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A hydrocolloid based biorefinery approach to the valorisation of mango peel waste

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ABSTRACT

An environment-friendly hydrothermal process was developed to extract pectin from a horticultural waste, namely mango peels, at a reasonable temperature and pressure. High average yields of pectin (15.7%, 27.2%) were obtained from the two mango cultivars namely, Calypso (Australian) and Totapuri (Indian). The physico-chemical characteristics were determined in terms of the degree of esterification (77–89%), molecular weights (M_w) (49,742–49,862 Da), FTIR and gelling tests. The spent liquor was hydrolysed to recover the specific polyphenols and sugars. The polyphenols in the hydrolysed liquid were quantified as gallic acid (525.1–1405.6 mg/l), mangiferin (7.8–162.2 mg/l), quercetin (40.6–49.2 mg/l) and ellagic acid (38.8–69.3 mg/l). The solid residue obtained as a solid co-product after pectin extraction was found to be rich in cellulose (38%) and lignin (16%). In this paper, an attempt has been made to address the challenges of the extraction of pectin from mango peels, in a conventional process. It was demonstrated that mango peels may be a promising unconventional source for recovery of food-grade quality pectin. Considerable efforts, which were earlier expended in the processing, were reduced when recovery of other food-grade co-products was integrated with the process of hydrocolloid extraction.

1. Introduction

The worldwide production of mango has reached 42 million tonnes in 2013 (Matharu, Houghton, Lucas-Torres, & Moreno, 2016). In developing countries such as India and China, a major quantity of the fruit is processed in the form of pulp, jams, jellies and candies (Banerjee, Vijayaraghavan, Arora, MacFarlane, & Patti, 2016). The processing of mango leads to 30–50 wt% of the fruit being "processing waste". Mango peel constitutes of 40–60% fraction of the total mango processing waste. The major bioactives that are found in mango peel are soluble dietary fibre (31–33%), insoluble dietary fibre (32.1–34%), polyphenols (93–96.2 mg/g of dry peel) and carotenoids (3092.2 μg/g of dry peel) (Ajila, Naidu, Bhat, & Rao, 2007; Banerjee et al., 2016; Maran, Swathi, Jeevitha, Jayalakshmi, & Ashvini, 2015). Pectin is a bioactive hydrocolloid, which is widely used as thickener, gelling agent, emulsifier, pharmaceutical additive and prebiotic (Ranganna, 1986; Sriamornsak, 2003; Thakur, Singh, Handa, & Rao, 1997). It contains 1,4-α-linked galacturonic acid and 1,2-linked rhamnose with side branches of either 1,4-linked β-D-galactose or 1,5-α-linked L-arabinose (Ciriminna, Chavarría-Hernández, Inés Rodríguez Hernández, & Pagliaro, 2015). Generally, 300–1000 saccharide units are present in the structure of pectin, which helps in the formation of gels in an aqueous solution (Ciriminna et al., 2015). The production of pectin is confined to certain regions of the world, such as Germany, Mexico, Brazil and China (Ciriminna et al., 2015). The production of pectin is governed by the presence of regional feedstock such as apple, sunflower, citrus and beetroot. The major importers of pectin are USA, Germany, Japan, France, Russia and India. The cost of pectin per kg is approximately $11–13 (Ciriminna et al., 2015). Mango peels were found to be an alternative source for pectin production; the peels were also identified as being a good source of high methoxyl and low methoxyl pectins (Berardini, Fezer, et al.,

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2005; Jamsazzadeh Kermani, Shpigelman, Pham, Van Loey, & Hendrickx, 2015; Panchev, Kirtchev, & Kratchanov, 1994).

A review of mango peel pectin extraction as described in the literature often lacks an emphasis on the by-products. Conventionally, pectin is extracted in an acid hydrolysis process. The peel slurry is heated at 90–120 °C for a minimum of 2.5 h. Among mineral acids such as nitric acid, hydrochloric acid and sulphuric acid are used in this slurry (Geerkens et al., 2015; Thakur et al., 1997). Microwave and sonication-assisted extraction were found to be good alternatives to conventional extraction (Wang et al., 2016; Maran et al., 2015). In the reported studies, when the mango peel slurry was exposed to microwave power of 413 W in the presence of an acid (pH 2.7) for 134 s, 28.8 per cent of pectin was recovered. A Pakistani variety of mango peel was used in a study, in which heating the slurry for 90 min at 90 °C in presence of acidified water (pH 2.5) recovered around 16.6% of pectin. Ultrasonication (180W) under the same experimental condition resulted in a reduced duration of 20 min, which helped in extraction of 15.8 percent of pectin. A degree of esterification, which was more than 65 percent, was obtained in both conventional and ultrasonication methods (Kauser, Saeed, & Iqbal, 2015). Apart from high DE pectins, low DE pectins were obtained from mango peels by using lemon juice assisted sonication (Banerjee et al., 2016).

In the acid based conventional processes, often the pectin is contaminated with traces of acid (Clark, Pfaltzgraff, Budarin, & De Bruyn, 2013). Apart from this, the by-product of this process generates large volumes of acidified water, which requires additional treatment before disposal (Horan, 1994). In a case study that was presented by Dalentoft and Jenson (1994), the pectin by-product from Copenhagen Pectin A/S effluent was found to be rich in organic content and had a large amount of nitrogen. The pectin wastewater contained 10,000–12,000 mg/L COD (chemical oxygen demand) and 5000–6000 mg/L BOD (biological oxygen demand) (Dalentoft & Jenson, 1994). As per the regulatory requirement, the defined range for chemical parameters prior to discharge were; BOD < 15 mg/L, nitrogen < 8 mg/L, suspended solids < 20 mg/L and pH– 6.5–7.5. The authors described a two-step treatment involving pre-denitrification, anaerobic treatment and aerobic treatment to achieve the different limits set by the regulatory authority (Dalentoft & Jenson, 1994). In the absence of the cost parameters in literature, it may be observed in the above study that industrial food by-product effluent regulations are strict; therefore, considerable efforts are required to treat such waste streams.

To address these challenges, an alternative acid-free hydrothermal treatment by using microwaves was developed in a recent study in which, 11.6% of high methoxyl pectin along with mesoporous cellulose was extracted from mango peels (Matharu et al., 2016).

As described above, mango peels are rich in nutrients apart from pectin. Therefore, a complete valorisation of the waste may be achieved in the form of an integrated process. Conventionally, organic solvent based extraction has been a popular method for polyphenols, which, however, is limited at an industrial scale due to a large quantity of organic solvent that the method requires. An Indian patent application 443/DEL/2003 discloses an organic solvent (acetone or ethyl alcohol) based recovery of polyphenols from Indian mango peels (Rao, Gururaja, & Matheyam, 2003). The patent application WO 2013/141722 A2, discloses an integrated process for mango waste (peels and seeds) to recover polyphenols and lipids using organic solvents (Taboada & Siacor, 2013).

The presence of free hydroxyl and carboxyl groups in pectin helps in linkage of the pectin chain with cellulose, lignin, and hemicellulose. Conventional acid hydrolysis may cleave these linkages, which results into solubilisation of the protopectin. The studies on the hydrothermal extraction of capsaicinoids from pepper and polycyclic aromatic hydrocarbons from soil matrix have reported that the high yield of such compounds was governed by a complex interaction of many factors (Carr, Mammucari, & Foster, 2011). The effect of pressurized heating on physico-chemical parameters of water, such as dissociation constant dynamic viscosity and surface tension was investigated in a study. Under the ambient conditions (25 °C), the dissociation constant of water was calculated to be 1.0×10^{-14}. At an elevated temperature, the thermal agitation was also accelerated, which resulted in the disruption of hydrogen bonds and other intermolecular forces. An increase in the dissociation constant was observed along with decrease in the dynamic viscosity and surface tension, respectively (Carr et al., 2011; Plaza & Turner, 2015). A temperature increase in the range of 298–498 K was found to decrease the dielectric constant of water from 80 to 27 which lies between dielectric constants of methanol and ethanol. Hence, the dissolution of the organic components increased in water at elevated temperatures (Miller, Hawthorne, Gizir, & Clifford, 1998; Ong, Cheong, & Goh, 2006; Teo, Tan, Yong, Hew, & Ong, 2010). The nature and composition of the bioactive compounds was considered to be an important factor in these studies. The bioactives that are rich in conjugated systems, demonstrated a higher solubility due to the delocalized electrons in a cluster, which improves the interaction with the solvent under hydrothermal treatment conditions (Carr et al., 2011).

The changed properties of water under the hydrothermal treatment may help in recovery of the pectin from mango peels. As water behaves like an acidic solvent under hydrothermal treatment, an enhanced separation of pectin from the biomass matrix may be obtained. Due to the structural disruption, polyphenols and sugars may also be co-extracted with pectin, leaving residual peel more porous. Thus, to further increase the overall value of the product, the characterization of the co-products was done in order to evaluate their potential as novel nutraceutical additives or as precursors to biofuels/bio-based chemicals. The solid residue as a co-product in the process was also studied for its carbohydrate composition.

Thus, a green biorefinery approach was applied to valorise the mango peels, which in addition to generating value-added products, eliminates the use of hazardous solvents and chemicals in the process of extracting pectin.

2. Materials and methods

The mango peels that were used in the study were obtained from the local market. The two commonly processed varieties; Totapuri (Indian) and Calypso (Australian) were used in the study to compare the pectin yields and impact of geographical differences on the quality. The studies on Indian peels were performed at the Indian Institute of Technology Bombay (IITB), India while the studies on Australian peels were conducted at Monash University, Australia. All the reagents and chemicals, unless specified, were procured from Sigma Aldrich. The HPLC standards such as mangiferin, ellagic acid, gallic acid and quercetin were procured from Sigma Aldrich.

The mango peels were blanched in hot water at 90 °C for 5 min. Further, the blanched peels were dried at 50 °C for 7 h in a hot air food drier (until a constant weight was achieved). The dried peels were ground in a kitchen mixer grinder and the powder was passed through a 500μ sized sieve. The dried powder was packed in an air tight container and stored at 4 ± 1 °C until further use.

2.1. Pectin extraction

A slurry of peel was prepared in water in the ratio, 1:20 w/v (1 g of solid to 20 ml of water). The solid-to-liquid ratio was optimized