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Original Research Paper

Comparing yields from the extraction of different citrus peels and spray drying of the extracts

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ABSTRACT

Citrus peel has been reported to contain bioactive compounds, such as phenolic compounds, ascorbic acid and carotenoids. A comparison of the quality and quantity of materials produced by extraction and spray drying of different citrus peels (orange, lemon, lime, and mandarin) has been conducted. The average total phenolic contents (TPC) of all citrus peel extracts were between 4.9 and 6.9 mg gallic acid equivalent (GAE)/g fresh weight (FW) citrus peels. Lime peel extract showed the highest antioxidant content (TPC of 6.9 mg GAE/g FW peel and SC_{50} of 740 μg/mL) and the lowest TPC recovery after spray drying (84%) compared with other types of peel extract. Regarding the yield (or solids recovery) from spray drying, lemon and mandarin peel extracts were found to be the most difficult to spray dry (yields/recoveries of 78% and 73%, respectively). The differences in composition, such as citric acid and sugars contents, may explain some of the differences between the spray drying yields of the extracts.

1. Introduction

Citrus residues, with a world production estimation of 15×10^6 tons/year, have been described as an underutilized material [1]. Citrus residues are often disposed as waste, which may cause major environmental problems due to their high associated chemical and biological oxygen demand (COD and BOD) [2]. Therefore, the processing of citrus residues is expected to minimize the environmental impact and to add value to this residue. Citrus peel, which accounts for almost one-fourth of the whole fruit mass, may be beneficial for pharmaceutical and food industries due to the high content of bioactive materials [1,3].

Orange, mandarin, lemon, and lime are the main types of citrus fruit grown in the world. These citrus fruits represent 94% of the total production of citrus fruits in Australia, with orange as the citrus fruit that makes up the largest percentage of production (78%) [4]. The presence of bioactive compounds, such as phenolic compounds, ascorbic acid and carotenoids, in citrus peels has been reported before [5]. These compounds are naturally occurring antioxidants which have the ability to scavenge reactive oxygen and nitrogen species and prevent oxidative damage to important biological macromolecules [6].

In the manufacture of these nutraceutical materials, the initial process required is the extraction of citrus peel using a suitable solvent. One of the common traditional extraction techniques is Soxhlet extraction. The Soxhlet extraction method is relatively simple, inexpensive, and also provides repeated contact of samples with fresh solvents, which helps the transfer equilibrium [7]. However, the process is time-consuming, which motivates the development of other techniques. Several modern extraction techniques, such as ultrasound and microwave techniques, for producing phenolic compounds from citrus peels have been reported previously [3,8].

After solvent extraction, the liquid extracts can be dried in order to lower storage and transportation costs, and also produce materials with better storage stability [9]. Spray drying is a commonly used method to convert feed from a liquid state into a powder form [10]. In the spray-drying process, the water of the feed solution is removed very quickly. Previous studies reported that spray drying might be suitable for the drying of heat-sensitive materials, such as phenolic compounds in citrus peel extracts [10,11].

In a previous study, the TPC recovery during spray drying of one type of citrus peel extract (orange peel extract) has been reported [11]. Several authors have also previously reported the antioxidant contents of different types of citrus peel extracts [3,5,12–14]. However, the comparison of yield (solid recovery) and TPC recovery in the spray drying of different citrus peels has not been evaluated so far. To complement those previous studies, a comparison between

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the performance of combined extraction and spray drying for different citrus peels has been made. The amounts of TPC and antioxidant activity using the DPPH (2,2-diphenyl-1-picrylhydrazyl) test method in the materials were measured. The spray drying yields (solid recovery) and TPC recovery of different citrus peel extracts have also been compared.

2. Materials and methods

2.1. Chemicals

Gallic acid (G7384), sodium carbonate (S2127), Folin Ciocalteu's reagent (FCR) (F9252), DPPH (2,2-diphenyl-1-picrylhydrazyl), and ethanol used in this study were analytical grade and were obtained from Sigma Aldrich. Whey Protein Isolate, or WPI, (Balance Ultimate Body Performance, Vitaco Health NZ Limited) was purchased from a local store.

2.2. Solvent extraction

Four types of citrus peel were used in this study: Navel orange, mandarin, lemon, and lime. Citrus fruits were obtained daily from the local supermarket (Coles Broadway, Sydney). The fresh citrus fruits were first washed, surface dried, and then peeled. The citrus peels were then cut into fine pieces with a Homemaker mini food processor (Model No. CH-1106). Prepared citrus peels were stored in a refrigerator at 4 °C until extraction that the same day.

The extraction was carried out in a Soxhlet extraction unit using deionized (DI) water as the solvent. The results of a preliminary extraction experiment suggested that there was no significant effect of solvent to solid ratio on the yield of phenolic compounds from Soxhlet extraction, so a low solvent to solid ratio was preferable to minimize the use of solvent. However, Soxhlet extraction has limitations, as a relatively large amount of solvent is required during the process [7]. For these reasons, the amounts of citrus peel and deionized water used in each extraction in this study were 20 g and 280 mL, respectively, resulting in a solvent to solid ratio of 14.

A preliminary experiment was also conducted in order to determine the extraction time required to achieve the equilibrium concentration. After the extraction was completed, the liquid extract was cooled down and then stored in the refrigerator prior to the determination of the TPCm and antioxidant activity.

2.3. Spray drying

The aqueous citrus peel extracts, with an addition of WPI (5% of the total solids concentration in the extracts), were dried using a Buchi B-290 mini spray dryer. The atomization air flow rate was controlled by using a rotameter (Q flow by Vogtlin, Switzerland). The operating conditions were shown in Table 1, which were determined to provide a peak yield, as reported in a previous study [11]. The yield (or recovery) from spray drying has been calculated using Eq. (1). The total solid content of the feed was determined by drying in a fan circulated oven (Thermoline Scientific, Australia) at 85 °C for 24 h.

$$\text{Yield } (\%) = \frac{\text{mass of solid in the collecting vessel}}{\text{mass of solid in the feed}} \times 100\% \quad (1)$$

Table 1
Spray drying conditions used in this study.

Variable	Conditions
Drying air inlet temperature	125 °C
Atomization air flow rate	601 L/h (50 mm on rotameter)
Liquid feed pump rate	4 mL/min (10% of maximum value)
Main drying air flow rate	38 m^3/h (100%, maximum)
Volume of extracts	Approximately 250 mL
Spray drying duration	Approximately 60 min
Outlet temperature	60 ± 1.6 °C
Moisture content of the powder	0.07 ± 0.01 g/g dry material

2.4. Determination of total phenolic content (TPC)

The determination of TPCs of the extract and powder was conducted using the procedure described by Singleton et al. [15]. Sample solution (0.1 mL) was first transferred to a container, followed by the addition of 7.9 mL of DI water and 0.5 mL of Folin Ciocalteu's reagent. Na_2CO_3 (1.5 mL; 20% w/w) was then added into the same container to make a total volume of 10 mL. The container s kept in the dark for two hours and the reaction allowed to proceed at a basic pH. The absorbance was read using a UV–Vis spectrophotometer (Cary 50 by Agilent Technologies, U.S.A.) at a wavelength of 765 nm. Gallic acid solution was used as positive control, and the TPC was expressed as mg GAE/g of material.

2.5. Antioxidant activity

The antioxidant activity of the material was determined using the DPPH method, as proposed by Brand-Williams et al. [16]. Equal volumes (total volumes of 4 mL) of the DPPH/ethanol solution and of the peel extract solutions (at different concentrations, which are shown in Table 2) were used. The mixtures were then incubated at room temperature. After 30 min, the absorbances of the mixtures were recorded using a UV–Vis spectrophotometer (Cary 50, Agilent Technologies, U.S.A.) at a wavelength of 517 nm. The free radical scavenging activity of the extracts was calculated using Eq. (2):

$$\text{Radical scavenging activity } (\%) = \frac{\text{Abs control} - \text{Abs sample}}{\text{Abs control}} \times 100 \quad (2)$$

where Abs control and Abs sample are the absorbance of the DPPH/ethanol solution and the actual sample at 517 nm, respectively. The percentage scavenging activity was plotted as a function of sample concentration, and the concentration of sample required for 50% scavenging activity (SC_{50}) was determined by interpolation.

2.6. Statistics

Data in this study were obtained from a minimum of four independent experiments, and are presented as mean values with standard deviations. For statistical analysis, differences were tested for significance by using analysis of variance (ANOVA) with a significance level of $P \leqslant 0.05$. To further assess the difference between group mean values, the Fisher's Least Significant Difference (LSD) test was performed. This protected LSD test was only performed if the ANOVA test showed a significant difference between groups.

Table 2
Range of concentration used in DPPH test method.

Type of peel extract	Range of concentration tested (μg/mL)
Lime peel extract	600–1500
Navel orange peel extract	1000–1500
Lemon peel extract	675–1250
Mandarin peel extract	580–1750

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