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Concurrent extraction of oil from roasted coffee (*Coffea arabica*) and fucoxanthin from brown seaweed (*Saccharina japonica*) using supercritical carbon dioxide

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ABSTRACT

The present study aimed to extract carotenoid from brown seaweed, *Saccharina japonica* (SJ) and oil from roasted coffee (RC) concurrently using supercritical carbon dioxide and evaluate its characteristics. Response surface methodology was employed to determine the optimum conditions for the extraction of oil with maximum carotenoid content. The kinetics of the extraction process was studied by fitting a curve for experimental data and comparing it with two previously proposed models. The fitted curve showed good agreement with mass transfer model. The global yield and oil recovery were 3.51 and 87.31% respectively. The fatty acid analysis showed a good balance between the saturated and polyunsaturated fatty acids. The fucoxanthin and total phenolic content of the oil were 2.08 ± 0.06 mg/g and 287.71 ± 13 mg/100 g, respectively. The oil also showed high antioxidant activity, oxidative stability, and better flavor profile as compared to SJ oil. Therefore, the current study demonstrated the extraction of a special oil concurrently from marine and terrestrial resources with a potential for application in food and related industries.

1. Introduction

Recently, special oils containing bioactive compounds extracted from different combinations of vegetables have attracted huge interest from various industries. These oils could be used as an ingredient in nutraceutical, pharmaceutical and cosmetics formulations and as well as a direct addition in foods [1].

Brown seaweeds are the most common edible seaweeds in China, Japan, and Korea. *Saccharina japonica* (SJ) is one of highly cultivated brown seaweed in South Korea. The amount of SJ produced by farming in 2013 amounted to 373,264 t (wet weight); an additional 4 t (wet weight) were collected from natural populations in Korea [2]. *S. japonica* lipids are rich in highly unsaturated fatty acids such as α-linolenic acid, arachidonic acid, and eicosapentaenoic acid (EPA) [3]. SJ lipids contain interesting phytochemicals such as fucoxanthin (FX), fucosterol and polyphenols [4]. FX is an orange colored carotenoid present in edible brown seaweeds, including SJ [5].

Several biological activities of FX and its metabolites have been reported including; antioxidant activity [6], anticancer activity [7] anti-obesity effect [8] neuroprotective activity [9]. However, seaweeds also have characteristic off-flavor which is the major obstacles for versatile applications in the food and cosmetics industries [10]. Masking or removing the off flavor could a possible strategy to these obstacles.

Coffee is the most traded commodity next to petroleum [11]. Coffee contains up to 17% oil depending on its origin and species it also has several compounds with known biological activity. The roasting process of coffee beans produces characteristic coffee flavor and the brown-colored compounds called melanoidins [12,13]. Melanoidins have been proved to possess antioxidant capacity and metal-chelating properties [14].

Extraction of bioactive compounds including carotenoids has been conducted using organic solvents. However, this technique has several disadvantages such as usage of a large amount of organic solvent, consuming too much time and extra purification step to separate the solvent. In addition, this method is not suitable for extraction of chemical, oxygen, and light sensitive substances such as carotenoids [15]. In order to mitigate the afore mentioned problems, extraction based on

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supercritical carbon dioxide (Sc-CO_2) has got popularity as carbon dioxide is a readily available, cheap, environmentally friendly solvent [16,17]. The low critical pressure (73.8 bar) and temperature (31.1 °C) of carbon dioxide is also another advantage for extraction of heat-sensitive substances [18]. Moreover, Sc-CO_2 solvent property can be adjusted by changing pressure and temperature to extract oil [19]. The use of co-solvents such as vegetable oils with Sc-CO_2 has been proved to be efficient mechanism to extract carotenoids [20–22].

Therefore, the aim of the present study was to extract a special oil rich in FX from SJ and RC using supercritical carbon dioxide with simultaneously extracted coffee oil acting as co-solvent. The study also investigated the kinetics of the extraction process. Moreover, the extracted oil was also evaluated for antioxidant activity, fatty acid and flavor compounds analysis, storage stability, and thermal and optical properties.

2. Experimental

2.1. Materials

Green coffee beans, *Coffea arabica*, from Ethiopia, (Yirgacheffe grade 1) was supplied by Global Soft Commodities GSC International Coffee®, Seoul, Republic of Korea. The brown seaweed (*S. japonica-(J.E. Areschoug) C.E. Lane, C. Mayes, Druehl et G.W. Saunders)*) was collected from Geumil-eup, Wando-gun, Jeollanamdo, Republic of Korea. Carbon dioxide (99.99% pure) was supplied by Kosem (Yangsan, Korea). All other chemicals used in the study were of HPLC or analytical grade.

2.2. Sample preparation

The SJ was freeze dried and crashed with mechanical blender (PN SMKA-4000 mixer, PN Co., LTD, Republic of Korea) and sieved using a 710-μm stainless steel sieve. Then the powder was placed in a zip-lock plastic bag and kept in a refrigerator at −20 °C until needed for analysis [23].

Green coffee was medium roasted using home scale roaster (Heathware Precision Coffee Roaster, Wheeling, IL, USA) at 220 °C for 900 s then the roasted coffee was finely ground by mechanical and sieved as indicated above. Sample that passed though the sieving mesh was immediately used for extraction of oil to protect the possible loss of flavor upon storage [24].

2.3. Methods

2.3.1. Selection of extraction conditions

First, response surface methodology (RSM) was used to evaluate the effect of independent variables, temperature (40-50 °C), pressure (200-300 bar) and mixing ratio of SJ to RC (27–75%) on the response, FX content of the oil. Then the combinations of the independent variables which maximize the FX content in the oil was further studied to investigate the nature of extraction kinetics. The RSM was conducted using central composite design (CCD) with three numeric factors at three levels. The design consists of a total of 13 runs with triplicate analysis at the central point to minimize the error [25]. Table 1 shows the true and coded values of the independent variables. The experimentally obtained data were fitted to the second order polynomial equation (Eq. (1)). Such equations can sufficiently describe the relationship between the independent variables and the response of interest [25].

$$Y = \beta_0 + \sum_{i=0}^{3} \beta_i X_i + \sum_{i=0}^{3} \beta_{ii} X_i + \sum \sum_{i<j=1}^{3} \beta_{ii} X_i X_j \quad (1)$$

where Y represents the response, Xi and Xj are the independent variables affecting the response, and β_0, β_i, β_{ii}, and β_{ij} are the regression coefficients for intercept, linear, quadratic and interaction terms respectively.

Table 1
Central composite design showing the combination of coded and real values of the independent variables, the predicted, and actual values of the response.

Run	Temperature (°C)	Pressure (bar)	SJ to RC Ratio (%)	Fucoxanthin (mg/g)	
				Predicted	Actual
1	50 (1)	300 (1)	25 (−1)	0.42	0.42
2	50 (1)	200 (−1)	75 (1)	1.93	1.93
3	40 (−1)	300 (1)	75 (1)	2.15	2.15
4	40 (−1)	200 (−1)	25 (−1)	0.48	0.48
5	40 (−1)	250 (0)	50 (0)	0.86	0.85
6	50 (1)	250 (0)	50 (0)	0.94	0.93
7	45 (0)	200 (−1)	50 (0)	0.92	0.91
8	45 (0)	300 (1)	50 (0)	0.92	0.91
9	45 (0)	250 (0)	25 (−1)	0.48	0.47
10	45 (0)	250 (0)	75 (1)	1.84	1.83
11	45 (0)	250 (0)	50 (0)	0.86	0.86
12	45 (0)	250 (0)	50 (0)	0.86	0.88
13	45 (0)	250 (0)	50 (0)	0.86	0.87

The best oil extraction conditions which maximize the FX content were selected using desirability function [26] with the help of Design-Expert software (Design-Expert v.7 Stat-Ease, Minneapolis, Minnesota, USA).

2.3.2. Oil extraction

The sample was filled into the reactor according to experimental design stated in Section 2.3.1. Fig. S1 shows the pattern of sample filling and the direction of the solvent flow. The extraction was conducted in an extraction vessel whose schematic diagram and description have been indicated in our previous study [27]. For constructing the extraction curve and modeling the kinetics of the extraction process, the optimum extraction condition predicted by the RSM was used. The CO_2 flow rate and the extraction time were kept constant at 27 g/min and 3 h respectively for all extraction processes.

2.3.3. Global yield and recovery of oil

The global yield was evaluated using a ratio of the mass of oil and mass of the solid raw material subjected to extraction (Eq. (2)). The oil recovery yield was calculated by the ratio of global yield and initial oil content obtained from soxhlet extraction using n-hexane for 16 h as per Eq. (3) [28].

$$\text{Global Yield (\%)} = \left(\frac{M_{extract}}{M_{initial}}\right) \times 100 \quad (2)$$

where $M_{extract}$ and $M_{initial}$ are the mass of extracted oil and the raw material used for extracting oil respectively.

$$\text{Recovery (\%)} = \left(\frac{\text{Global Yield}}{\text{Soxhlet yield}}\right) \times 100 \quad (3)$$

2.3.4. Extraction curve and modeling

To explain the nature of the extraction curve and to compare experimental results with model predictions, we have reviewed and selected two models from two previously published studies. The empirical model reported by Naik et al. [29] and the mass transfer model reported by Handayani et al. [30] were used in this study.

Naik et al. [29] empirical model defined as follows (Eq. (4)) :

$$Y = \frac{Y_\infty t}{b + t} \quad (4)$$

where Y, represents the yield (%) at any time t (min); b is the modifiable parameter of the model, Y_∞ is the total extractable oil in the sample at an infinite time.

The Handayani et al. [30] model defined as follows (Eq. (5)):