The Journal of Supercritical Fluids 128 (2017) 208–217

Contents lists available at ScienceDirect

The Journal of Supercritical Fluids

journal homepage: www.elsevier.com/locate/supflu

Impregnation of mango leaf extract into a polyester textile using supercritical carbon dioxide

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ARTICLE INFO

Keywords:
Mango leaves
Supercritical fluid impregnation
Polyphenols
Mangiferin
Antioxidant activity
Antimicrobial activity

ABSTRACT

A mango leaf extract (MLE) has been used in conjunction with supercritical CO_2 to impregnate a polyester textile. MLE was produced using CO_2/methanol (50%) at 120 bar and 100 °C. The extract presented antioxidant (3.28 ± 0.1 μg DPP/μg extract), bacteriostatic (4.53 mg/mL) and bactericidal (7.24 mg/mL) activities. The effects of impregnation conditions, i.e., depressurization (0.6, 1.1 and 25 bar/min), temperature (35 and 55 °C) and pressure (400 and 500 bar), were analyzed. The most favorable conditions for the total polyphenols loading were obtained at 500 bar, 55 °C and 25 bar/min. Under these conditions the highest bacterial growth inhibition was observed with the impregnated polyester; however, the highest antioxidant capacity (AAI = 4.04 ± 0.16) was observed on using the same pressure but a low temperature (35 °C). The effect that pressure and temperature had on the impregnation efficiency of the major impregnated polyphenols was also studied.

1. Introduction

Textiles are mainly used to cover the human body and they offer protection against adverse conditions. However, market and consumer needs are pushing the textile industry to be ever more innovative and efficient, which in turn leads to cutting edge R & D programs, the production of highly functional products and the promotion of strategic partnerships. In this context, functional fabrics are one of the most advanced developments as they represent materials with new properties and added value [1].

For example, Silva et al. [2] studied the impregnation of microcapsules that contained limonene into a wool/polyester fabric. The impregnation process was carried out in several steps and temperatures of 100 °C were employed during drying. In the first stage, the textile was immersed in the bath and then passed through a two-roller foulard for the impregnation step. The textile material was subsequently processed in a thermofixation oven, where drying took place (100 °C), while microcapsule fixation was achieved by thermal crosslinking (140 °C). Silver has several effects on microorganisms, including impeding the electron transport system and preventing DNA replication [3]. Processes such as master batch impregnation [4], layer-by-layer deposition [5], RF-plasma-mediated deposition and vacuum-UV [6] and sonochemical [7] coating have previously been used to fabricate textiles that contain antimicrobial metals. These impregnation methods also require high temperatures.

Supercritical Solvent Impregnation (SSI) is a 'green' technology that does not produce any harmful byproducts and can represent an efficient and advantageous alternative to traditional impregnation methods. As mentioned above, the more commonly used impregnation processes generally require high processing temperatures, which can deteriorate thermosensitive active compounds, or involve the use of organic solvents, which must then be removed through numerous drying or evaporation steps. One of the main advantages of the SSI technique is that the solute loading and depth of impregnation can be tuned by changing process conditions. This possibility gives rise to final products that are free from organic solvent residues since supercritical CO_2 is released as a gas after depressurization. Finally, the technique allows the process to be carried out under relatively mild conditions in an oxygen free environment, which is often desirable when the objective is to impregnate natural-based compounds with biological activity [8,9].

The SSI technique has recently been used in several fields within the chemical industry. Most common examples involve wood impregnation [8–11], supercritical aerogel impregnation [12–14], polymer dyeing [15–17] and drug-loaded implants [18–22]. Bach et al. and, more recently, Banchero reviewed the studies in which supercritical CO_2 impregnation processes were applied to the dyeing of synthetic and natural textiles [23,24]. The reviews mainly focus on the dyeing of polyester textile (PET) and they provide an interesting overview of the phenomena that occur during this process. However, the supercritical CO_2 dyeing process differs from the supercritical CO_2-assisted

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http://dx.doi.org/10.1016/j.supflu.2017.05.033
Received 31 March 2017; Received in revised form 31 May 2017; Accepted 31 May 2017
Available online 02 June 2017

impregnation of bioactive compounds in terms of experimental conditions and the kind of material that can be impregnated [18].

Mangifera indica L. is a medicinal plant that belongs to the *Anacardiaceae* family and it is native to Southeast Asia and is widely distributed in tropical regions. Mango is the third most popular tropical fruit worldwide and its production generates considerable agro-industrial wastes, such as peel, kernel seeds, leaves, and stem bark derived from pruning activity and industrial fruit processing. The results of numerous studies have shown that mango leaves are an important source of polyphenols with potent antioxidant and pharmaceutical properties [25,26]. Mangiferin is one of the predominant polyphenols in mango leaves and it has multiple pharmaceutical properties such as antioxidant, antibacterial, antifungal, antidiabetic, immunomodulatory, anti-inflammatory, and analgesic properties, with potential uses in the treatment or prevention of chronic diseases including cancer and neurodegenerative and cardiovascular diseases [25–29]. Other valuable phenolic compounds with interesting pharmaceutical properties, such as quercetin, gallic acid, gallotannins, and iriflophenones, have also been identified in mango leaves [30–32]. The high content of potent antioxidant polyphenols is the reason for the great potential of mango leaf extracts in cosmetic, nutraceutical, pharmaceutical or food applications.

In previous studies, the authors investigated the recovery of active compounds from mango leaves by different high-pressure techniques such as Supercritical Fluid Extraction (SFE), Pressurized Liquid Extraction (PLE) and Enhanced Solvent Extraction (ESE) [31,33,34]. However, the use of supercritical CO_2 processing to impregnate textiles with a natural extract from mango leaves has not been investigated previously.

The main objective of this study was to evaluate the supercritical fluid impregnation of a polyester textile with a mango leaf extract. Polyester is the king of synthetic textile fibers because of the combination of excellent bulk properties and low production costs. In the first stage, we obtained the extract from mango leaves using enhanced solvent extraction (CO_2 and 50% methanol) at 120 bar and 100 °C. The resulting extract (MLE) was characterized in terms of total and major polyphenols contents and the antioxidant and antimicrobial properties (minimum inhibitory concentration, bactericidal activity). In the second stage, this extract was used in the SSI of a polyester textile. The effect of several impregnation conditions (depressurization rate, pressure and temperature) on the total polyphenols and the individual major mango polyphenols (gallic acid, mangiferin and iriflophenone glucoside) was investigated. The quality of the textile after the impregnation process was evaluated in terms of the phenolic loading and the antioxidant and antibacterial properties. The latter properties were compared with the functional properties of the crude extract.

2. Materials and methods

2.1. Materials

The raw material used in the extraction process consisted of leaves of *Mangifera indica* L. (c.v. Kent) and these were provided by the Experimental Farm 'La Mayora', Superior Centre of Scientific Research (CSIC), Malaga, Spain. All leaves were dried at room temperature to constant weight and kept frozen in the absence of light. In the impregnation process MLE and polyester fiber were used as the raw materials. Polyester textiles were obtained from a local market. The carbon dioxide (99.995%) used in both processes (extraction and impregnation) was supplied by Abello-Linde S.A. (Barcelona, Spain).

2,2-Diphenyl-1-picrylhydrazyl free radical (DPPH), mangiferin (1,3,6,7-tetrahydroxyxanthone C2-β-D-glucoside), and gallic acid were provided by Sigma-Aldrich (Steinheim, Germany). Peptone, sodium chloride and yeast extract used for the microorganism cultivation, barium chloride and sulfuric acid were purchased from Sigma-Aldrich. A bacterial culture of *Escherichia coli* CECT101 was used.

The organic solvents methanol, acetonitrile, and formic acid, all HPLC gradient grade, were provided by Panreac (Barcelona, Spain). The water used in all experiments was double-distilled milli-Q grade.

2.2. High pressure extraction procedure

Extraction was carried out in high extraction equipment from Thar Technology (Pittsburgh, PA, USA, model SF100) with a 100 mL extraction vessel and two high-pressure pumps, one for carbon dioxide and the other for the cosolvent.

The operating methodology involved loading the extraction cartridge with approximately 30 g of dried mango leaves. The extract was collected in a cyclonic separator and transferred to glass bottles and the samples were stored at 4 °C with the exclusion of light. The experimental procedure was explained in more detail in a previous publication [33].

Enhanced Solvent Extraction was selected to obtain the MLE. In this technique mixtures of CO_2 with high proportions of polar solvents (superior than 10%) are used as extraction solvents. ESE is less common than SFE and PLE, but previous studies have shown that it is a highly efficient technique to extract polar compounds from different raw materials [35,36], including phenolic compounds from mango leaves [31,33].

The extraction of mango leaves was carried with a mixture of CO_2 + 50% methanol at 120 bar, 100 °C and 10 g/min for 3 h. ESE carried out with CO_2 + 50% cosolvent has proven to be more efficient than SFE, and as efficient as PLE, to extract polar phenolic compounds from mango leaves and it has the advantages of being a more solvent-efficient method [31]. Methanol was selected as the cosolvent because previous studies have shown that methanolic mango pulp and peel extracts presented antimicrobial activities superior to those reported for ethanolic extracts [37]. In addition, some flavonoids with antibacterial activity have been isolated from methanolic mango leaf extracts [38]. Pressure and temperature were also set according to the results of previous studies, where it was found that a high temperature (100 °C) favored the recovery of phenolic compounds from mango leaves without affecting the antioxidant activity of the extracts, and that an increase in pressure did not have a significant effect on the process [31].

The extract obtained (MLE) was characterized in terms of antioxidant and antimicrobial activity and also according to the total phenolic content and the content of the predominant polyphenols.

2.3. High pressure impregnation procedure

Supercritical impregnation of the polyester textile was carried out in equipment from Thar Technology (Pittsburgh, PA, USA, model SF100). The impregnation procedure was carried out in batch mode. Methanol, 5% (v/v), was used as the cosolvent for the MLE system in order to increase its solubility in supercritical CO_2 (SC-CO_2). The methanolic MLE (5 mL) was introduced at the bottom of the impregnation cell (vessel), which had a capacity of 100 mL, and three polyester samples (approximately 100 mg each) were placed at the top of the impregnation cell in a stainless steel support so that the CO_2 could come into contact first with the extract and then with the textile. In addition, polyester textile and the MLE were separated to avoid direct contact between them. CO_2 was introduced into the impregnation cell and the conditions were maintained for a predetermined time period (static method). The impregnation cell was heated to the desired temperature prior to starting the experiment. A washing step was included at the end of the impregnation period in order to remove the excess unfixed particles and the methanol from the final product. This step involved passing a clean flow of SC-CO_2 through the system for 30 min. Finally, the system was depressurized. When pressure release was complete, the polyester fiber was removed from the impregnation cell and the samples were stored prior to analysis. Each experiment (with 3 samples)