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Deacidification of cranberry juice by electrodialysis: Impact of membrane types and configurations on acid migration and juice physicochemical characteristics

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ABSTRACT

Cranberry is a typical fruit from North America having a very high acidity that makes raw juice hardly acceptable for consumers. In this study, the reduction of juice acidity was investigated using electrodialysis (ED) with different types of membranes and cell configurations. Electrodialysis is an ecofriendly membrane technology used in a large range of food applications. Three different ED configurations were tested at the laboratory scale: bipolar and anion-exchange membranes (ED2MB), bipolar and ultrafiltration membranes (ED2MBUF), and cation-exchange and ultrafiltration membranes (EDUF). Each configuration was evaluated in terms of juice physicochemical parameters (titratable acidity, conductivity, total soluble solids, color, anthocyanins, organic acids and mineral contents) and electrodialytic parameters (membrane conductivity and thicknesses, global system resistance). In ED2MB configuration, a 40% deacidification rate was reached after 3 h of treatment (80% after 6 h) whereas 0% and only 8% were obtained after 3 h with ED2MBUF and EDUF configurations, respectively. Furthermore, a selective migration of organic acids was observed with the ED2MB configuration: citric acid (22 ppm/min), malic acid (11 ppm/min) and quinic acid (6.5 ppm/min). Consequently, ED2MB configuration allows the deacidification of cranberry juice and the production of pure acids (no waste generated) without any chemical consumption due to the bipolar membrane in-situ generation of proton and hydroxyl species from water.

1. Introduction

Cranberry is a typical fruit from North America, belonging to the family "ericaceous" and the gender "Vaccinium". This fruit is well recognized for its beneficial effects on human health due to its high concentrations in polyphenols such as anthocyanins (galactosides and arabinosides of cyanidin, peonidin, malvidin and myricetin) and proanthocyanidins (PACs) [1]. Several studies demonstrated that cranberry juice has a preventive effect against urinary tract infection and reduces ex-*vivo* adherence of *Escherichia coli* to vaginal epithelial cells [1,2]. Proanthocyanidins present in cranberry juice could reduce the bacteria colonies in vaginal epithelial cells [3,4]. These compounds could also prevent gastric ulcers caused by *Helicobacter pylori* [5], reduce cardiovascular risk factors [6] and inhibit the formation of bacterial complexes in dental plaques [7].

Consumers have been attracted by the health benefits attributed to cranberry juice but its very high organic acid content and low pH create side effects that limit its consumption. Hence, in clinical trials, high rates of withdrawals (around 40%) were observed after cranberry juice consumption due to undesirable effects (diarrhea, vomiting and bloating) [1,5,8]. Organic acids responsible for the high titratable acidity of cranberry juice are citric, quinic, malic, and succinic acids [9]. Quinic acid is present in many fruits and vegetables such as lemon, melon, peach, apple, red pepper and tomato but its concentration in cranberry juice is higher than in other fruits. Quinic acid is the second most important acid in concentration in the cranberry juice and is the reference compound for detecting cranberry juice adulteration [10–13].

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In order to respond to consumers demand, a selective process to deacidify cranberry juice without changing its physicochemical and organoleptic properties needs to be developed. Electrodialysis, an electrochemical process based on the migration of charged molecules through ion-exchange membranes under an electric field, was tested in various laboratory applications such as fruit juice deacidification, wine stabilization and anthocyanins enrichment in cranberry juice [9,14,15]. Electrodialytic deacidification is an ecofriendly process since there is absence of chemical reagents, no waste generation and production of a high quality food product. Indeed, calcium salt precipitation and ion-exchange resin methods, which are currently used for juice deacidification, present significant disadvantages such as addition of chemical reagents, modification of juice aroma and generation of a large quantity of effluents [16]. Consequently, electrodialytic deacidification, is a very interesting method in a context of sustainable development. Very recently, a study demonstrated the feasibility of cranberry juice deacidification by electrodialysis with bipolar membrane but at a final deacidification rate of 22.84% after 6 h of treatment with a two steps ED configuration [17]. In addition, due to the configuration tested (combination of bipolar, cation-exchange and anion-exchange membranes), decrease in anthocyanin contents and modification of juice color were observed after 6 h of treatment.

In this context, the objectives of this work were (1) to compare different cell configurations and type of membranes in terms of organic acid migration rates and electrodialytic parameters (thicknesses and electrical conductivities of membranes and global system resistance) and (2) to study the evolution of the cranberry juice physicochemical properties including conductivity and pH during the three electrodialytic deacidification processes tested.

2. Materials and methods

2.1. Cranberry juice

The clarified and pasteurized cranberry juice was obtained from fresh fruits (Fruit d'Or, Notre-Dame-de-Lourdes, Quebec, Canada). The juice was stored at −20 °C and thawed at 4 °C before each experiment. The physicochemical characteristics of the cranberry juice used for the experiment are presented in Table 1.

Table 1
Physicochemical characteristics of the clarified and pasteurized cranberry juice.

pH	2.4 ± 0.1
Conductivity (mS/cm)	2.8 ± 0.2
Titratable acidity (g/L of citric acid monohydrate equivalents)	9.5 ± 1.7
Total soluble solids (° Brix)	7.0 ± 0.5
Colorimetry	
L*	27.1 ± 0.2
a*	1.5 ± 0.4
b*	−0.4 ± 0.1
Total proanthocyanidins (mg/L)	106 ± 11
Total polyphenols (mg equivalent gallic acid/L)	680 ± 70
Anthocyanin contents (mg/L)	
Cyanidin-3-galactose	13.8 ± 1.2
Cyanidin-3-glucose	0.3 ± 0.0
Cyanidin-3-arabinose	14.4 ± 1.3
Peonidin-3-galactose	20.0 ± 1.9
Peonidin-3-glucose	1.8 ± 0.2
Peonidin-3-arabinose	10.8 ± 1.0
Organic acid contents (mg/L)	
Citric acid	9800 ± 800
Quinic acid	8200 ± 300
Malic acid	5000 ± 500
Succinic acid	7600 ± 1000

2.2. Electrodialytic configurations

Electrodialysis experiments were performed using a MP type cell (ElectroCell AB, Täby, Sweden) with an effective surface area of 100 cm^2. Three different ED configurations were tested (Fig. 1).

2.2.1. ED2MB

In this configuration, two compartments (raw juice and organic acids recovery compartments) were formed by stacking two bipolar membranes (BP-1, Tokuyama Soda Ltd., Tokyo, Japan) and one food grade Neosepta anion-exchange membrane (AMX-SB, Tokuyama Soda Ltd., Tokyo, Japan) (Fig. 1a). Bipolar membranes allow the formation of organic acid through the production of H^+ in C1 compartment while the anionic membrane allows the migration of organic acids according to their p*K*a.

2.2.2. ED2MBUF

In comparison with the previous configuration, the anion-exchange membrane (AEM) was replaced by a polysulfone ultrafiltration membrane (UF) with a molecular weight cut off of 3 kDa (GE, Polysulfone, France) (Fig. 1b). The UF membrane would facilitate the migration of organic acid due to its large cut off, so theoretically the desired acidification rate can be reached more quickly.

2.2.3. EDUF

In this configuration, the two bipolar membranes of configuration ED2MBUF were replaced by cation-exchange membranes (CMX-SB, Tokuyama Soda Ltd., Tokyo, Japan) (Fig. 1c).

For each configuration, the cell had three closed loops, connected to separate external reservoirs and allowing recirculation of the three solutions (acid recovery, cranberry juice and electrode rinsing solutions) during treatment. The solutions were circulated using three centrifugal pumps and the flow rates controlled by flow-meters (Aalborg Instruments and Controls, Inc., Orangeburg, USA). The anode was a dimensionally stable electrode (DSA-O_2) and the cathode was a food grade stainless steel electrode. The anode/cathode voltage difference was supplied by an electrical power supply (Model HPD 30-10, Xantrex, Burnaby, Canada).

2.3. Protocol

A 20 g/L NaCl solution was circulated in the electrode rinsing compartments of all three configurations but a 2 g/L KCl solution was used in the recovery compartment of the ED2MB and EDUF configurations. In ED2MBUF configuration, a 15 g/L citrate solution was used in the recovery compartment instead of KCl to avoid migration of potassium in the cranberry juice through UF membrane which could have a negative effect on juice composition and flavor. Cranberry juice and KCl/citrate solutions flow rates were both maintained constant at 400 mL/min whereas a flow rate of 450 mL/min was used for the NaCl solution. The volume of cranberry juice and KCl solutions were 800 mL whereas for electrolyte solution it was 1 L and the volume remained constant during experiment. Treatments were performed at room temperature under a constant electric field of 10 V corresponding respectively to averaged current densities of 38, 10 and 11 A/m^2 for ED2MB, ED2MBUF and EDUF configurations. The current density for ED2MBUF and EDUF configurations is four times lower than ED2MB configuration since ultrafiltration membranes are less conductive than ionic exchange membrane leading to a lower current density. The treatment duration was set for 3 h except for the ED2MB configuration which was prolonged to 6 h, to determine the full potential of this configuration. In all configurations, three replicates were performed. The electrical conductivity and the pH of both the cranberry juice and the recovery solution were recorded every hour.