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Fucoxanthin-rich oil encapsulation using biodegradable polyethylene glycol and particles from gas-saturated solutions technique

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ABSTRACT

Fucoxanthin is a precious marine carotenoid with multiple bioactivities such as antioxidant, anti-obesity, and anti-cancer effects. Despite its valuable properties, fucoxanthin possesses high sensitivity when exposed to aerial, thermal, or light factors. Our study aims to extract fucoxanthin-rich oil (FO) from *Saccharina japonica* using supercritical carbon dioxide with sunflower oil (SFO) as co-solvent and encapsulate the extracted oil using particles from gas-saturated solutions (PGSS) process with polyethylene glycol (PEG) as a biodegradable coating. The encapsulation efficiency (EE) of the extracted oil based on fucoxanthin content was optimized using response surface methodology with Box-Behnken design (BBD). Three independent factors, including mixing ratio of FO and PEG, temperature, and pressure, were optimized. The encapsulated particles (EP) at optimized conditions were characterized for their powder properties and storage stability. FO and EP were measured in antioxidant activities and fatty acids content. The optimum EE was approximately 82%, implying the capability of PGSS and PEG in encapsulating nutritious oil.

1. Introduction

Fucoxanthin is a marine carotenoid abundantly recovered from brown algae (*Phaeophyceae*), for instance, *Laminariaceae*, *Sargassaceae*, and *Alariaceae* families [1]. In aid of fucoxanthin colorant property, brown algae have long been consumed as a natural pigment in various traditional Asian dishes. Presently, fucoxanthin has been selling on functional food market as a natural anti-obesity nutrient. A study on the anti-obesity behavior of fucoxanthin revealed that a daily intake of fucoxanthin can induce uncoupling protein 1 expression in abdominal white adipose tissue (WAT), leading to the reduction of body fat weight [2]. Consequently, this effect helps to regulate the cytokine secretions of WAT, leading to the improvement of insulin resistance and decreasing of blood glucose levels, hence preventing diabetes [3]. Furthermore, several biological benefits of fucoxanthin were also discovered. Kumar et al. [4] and Lopes-Costa et al. [5] successfully determined anti-cancer effects of fucoxanthin in colon cells, meanwhile, its antioxidant activity was reported by Fung et al. [6] when studying on *Undaria pinnatifida* extracts. In evaluation with other types of carotenoids, such as astaxanthin and β-carotenes, fucoxanthin inhibition on colon cancer cell viability is proved to be higher [7]. Likewise, Hosokawa et al. [8] explored higher antioxidant activities of fucoxanthin when compare with α-tocopherol, a typical antioxidant source in diet supplements. Those advantageous benefits of fucoxanthin might be accounted to its unusual allenic carbon, and several functional groups on its carbon skeleton structure, such as monoepoxide, hydroxyl, carbonyl, and carboxyl moieties [9].

The main challenge when producing and preserving fucoxanthin products is its feeble stability against light, aerial exposure, and thermal processing. A study on the stability behaviors of fucoxanthin in canola oil [10] proved that the presence of light, air, and heat, either individually or synergistically, promoted substantial degradation and transformation of total, all-*trans* and 9′-*cis* fucoxanthin into its isomers such as 13-*cis* and 13′-*cis* fucoxanthin. Therefore, suitable methods to protect fucoxanthin products are needed to overcome this limitation.

Encapsulation has progressively developed in the food and pharmaceutical industry [11]. The goal of the process is to incorporate a food ingredient within a suitable coating material, and thus, preserving the bioactive compounds from extreme external factors such as temperature, humidity, and oxidation. Conventional encapsulation processes, for example, spray drying, fluidized bed coating, coacervation, and emulsification are widely used in manufacturer scales [11,12]. However, these techniques either required high thermal operations or involved organic solvents that, for safety requirements, need additional processes to be removed from the final products.

To date, supercritical carbon dioxide ($ScCO_2$) has become a

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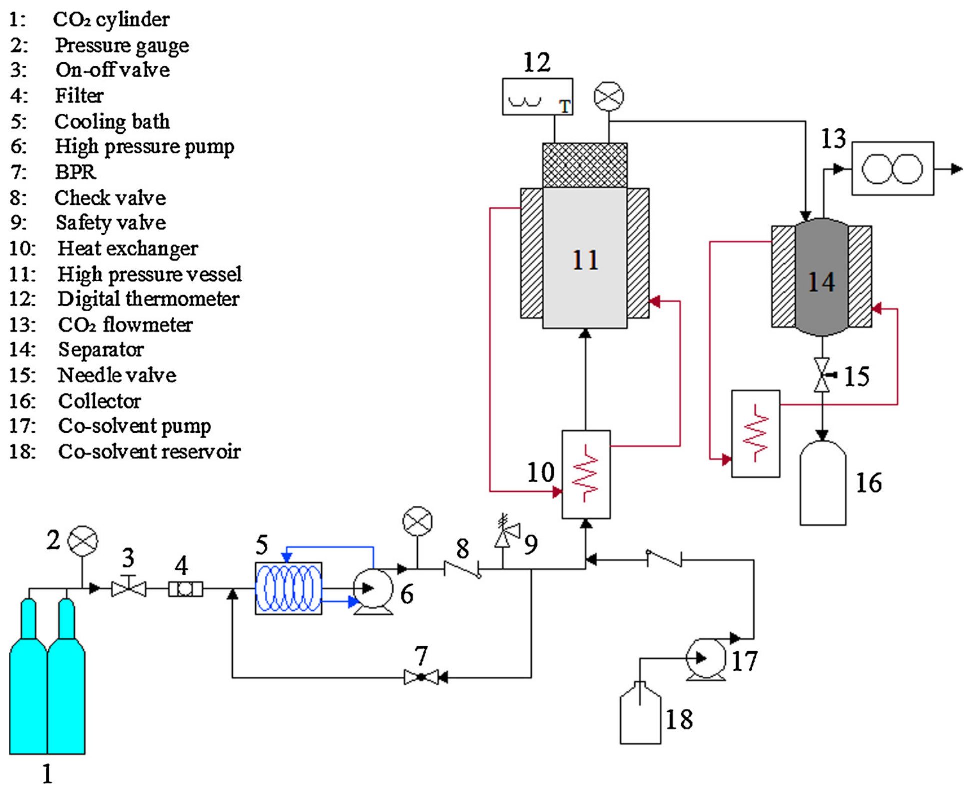

Fig. 1. Schematic diagram of $ScCO_2$ extraction system.

prominent green technique in terms of chemical synthesis [13], nutrient recovery [14], and particles formation [15]. Based on its neutral and non-toxic properties, low viscosity but high diffusivity and solubility, tunable polarity, and easy removability, $ScCO_2$ can play a role of a powerful solvent in extracting key compounds that consequently help to alternate the use of harmful organic solvents. In addition, the low operational temperature is a great advantage of $ScCO_2$ in dealing with volatile and heat-sensitive compounds such as carotenoids, essential oils, and vitamins [16–18].

Fucoxanthin and several functional components such as fatty acids and fucosterol can be recovered together in the lipid fraction of the seaweed [19]. However, the total lipid content in brown seaweed is quite low (less than 7%) [20]. Recently, vegetable oils, with a large number of fatty acids, mono- and diglycerides [21], were employed to enhance the extraction yield of carotenoids from raw materials. For example, soybean oil and olive oil were used to carry astaxanthin [22], canola oil to carry carotenes [23], and hazelnut oil to carry lycopene from tomato extracts [24]. Besides, vegetable oils were also reported to induce the stability of carotenoids [25]. Therefore, the co-extraction of vegetable oil and fucoxanthin can be a novel approach in enriching this marine nutrition.

Particles from gas-saturated solutions (PGSS) is an application of $ScCO_2$ in term of particles [26], composites [27], or encapsulates formation [28]. Under the supercritical condition, the mixture of nutrients and coating substances gets saturated with carbon dioxide (CO_2). An instant depressurization of the CO_2-saturated solutions through a micro-nozzle can generate a sudden cooling effect (Joule-Thomson effect), allow the coating and core materials to be co-precipitated, and thereby leading to the encapsulation of key compounds. Another advantage of PGSS is the ability of $ScCO_2$ to reducing the melting point of the coating polymer [29], hence decrease working temperature to a middle level, which is more suitable for heat-sensitive nutrients like fucoxanthin.

Polyethylene glycol (PEG) has long been engaged as a polymeric carrier for drug delivery systems [30]. Based on a durable shielding effect, PEG can prolong stealth effect of the drug as well as its storage stability. Furthermore, with a relatively hydrophilic property, PEG also enhance the solubility of the drug powder. On the other hand, the solubility of CO_2 in PEG increases proportionally with pressure. In the pressure range 20–250 bar and temperature range 50–80 °C, the solubility of CO_2 in PEG is independent with the polymer molar mass and can gain up to 25% gas weight [31]. Moreover, the melting point of PEG was also observed reducing under $ScCO_2$ condition in the same study. Therefore, the polymer has been applied in numerous studies about the encapsulation of food and medical key compounds using PGSS [32].

The aim of our study is to employ PEG as the coating material in the encapsulation of fucoxanthin-rich oil (FO) extracted from *Saccharina japonica* with PGSS technique. The FO was extracted using a lab-scale $ScCO_2$ extraction system with sunflower oil (SFO) as co-solvent. Single factor analysis (SFA) and response surface methodology (RSM) with Box-Behnken design (BBD) were applied to design the encapsulation experiments, concerning three independent variables including mixing ratio, temperature, and pressure. The encapsulation efficiency (EE) based on fucoxanthin content was the principle response variable. The FO was characterized in terms of fucoxanthin content and fatty acid composition. The encapsulated particles (EP) at optimized conditions were characterized for particle size analysis (PSA), morphology, and physical properties. Together, FO and EP were tested for antioxidant activities and storage stability.

2. Materials and methods

2.1. Sample preparation

S. japonica (sundried sample) was purchased from Gijang market (Ilgwang-myeon, Gijang-gun, Busan, Republic of Korea), in April 2017. The dried seaweed was firmly ground and sieved using a 710-µm stainless steel sieve. The ground seaweed powder was stored under −40 °C prior to extraction.