Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of Membrane Science 254 (2005) 101–109
journal of MEMBRANE SCIENCE
www.elsevier.com/locate/memsci

Electromigration feasibility of green tea catechins

David Labbé [a,b], Monica Araya-Farias [a,b], Angelo Tremblay [a,c], Laurent Bazinet [a,b,*]

[a] *Institute of Nutraceuticals and Functional Foods (INAF), Université Laval, Sainte-Foy (QC), Canada G1K 7P4*
[b] *Department of Food Sciences and Nutrition, Pavillon Paul Comtois, Université Laval, Sainte-Foy (QC), Canada G1K 7P4*
[c] *Department of Social and Preventive Medicine, PEPS, Université Laval, Sainte-Foy (QC), Canada G1K 7P4*

Received 8 March 2004; received in revised form 24 September 2004; accepted 25 October 2004
Available online 17 March 2005

Abstract

Epigallocatechin gallate (EGCG) is regarded as the most important of the tea catechins. Therefore, methods for producing tea extracts with high EGCG content have been developed. However, these methods have the disadvantages to use solvent or to allow the purification of small volumes. The aim of this exploratory study was to evaluate the feasibility of selectively extracting catechins and caffeine from a green tea solution using an electrodialysis cell. Commercially available membranes (AMX-SB, AFN, PC-400 D and UF-1000 Da) were tested for their potential to allow migration of green tea catechins. This study demonstrated that epigallocatechin (EGC) and EGCG from a green tea infusion can migrate at a high rate through an electrodialysis (ED) system. Among tested membranes, the UF-1000 Da membrane can achieve an EGC and EGCG migration as high as 50%. The other studied catechins and caffeine had no significant migration rate through either the anionics or the UF membranes. Thus, this method combined to a previously developed EGCG preconcentration procedure might allow the production of a green tea extract with highly active biological compound.

Keywords: Electromigration; Ultrafiltration membrane; EGCG; Catechins; Separation

1. Introduction

Tea is the second most consumed beverage around the world, after water, for its attractive aroma and flavour, as well as for its health-related benefits [1,2]. Green tea, a non-fermented tea, contains a large amount of catechins, up to 20–30% of the green tea dry weight [1–3]. Catechins are antioxidants [4–6] having a beneficial biological activity [3,7]. Five of the six main catechins present in green tea and known to possess biological properties are: (−)-epicatechin (EC), (−)-epigallocatechin (EGC), (−)-epicatechin gallate (ECG), (−)-epigallocatechine gallate (EGCG), and (−)-gallocatechin gallate (GCG) [8]. Presence of OH groups on catechins imply a susceptibiliy to the effect of pH and thus, an ionization of the molecule [9].

EGCG is regarded as the most important of the tea catechins because of its high content in tea and the fact that its activity is mirrored by green tea extracts. Therefore, methods for producing tea extracts with high EGCG ratios have been developed. Copeland et al. [10] have developed a method to produce EGCG from a decaffeinated aqueous brew of commercial green tea. In this method, the precipitate containing the EGCG is redissolved after decaffeination with chloroform and further purified by solvent partition with ethyl hexanoate and propyl acetate. In recent years, high-speed counter-current chromatography (HSCCC) has become a very useful tool for fractionation of both crude extracts and semi-purified fractions. This technique has been successfully used to produce purified catechin from crude green tea extracts [11,12]. However, these methods have the disadvantages to use solvent (method of Copeland et al. [10]) or to allow the purification of small volumes in the case of HSCCC.

* Corresponding author. Tel.: +418 656 2131x7445; fax: +418 656 3353.
E-mail address: laurent.bazinet@aln.ulaval.ca (L. Bazinet).

0376-7388/$ – see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.memsci.2004.10.048

Due to the presence of one or more electrical charges on the catechins molecule, electrodialysis (ED) would seem to have potential for extracting these compounds from green tea brewing. ED is an electrochemical separation process by which ionic (electrically charged) species are transported, from one solution to another, by crossing one or more perm-selective membranes, under the influence of an electrical current [13]. ED uses an electric field as the driving force and charged membranes to perform the separation: electrodialysis is a combined method of dialysis and electrolysis [14]. By circulating a green tea solution in an electrodialyser, it may be possible to use the driving force of the electrical field to force the catechins to migrate through the membrane out of the main body of the solution.

The objective of this study was to evaluate the feasibility of selectively extracting catechins and caffeine from a green tea solution using an ED cell. We also wanted to determine specifically which commercially available membrane allowed the optimal migration of green tea catechins. The electrodialytic parameters were recorded all along the process, and the concentrations of catechins and caffeine were measured on samples collected to assess the feasibility of the electromigration process.

2. Materials and methods

2.1. Materials

2.1.1. Green tea

The green tea was a non-biological Japanese green tea (lot 12423TKA) obtained from local retailer La Giroflée (Québec City, QC, Canada). The green tea was stored at room temperature in a dark and dry space.

2.1.2. Catechins and caffeine standards

(−)-Epicatechin, (−)-epigallocatechin, (−)-epicatechin gallate, (−)-epigallocatechine gallate, (−)-gallocatechin gallate and caffeine standards were from Sigma Company (Saint-Louis, MO).

2.1.3. Membranes

Three (3) anionic and one (1) ultrafiltration membranes, all commercially available, were selected according to their physico-chemical characteristics (Table 1).

2.2. Methods

2.2.1. Electromigration configuration

The module used was an MP type cell (100 cm^2 of effective electrode surface) manufactured by ElectroCell (Täby, Sweden). The cell consisted of several compartments separated by cationic and tested membranes (Fig. 1). The compartments defined three closed loops containing the solution to be treated (green tea brewing), an aqueous potassium chloride solution (2 g/L KCl) and an electrolyte solution (20 g/L NaCl). Each closed loop was connected to a separate external reservoir to allow continuous recirculation of the solutions. The electrolytes were circulated using three centrifugal pumps, and the flow rates were controlled using flowmeters. The anode, a dimensionally stable electrode (DSA), and the cathode, a 316 stainless-steel electrode, were supplied with the MP cell. The anode/cathode voltage difference was supplied by a variable 0–100 V power source.

2.2.2. Protocol

The membranes were tested under the same conditions, for their capability to allow the migration of catechins. A 20 g of green tea were brewed in 1000 mL of double-distilled water to performed catechin solubilization (according to Wang et al. [15]). Brewing was made at 70 °C in a thermostated water-bath during 40 min, filtered on a Whatman #1 and stored at 4 °C until electromigration treatment was performed. Electromigration was performed in a batch process with a constant current density of 1 A, electrolyte solution volumes of 6 L and a 1.5 L volume of green tea solution. Electromigration was stopped after 1 h of treatment. The initial pH of the green tea brewing varied from 5.6 to 5.8. During each treatment, samples of green tea solution were taken at the beginning of the treatment before applying voltage, every 5 min until 20 min of treatment and thereafter every 10 min. Anode/cathode voltage difference, conductivity and temperature were recorded throughout the process. Concentrations of catechins and caffeine were determined on samples stored at 4 °C by HPLC.

2.2.3. pH

The pH was measured with a pH-meter model SP20 (epoxy gel combination pH electrode, VWR Symphony), produced by Thermo Orion (West Chester, PA).

2.2.4. Conductivity

Conductivity was measured at 4 °C with a YSI conductivimeter (model 3100-115 V, Yellow Springs, OH) and an immersion probe (model 3417, $k = 1\ cm^{-1}$, YSI).

Table 1
Physico-chemical characteristics of the three anionic membranes and the ultrafiltration membrane

	UF-1000 Da	AMX-SB	AFN	PC-400 D
Electrical resistance ($\Omega\ cm^2$)	N/D	2.0–3.5	0.2–1.0	10
Thickness (mm)	0.26–0.28	0.14–0.18	0.15–0.18	0.09–0.11
Burst strength (kg/cm^2)	N/D	4.5–5.5	2.0–4.0	4.0–5.0
Material	Cellulose ester	Polymer of polydivinylbenzene and polystyrene		Nature non communicated
		Minimal reinforcement		Reinforced with polyester