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Flax mucilage and barley beta-glucan aerogels obtained using supercritical carbon dioxide: Application as flax lignan carriers

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ABSTRACT

Flax lignan, secoisolariciresinol diglucoside (SDG), has been implicated in the prevention of hormonally related cancers and other diseases. Polysaccharide aerogels, dried using supercritical carbon dioxide ($SCCO_2$), have low densities and high porosities, making them ideal as bioactive carriers. Barley β-glucan (5%) and flax mucilage (10%) hydrogels were first converted to alcogels and then dried using $SCCO_2$, and their characteristics were analyzed. SDG concentrate was incorporated into the gels prior to drying using several approaches. Both aerogels had low densities; however, surface area and % of hydrogel volume of mucilage (201 m^2/g and 57%) were higher compared to those of β-glucan aerogel (166 m^2/g and 38%). When SDG concentrate was added to the hydrated polysaccharide mixtures, regardless of the technique used, SDG contents were similar; however, the impregnation yield was less than 50%. $SCCO_2$-dried polysaccharide aerogels show promise for use as a delivery vehicle for nutraceuticals, including flax SDG.

Industrial relevance: The functional food and nutraceutical industry is looking for ways to enhance the delivery and efficacy of various bioactive ingredients. One approach is to use aerogels as a delivery system. Use of $SCCO_2$ for the preparation of aerogels offers the advantage of eliminating the use of organic solvents and being able to operate at relatively low temperatures to minimize the degradation of bioactives. This study demonstrates one potential application where polysaccharide aerogels are impregnated with flax lignans with the anticipation to increase bioavailability, which requires further testing.

1. Introduction

Flax seed contains lignans, phenolic antioxidant compounds linked to health benefits, especially for alleviating risk factors for cardiovascular disease and cancer (Crosby, 2005; Morris, 2001). The predominant lignan in the flax seed is secoisolariciresinol diglucoside (SDG), which is linked to other phenolic glucosides via ester linkages to form oligomers (Eliasson, Kamal-Eldin, Andersson, & Aman, 2003; Kamal-Eldin et al., 2001). It is assumed that upon ingestion ester linkages are cleaved to release free SDG, from which glucosidic groups are cleaved to release the aglycone form, secoisolariciresinol (SECO). SECO is either absorbed or metabolized by the colonic bacteria to the mammalian lignans, enterodiol and enterolactone (Crosby, 2005).

Flax seed also contains an underutilized but potential high-value ingredient known as mucilage, a soluble fiber, which makes up 10% of seed weight and is found in the outermost layer of the seed coat (Bhatty, 1993; Singer, Taha, Mohamed, Gibriel, & El-Nawawy, 2011). Two heterogenous polysaccharide fractions make up mucilage: a neutral arabinoxylan fraction (75%) and an acidic rhamnogalacturanan fraction (25%) (Wanasundara & Shahidi, 1997; Warrand et al., 2003). Flax mucilage consumption has health benefits of its own, with implications in diabetes and cardiovascular disease management, colon cancer prevention and reduction in the incidence of obesity (Singer et al., 2011). Therefore, enhanced utilization of mucilage is needed to increase its value as a health-benefiting ingredient.

Mucilage also has good water holding capacity and functional properties similar to other common gums as a stabilizer and thickener (Chen, Xu, & Wang, 2006; Mazza & Biliaderis, 1989; Stewart & Mazza, 2000). In addition, flax mucilage is able to form a thermo-reversible cold-set weak gel (Chen, Xu, & Wang, 2006; Cui & Mazza, 1996; Cui, Mazza, & Biliaderis, 1994). Similarly, β-glucan, a soluble fiber component of barley and oats, has been demonstrated to have beneficial functional properties, including gelation (Burkus & Temelli, 1999, 2000).

Delivery systems for bioactive components have been receiving growing interest (Ajazuddin & Saraf, 2010; Champagne & Fustier, 2007; Chen, Remondetto, & Subirade, 2006; de Vos, Faas, Spasojevic, & Sikkema, 2010; Heidarpour et al., 2011; Kailasapathy, 2009). One such delivery form is aerogels dried using supercritical carbon dioxide ($SCCO_2$) and formed using bio-degradable and renewable polysaccharides (Brown, Fryer, Norton, & Bridson, 2010; Chang, Chen, & Jiao, 2010; Comin, Temelli, & Saldaña, 2012a, 2012b; Mehling, Smirnova, Guenther, & Neubert, 2009; Miao et al., 2008; Quignard, Valentin, & Di

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Renzo, 2008; Robitzer & Quignard, 2011; Tsioptsias, Michailof, Stauropoulos, & Panayiotou, 2009), ensuring their potential for edible applications. $SCCO_2$ is a "green" solvent with a tunable solvent power that can be adjusted by changing the temperature and pressure. Due to the disappearance of interfacial tension under supercritical conditions, $SCCO_2$ can be used effectively for the drying of alcogels. Aerogels obtained by $SCCO_2$ drying have a high surface area and porosity and low density, showing great potential for bioactive delivery (Haimer et al., 2010; Mehling et al., 2009).

The main objective of this study was to utilize polysaccharide aerogels for the delivery of flax lignan, SDG. Currently, the main delivery forms of SDG seem to be ground flax seed, defatted flaxseed or flax hulls (Goyal, Sharma, Upadhyay, Gill, & Sihag, 2014), which are mostly sold as capsules. Considering the low oral bioavailability of SDG and SECO in flaxseeds, it is anticipated that using aerogels as a delivery system may potentially contribute to their enhanced bioavailability. The specific objectives were: a) to prepare aerogels from flax mucilage and barley β-glucan, and to compare the resulting aerogel properties, and b) to impregnate the aerogels with lignan concentrate using different approaches and to determine the impregnation yield for SDG.

2. Materials and methods

2.1. Materials

Low-molecular weight (198 kDa) barley β-glucan concentrate (83.3% purity) was obtained at a pilot plant as described by Burkus and Temelli (2006). Whole flax seeds (Agricore United, Winnipeg, MB, Canada) were soaked in distilled water at a ratio of 13:1 and stirred at 85 °C for 3 h. Flax seeds were separated using a sieve, and mucilage was precipitated by adding ethanol to the supernatant at a 1:1 (v:v) ratio. Precipitate was collected and dried overnight in an oven at 100 °C. Dried mucilage was then ground in a coffee grinder (Philips Model HD5112, Markham, ON, Canada) to promote even wetting in future steps. BeneFlax™, a flax lignan concentrate containing no less than 35% SDG, was kindly donated by Archer Daniels Midland Company (Decatur, IL, USA).

2.2. Hydrogel formation

The method of Burkus and Temelli (1999) was used to form 5% (w/v) hydrogels from the β-glucan concentrate. β-Glucan concentrate was added to 50 mL of distilled water with stirring. The mixture was covered with foil to reduce water loss, heated with constant stirring until boiling, and boiled for 5 min. Temperature was then reduced to 75 °C and the mixture was stirred for 1 h. To form 10% flax mucilage gels, flax mucilage was added to 50 mL of boiling distilled water with constant stirring. To assist in evenly hydrating the mucilage, the mixture was then covered, placed in a sonicator, heated to 70 °C and sonicated for 1 h.

The mixtures were then spooned into 10 mL plastic syringes, with the tips removed, covered with parafilm to prevent moisture loss and allowed to set overnight at ambient conditions, in the case of the β-glucan gels, and at 4 °C in the case of the mucilage gels.

2.3. $SCCO_2$ drying of hydrogels

Hydrogels were removed from their molds and cut into cylinders (2 cm long × 1.4 cm diameter). Gels were placed in 50 mL baths, at ambient conditions, with increasing concentrations of 20, 40, 60 and 80% (v/v) ethanol with a residence time in each bath of 1 h, and then in a 50 mL 100% ethanol bath overnight, or 4 × 1 h baths of 50 mL of 100% ethanol followed by a 50 mL bath of 100% ethanol overnight. Alcogels were then measured, weighed and placed into a 25 mL stainless steel cell, with a diameter of 1.4 cm, which was reduced in volume to approximately accommodate twice the length of the alcogel, using a stainless steel filter frit (2 μm) divider. The remainder of the cell was filled with 3 mm glass beads. The cell was placed into a laboratory scale supercritical fluid extraction system (Newport Scientific, Inc., Jessup, MD, USA), described previously for the extraction of lignans from flax seed (Comin, Temelli, & Saldaña, 2010). CO_2 of 99.95% purity (Praxair, Edmonton, AB, Canada) was pressurized to 15 MPa using a diaphragm compressor. The extraction vessel was heated to 40 °C with a heating jacket. Once the desired pressure and temperature had been reached, the conditions were held for 1 h, at which point CO_2 flow was started at 1 L/min (measured at ambient conditions) and maintained for 4 h. Upon depressurization, ethanol was collected in vials and CO_2 was vented. After 4 h, the system was depressurized at a maximum rate of 1 L/min, until tank pressure of 6.2 MPa was reached, and then CO_2 flow rate was increased to minimize depressurization time. Samples were held in zipper lock sealed bags overnight, to allow any CO_2 inside the aerogels to dissipate, and then they were measured and weighed.

2.4. Incorporation of flax lignans

BeneFlax was added to the gels using four different methods. The first two methods consisted of adding 1 g of BeneFlax to 50 mL of mixture before (BH) or after hydrating (AH) the polysaccharide powders. For the third method, a solution of 1 g of BeneFlax in 70% ethanol was prepared, followed by the addition of β-glucan or flax mucilage, and evaporation of the solvent at 70 °C at ambient pressure. Distilled water was then added to the mixture to rehydrate to a volume of 50 mL (EtOH-BH). For the fourth method, pre-made hydrogels were soaked in 4 × 50 mL solutions of 1 g of BeneFlax in 50 mL 70% ethanol for 1 h each, followed by 50 mL of 100% ethanol overnight (EtOH-AG). All experiments were performed in triplicate.

2.5. Determination of SDG loading in $SCCO_2$

A 5 g sample of BeneFlax was loaded into a 25 mL basket, and placed in the supercritical system, as described above. The cell was pressurized with CO_2 to 15 MPa and the temperature was held at 40 °C. Where ethanol was used as a co-solvent, 0.5 mL/min ethanol was pumped (Model 1330, Bio-Rad Laboratories Ltd., Mississauga, ON, Canada) into the pressurized CO_2 stream. The flow rate of the CO_2 was 1 L/min, measured at ambient conditions. Glass collection vials were connected to the depressurization valve and held in a refrigerated bath (−20 °C). Extract fractions were collected at different time intervals to determine SDG loading of $SCCO_2$. Ethanol was removed from the extracts by purging gently with nitrogen gas. Sample weights were determined gravimetrically (±0.0001 g). Extract samples were stored at −20 °C until ready to be analyzed by HPLC. All experiments were done in triplicate.

2.6. HPLC analysis

Aerogels were cut into quarters, crushed gently, added to 10 mL of 70% ethanol, and heated for 3.5 h in a 65 °C shaking water bath. Samples were then cooled to ambient temperature, centrifuged and 2 mL aliquots were transferred to HPLC vials to determine if any free SDG was present in the aerogels. Aliquots of the aerogel extracts (5 mL) were added to 5 mL of 2 N NaOH in 70% ethanol, and placed back into the shaking water bath for an additional 3.5 h at 60 °C. Samples were then neutralized using concentrated glacial acetic acid, centrifuged (1250 ×g), and 2 mL aliquots were added to HPLC vials. Powders of BeneFlax, β-glucan concentrate and flax mucilage and extracts from $SCCO_2$ loading determination were treated in the same manner as above, and concentrated or diluted as required.

HPLC analysis was carried out according to Comin et al. (2010) with some modifications. The column was a C 18–5 μm (4.6 mm × 150 mm) reverse phase column (Supelco, Sigma-Aldrich, St. Louis, MO, USA) and