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Enrichment of (−) epigallocatechin gallate (EGCG) from aqueous extract of green tea leaves by hollow fiber microfiltration: Modeling of flux decline and identification of optimum operating conditions

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ABSTRACT

Polyphenols are the important phytochemicals available in green tea leaves and epigallocatechin gallate (EGCG) is the most active one. A scalable method to enhance purity of EGCG from two stage aqueous extract from green tea leaves using hollow fiber microfiltration is reported in this work. Experiments were conducted in the range of 35 to 172 kPa transmembrane pressure drop and cross flow rate 10 to 30 l/h. The flux decline during filtration was quantified using a simple resistance in series model. A first order kinetic model adequately described the growth of fouling layer with time of filtration. A generalized criterion on locus of limiting flux with operating conditions (limiting transmembrane pressure and Reynolds number) was derived and the envelope of optimum operating conditions was identified. Limiting transmembrane pressure was 147 kPa at Reynolds number 282 and the limiting permeate flux at these conditions was 93 $l/m^2 h$. About 80% purity of total polyphenol content and 75% purity of EGCG were attained in the permeate. This work provided a basis for production and purification of green tea polyphenols in industrial scale.

1. Introduction

Tea is one of the most popular beverages around the world due to its taste, aroma and medicinal properties [1]. Three major kinds of tea, namely, black tea (fully fermented), oolong tea (partially fermented) and green tea (unfermented) are produced based on their manufacturing processes. Recently, green tea is receiving considerable attention for specific health benefits due to presence of large amount of caffeine and catechins [2]. Various ethno-pharmacological studies confirm that catechins are the most abundant tea polyphenols present in green tea leaves having antioxidant, anti-cancer, anti-mutagenic and anti-tumorigenic properties [3]. Family of catechins present in green tea leaves include epicatechin (EC), epigallocatechin (EGC), epicatechin gallate (ECG), epigallocatechin gallate (EGCG) and gallocatechin gallate (GCG) contributing to total polyphenol content [4]. Among these, EGCG is the most powerful catechin and it is 200 times stronger than the well-known anti-oxidants, like, vitamin E and C [5]. Thus, extraction and purification of these bioactive components are of great interest to the researchers as well as industries, like, pharmaceuticals, cosmetics and food sectors. Polyphenols are also used as preservative in olive, vegetable oils and other foods rich in fat [6]. Polyphenols are frequently used as UV filter for sunscreen formulation in cosmetic products [7,8]. Additionally, antioxidants, like, polyphenols exhibit antimicrobial properties and are used in bakery products to enhance shelf life [9].

Various techniques are used for extraction of polyphenols from green tea including solvent extraction followed by adsorption [10], microwave assisted solvent extraction [11,12], super critical fluid extraction coupled with adsorption [13]. However, all these processes are based on toxic organic solvents. In this regard, water based extraction of polyphenols from green tea is suitable for pharmaceutical and food applications. High-pressure liquid chromatography (HPLC) can be used to purify polyphenols but this method is of extremely small scale, expensive and time consuming [14]. Hence, bulk production of polyphenols using HPLC is not a viable solution for industrial scale. Some of the emerging technologies for extraction of polyphenols and other antioxidants include ultrasound assisted extraction, pulsed electric field, high voltage electric discharge along with laser ablation, nanotechnology based processes like, nanoemulsion, etc [15]. However, most of these processes are capital and energy intensive, difficult to scale up, having safety issues like, contamination by heavy metals (in case of pulsed electric filed), nanoparticles (in case of nanotechnology assisted processes) and finally consumer acceptability [16].

In this context, membrane based separation can be an alternative due to several advantages, like, low expense, good scalability, excellent

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Nomenclature

A_0	membrane surface area, m^2
J_w	pure water flux, $l/m^2 \cdot h$
k	rate of growth of the fouling layer, s^{-1}
L_p	permeability, m/Pa·s
Q	volume of permeate, m^3
Re	Reynolds number, $\frac{\rho u_0 d}{\mu}$
R_F	fouling resistance, m^{-1}
R_F^s	steady state fouling resistance, m^{-1}
R_M	membrane hydraulic resistance, m^{-1}
t	time, s^{-1}
v_w	permeate flux, $l/m^2 \cdot h$
v_w^o	pure water flux, $l/m^2 \cdot h$
v_w^s	steady state permeate flux, $l/m^2 \cdot h$

Greek symbols

ΔP	transmembrane pressure, kPa
Δt	sampling time, s
μ	viscosity of the permeating solution, Pa·s

Abbreviation

CFR	cross flow rate
BSA	bovine serum albumin
DMF	dimethylformamide
EC	epicatechin
EGC	epigallocatechin
ECG	epicatechin gallate
EGCG	epigallocatechin gallate
GCG	gallocatechin gallate
HPLC	high pressure liquid chromatography
MF	microfiltration
NF	nanofiltration
PVP	polyvinylpyrrolidone
RO	reverse osmosis
SEM	scanning electron microscopy
TMP	transmembrane pressure
TS	total solid
UF	ultrafiltration

separation and high efficiency. Bioactive functional macromolecules, like, pectin, protein, polymeric anthocyanins as well as micromolecules, like, sugar, flavanols, polyphenolic compounds can be separated efficiently using ultrafiltration (UF) membranes having wide cut-off ranging from 1 to 100 kDa [17,18]. Several reports are available for clarification of tea extract using membrane filtration. Microfiltration (MF) was used for removal of imidacloprid and acetamiprid from tea extract in a small laboratory scale batch cell [19]. Removal of 30–50% pectin and 7–20% caffeine was reported by a hybrid membrane process comprising MF followed by ultrafiltration (UF) [20]. Performance between MF and UF for clarification of black tea extract was compared and it was concluded that MF was preferable compared to UF based on quality parameters [21]. MF was used primarily to remove haze from black tea extract [22,23]. Rao et al. [24], studied the use of UF followed by adsorption by fining agents, like, silica gel and chitosan for clarification of green tea extract and showed that the extract had high shelf life at low temperature. Clarification of black tea extract using UF grade ceramic and polymeric membrane was also attempted [25,26]. They reported that UF membrane produced a clarified black tea with increased stability and significantly reduced haze. Zhu and Qin [27] showed that EGCG can be fractionated from other polyphenols in cured tea using cellulose diacetate and b-cyclodextrin (CDA-β-CD) asymmetric membrane (purity of the EGCG was around 87%). Around 80% purity of EGCG was reported by two stage infusion of green tea extracted followed by MF and UF in series [28]. 49% purity of EGCG among the polyphenols was achieved from water extract of green tea using ultrasound assisted UF (20 kDa cut off) preceded by pre-treatment using MF [29]. However, all the above studies were conducted in laboratory scale flat sheet membranes that could not be scaled up directly due to modified flow geometry of spiral wound modules [30]. On the other hand, processing in hollow fiber leads to several advantages including directly scalable design, lower transmembrane pressure drop (hence, energy saving) and higher specific throughput (permeate flux per unit transmembrane drop). It was noted that in majority of above references, microfiltration of tea extract was conducted at fixed operating conditions (transmembrane pressure drop and cross flow velocity). However, effects of operating conditions on limiting flux, solids and polyphenol permeation and flux decline were reported by Argyle and Bird [23]. But in that study, reconstituted black tea liquor in high concentration was used by dissolving black tea powder in water. Therefore, the effects of operating conditions on fresh green tea extract need to be addressed for appropriate scale up and this has been reported in present work using scalable hollow fiber platform. Operating conditions were identified for maximum yield and purity of EGCG from green tea extract. A simple resistance-in-series model was formulated and used for quantifying decline of permeate flux (membrane fouling) as well as identifying the limiting transmembrane pressure drop, thereby, pin-pointing the upper limit of operating conditions.

2. Theory

Flux decline during microfiltration of plant extract is due to development of a fouling layer of rejected solutes on the membrane surface [31–33]. Thus, the resistances encountered by the solvent during its permeation are membrane hydraulic resistance (R_M) and fouling resistance (R_F) assuming the irreversible membrane fouling is not significant due to efficient membrane cleaning which is the case here as described subsequently. The membrane hydraulic resistance is,

$$R_M = \frac{\Delta P}{\mu v_w^0} \tag{1}$$

where ΔP is the transmembrane pressure drop (TMP), μ is the viscosity of the permeating solution and v_w^0 is the pure water flux through the membrane.

The fouling layer resistance at any point of time 't' can be determined from the experimental time history of permeate flux. It can be represented as,

$$R_F = \frac{\Delta P}{\mu(v_w(t))} - R_M \tag{2}$$

Table 1
Specification of hollow fiber spinning unit.

Inner diameter of smaller needle, m	0.0005
Outer diameter of smaller needle, m	0.012
Air gap between extrusion point and gelation bath, m	0.25
Casting temperature, K	300
Pressure in polymer-melt tank, kPa	35
Water flow rate, m^3/s	3.3×10^{-7}
Flow rate of polymer solution, kg/s	5×10^{-5}
Inner diameter of hollow fiber, m	0.0006
Outer diameter of hollow fiber, m	0.0009
Inner diameter of cartridge, m	0.0118
Length of cartridge, m	0.18
Number of hollow fibers	70
Total membrane area, m^2	0.024