclusion but interactions between solutes and membrane like charge interactions, bridging and hydrophobic interactions may play an important role in the formation of fouling layers at the membrane surface (or within the membrane pores) which will exert some influence on passage of solutes through the membrane. The formation of fouling layers due to macromolecules like proteins and dietary fibres has been also reported in literature (Galanakis et al., 2014; Patsioura et al., 2011). Moreover, diafiltration conditions can be employed in order to remove contaminants with low molecular weight (MW) from valuable products with higher molecular weight in order to increase the product yield of the process (Aspelund and Glatz, 2010; Teixeira et al., 2014).

No literature is readily available on the performance of UF and NF membranes for the separation and purification of phenolic compounds from sugars in pomegranate juice. In the light of these considerations, this work investigated the performance of flat-sheet UF and NF membranes, with different membrane material and molecular weight cut-off (MWCO), for separating and concentrating phenolic compounds from clarified pomegranate juice. The performance of the selected membranes was compared in terms of permeate fluxes, retention towards sugars, total antioxidant activity and biologically active compounds (mainly total polyphenols and anthocyanins). To fulfil the final aim to purify the selected bioactive compounds from sugars, the membrane process was also studied in a diafiltration mode.

2. Material and methods

2.1. Pomegranate juice extraction and clarification

Pomegranates, of Calabria origin, were purchased from a local open market (Cosenza, Italy). Fruits were washed in cold tap water and drained. They were manually cut-up into two halves and then squeezed by using an electric juicer (Aristalco S.r.l., Treviso, Italy). The obtained juice, having a deep-red color, was pre-filtered with a cotton fabric filter. The extracting procedure gave an average juice yield of 40% (w/w).

The raw juice was clarified by using a laboratory unit supplied by Verind SpA (Milan, Italy) equipped with a cellulose triacetate UF membrane module (FUC 1582, Microdyn Nadir, Wiesbaden, Germany) in hollow fiber configuration with a nominal MWCO of 150 kDa ad a membrane surface area of 0.26 m^2. The juice filtration was conducted according to the batch concentration mode (the retentate stream was flowed back to the feed tank while the permeate stream was collected separately) up to a weight reduction factor (WRF) of 4.8. The WRF is defined as the ratio between the initial feed weight and the weight of the resulting retentate