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Coffee pectin production: An alternative way for agricultural waste management in coffee farms

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ABSTRACT: Chiang Rai Province is home to the largest Arabica coffee plantation, accounting for 16% of coffee plantation in Thailand. During the green bean production from coffee cherries, 45% of the coffee pulp is treated as agricultural waste. This study aimed to increase the value of the coffee cherry pulp by using it as an alternative source of pectin. A double extraction process was used to extract pectin: in the first extraction, acid was used to extract the coffee pulp, followed by the second extraction with base. Both acid and base solutions yielded from the extractions were combined prior to the pectin precipitation step. The pectin yield from this double extraction method was 2-fold higher than that yielded from previous methods. Furthermore, to reduce toxicity during the extraction method, we use citric acid to replace hydrochloric and nitric acids. Among three heating conditions during extraction: boiling, autoclave, and microwave, the boiling method gave the highest pectin yield at 15.9%. Unlike the high methoxyl pectin (HMP) yield from citrus, the coffee pectin from the boiling and microwave-assisted methods was categorized as the low methoxyl pectin (LMP). The LMP from coffee cherry can be used as prebiotic supplement or in wound dressing film production. Importantly, producing LMP not only has the potential to reduce post-harvest agricultural waste by 3800 tons per year in Chiang Rai Province but also provides value to agricultural waste and additional income to coffee growers.

KEYWORDS: coffee pectin, low methoxyl pectin, double extraction process

INTRODUCTION

Coffee is one of the most favorite drinks consumed around the world. There are more than 120 varieties of coffee, but the two most popular are *Coffea arabica*, commonly known as Arabica, accounting for 60–80% of global coffee production, and *Coffea canephora* also known as Robusta, which accounts for 20–40% of production [1]. Thailand is the third largest coffee production country in Asia, with 24 000 tons of Robusta coffee cherry produced in the South and 11 000 tons of Arabica coffee cherry produced in the North [2]. Of these, 4900 tons are produced in Chiang Rai Province, the largest producer of Arabica coffee in Thailand. During the green bean processing, the skin, pulp, mucilage and parchment are left as agricultural waste, accounting for 55% of coffee cherries. Coffee pulp is largely composed of carbohydrates, including pectin (20–35%), oligosaccharides and fiber (30%), along with proteins, minerals, and water. It has been estimated that 1 kg of coffee pulp can yield 49.8 g of dried pectin [3]. Commonly, pectin is largely produced in the fruit juice industry from citrus peel and apple pulp waste. Nonetheless, agro-waste from local fruits, such as banana, passion fruit, jackfruit, as well as coffee pulp, may also be an alternative source of pectin [4]. Pectin is a complex heteropolysaccharide made of D-galacturonic acid (GalA) monomers, which are linked to each other by α-(1→4) galacturonosyl linkages, part of the carboxyl groups of GalA are esterified with methoxyl groups. The percentage of esterified carboxyl groups determines the degree of esterification (DE), a feature that is used to decide the type of pectin as shown in Fig. 1.

The DE of HMP is higher than 50%, while that of LMP is lower than 50%. The varying DE makes the properties of HMP and LMP different. For example, HMP requires high amount of sugars for gelation and it is very sensitive to acid, while LMP can form a soft gel in the presence of ions, such as calcium

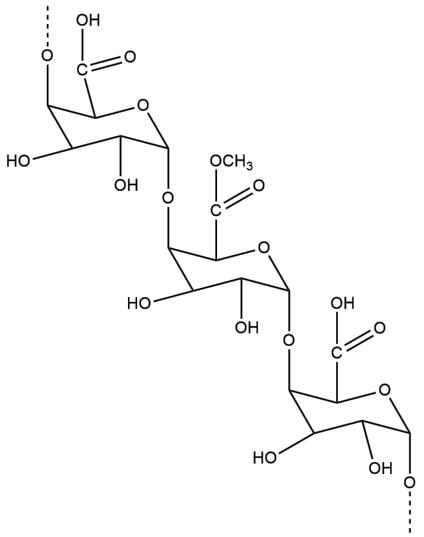

Fig. 1 Structure of pectin.

ions (Ca^{2+}) [5]. Both types of pectin are used in the food industry, but HMP is mainly used for thickening and gelling properties. The LMP is used instead of HMP in low sugar content foods, film in the packaging industry or as supplementary fiber [6]. Though agricultural waste such as citrus peel, apple pomace and sugar beet constitutes the main source for pectin production [7], coffee pulp could be an alternative source. Many extraction methods have been reported regarding pectin extraction from coffee pulp. The classical method was reported by Garcia et al [8] in 1991, who reported extraction of LMP from coffee pulp at a yield of 10.65% with DE of 23.81. In their protocol, they used hydrochloric acid combined with ethanol in 1:1 ratio at a final pH of 2.0 during extraction. Reichembach and Petkowicz (2020) reported extraction of HMP from coffee pulp with DE of 63.2 and 14.6% yield using 0.1 M nitric acid [6].

The main aim of this study was to develop a suitable pectin extraction method from coffee pulp. Furthermore, the study investigated the optimal conditions for higher yield and usage of more environmentally friendly, less toxic chemicals than those used in previous studies.

MATERIALS AND METHODS

Sample storage and preparation

Coffee cherry pulp from wet processing (*Coffea arabica* var. Catimor) was supplied by Akha Coffee Co. Ltd., from a coffee plantation in Mae Suai District, Chiang Rai Province, Thailand during the 2018–2019 harvest. The fresh coffee cherry was stored at −20 °C prior to extraction. At least 24 h before extraction, the pulp was soaked in 1% (w/v) of sodium bisulfite solution in 1:1 ratio at room temperature and pulp residual was filtrated out prior to extraction.

Conventional extraction

The extraction method was modified from that reported by Rakitikul et al [9], whereby 100 g of coffee pulp was macerated with 200 ml of 1% (v/v) nitric acid at a final pH of 3.0. The mixture was then boiled for 3 h at 90 °C with constant stirring. After extraction, pectin solution was cooled down to room temperature and filtered through cheesecloth. The filtrate was then adjusted to pH 5.0 with 0.5 N sodium hydroxide solution. One volume of filtrate was precipitated with two volumes of 95% ethanol for 12 h at 30 °C. The formed gel was filtrated through the cheesecloth and dried in a drying oven (BINDER model FD240, Germany) at 60 °C for 24 h. The dried pectin was kept in desiccator for further study. The extraction experiments were carried out in three replicates.

Double extraction method

The extraction method was adapted from that reported by Andres et al [10]. The coffee pulp was macerated with of 0.1 N citric acid (pH 3) in a ratio of 1:2 (w/v) prior to treatment using three different heating conditions; boiling, autoclave and microwave. For the boiling treatment, the solution was boiled for 3 h at 90 °C with constant stirring; for the autoclave extraction, the solution was heated at 121 °C, 15 psi for 15 min; and for the microwave-assisted extraction, the solution was heated in a microwave oven at 900 watt of power for 15 min. After extraction, all three suspensions were cooled down to room temperature and filtered through cheesecloth. The filtrates (hereafter called acid solution) were kept separately for further pH adjustment using the solution from the second extraction. The pressed coffee pulp cake of each treatment was mixed separately with 0.5 N NaOH in 1:1 ratio (w/v) and the suspension was stirred constantly for 1 h at 30 °C. The suspension from alkali extraction was then filtrated through the cheesecloth and then both acid and alkali solutions were combined, and the pH adjusted to 5.0 prior to precipitation with two volumes of 95% ethanol for 12 h at 30 °C. The formed gel was filtrated through the cheesecloth and dried in an oven at 60 °C for 24 h. The dried pectin was kept in desiccator for further study. The

extraction experiments were carried out in three replicates.

Analysis of Fourier transformed infrared spectroscopy (FTIR)

Crude coffee pectin was subjected to FTIR spectroscopy using a Perkin Elmer FTIR spectrometer model Spectrum GX (Perkin Elmer Co., MA, USA). Samples were ground into a fine powder using a mortar and a grinder prior to compression into KBr discs. The characteristic spectra were recorded in the range of 4000–450 cm^{-1}, at a resolution of 4 cm^{-1} [11].

Determination of degree of esterification (DE)

The degree of esterification of pectin was determined by the titrimetric method. A volume of 0.2 g of crude pectin was dissolved in 20 ml of distilled water at 40 °C and stirred until homogenized. After that, phenolphthalein reagent was added to the samples as an indicator, and titrated using 0.1 N sodium hydroxide, the volume of used sodium hydroxide solution was recorded as V_1. Then, 10 ml of 0.1 N sodium hydroxide was added to samples and stirred 15 min for hydrolysis, followed by addition of 10 ml of 0.1 N hydrochloric acid and stirring until pink color disappeared. The total sodium hydroxide volume was recorded as V_2. The DE was calculated as shown in equation (1) [12]:

$$\%\,\mathrm{DE} = \frac{V_2}{V_2 + V_1} \times 100. \tag{1}$$

Pectin yield

A volume of 100 g of fresh coffee pulp yielded 20 g dry weight, and this was used as the initial weight for yield calculation. Since 100 g of fresh coffee pulp has been estimated to contain 4.98 g of dried pectin [13], this value was used for calculation of pectin yield and extraction efficiency as stated in equations (2) and (3), respectively [14]:

$$\text{Pectin yield}\,(\%) = \frac{\text{weight of dried pectin (g)} \times 100}{\text{weight of dried peel used in extraction}} \tag{2}$$

$$\text{Extraction efficiency}\,(\%) = \frac{\text{weight of dried pectin (g)} \times 100}{4.98} \tag{3}$$

where 4.98 is the estimated pectin in gram yielded from 100 g fresh coffee pulp.

RESULTS AND DISCUSSION

Effect of extraction methods on the yield of coffee pectin

Each of 100 g of coffee pulp was subjected to conventional method extraction and double extraction method, in order to compare the type of acid and base and effect on the yield, % DE and color of dried pectin (Table 1).

The pectin from the double extraction method was determined as LMP and the yield was higher than the conventional method (10.7%). The yield (15.9%) was also higher than that reported by Reichembach and Petkowicz [6], whose experiments resulted in HMP with 10.7% yield using 1% (v/v) nitric acid. Moreover, the yield was higher than that found by Garcia et al [8], who used hydrochloric acid combined with ethanol in 1:1 ratio at the final pH 2.0 to extract LMP from coffee pulp with a yield of 10.7% and DE of 23.8. The type of pectin is dependent on the source of the plant and the acid or base used during extraction. Generally, the acid extraction yielded HMP, while basic extraction yielded LMP as a result of saponification of the ester groups [10]. Considering the results of this experiment, the double extraction method, during which coffee pulp was initially extracted using acid followed by using base, yielded LMP and a higher yield than the previously reported method. In order to avoid use of hazardous materials, citric acid was used as an alternative acid source instead of the strong acids (e.g. nitric acid and hydrochloric acid) used previously. Considering this, it is possible that the observed results could also be due to using a different acid than the one used originally. In the next section, the effects of heating condition are discussed.

Effect of heating condition during extraction method

In this part, three heating conditions: boiling, autoclave and microwave, were applied during double extraction method. The yield, extraction efficiency, DE, and type of pectin were compared (Table 2). The best heating condition was boiling method, which gave 15.9% yield, while autoclave and microwave-assisted methods yielded 10.7% and 9.3%, respectively. The pectin from autoclave heating method was of the HMP type, suggesting that despite of the solvent used in the extraction, the heating condition also affected the type of pectin. Furthermore, the double extraction method with citric acid as a solvent could naturally yield LMP

Table 1 Effect of extraction method on the characteristics of coffee pectin.

Analysis	Conventional extraction method	Double extraction method
Extraction solvent	1% (v/v) nitric acid, pH 3.0	0.1 N citric acid, pH 3 and 0.5 N NaOH
Extraction condition	Boiling at 90 °C, 3 h	Boiling at 90 °C, 3 h
Pectin yield (%)	10.7 ± 0.3	15.9 ± 0.4
Degree of esterification	62.1 ± 0.8	28.6 ± 0.2
Type of pectin	High methoxyl	Low methoxyl
Pectin color	Dark green	Light brown

Table 2 Effect of heating conditions on the characteristics of coffee pectin.

Analysis	Heating condition		
	Boiling	Autoclave	Microwave
Pectin dry weight (g)	3.14 ± 0.04	2.14 ± 0.05	1.85 ± 0.07
Pectin yield (%)	15.9 ± 0.4	10.7 ± 0.3	9.3 ± 0.6
Extraction efficiency (%)	63.1	43.0	37.2
Degree of esterification	28.6 ± 0.2	50.2 ± 0.1	44.4 ± 0.1
Type of pectin	LMP	HMP	LMP

from the coffee pulp, while most of the commercial LMP was generally produced by de-esterification of HMP via enzymatic treatment [15]. Thus using this method and citric acid as solvent would greatly reduce production cost and number of steps required to produce LMP.

FTIR analysis of the coffee pectin

The spectra of pectin from commercial HMP and LMP and coffee pectin extracts using three heating treatments are shown in Fig. 2 and Fig. 3, respectively. The strong evolving peaks at 1747 cm^{-1} depicted esterified carboxyl groups (a), while peaks at 1022 and 1106 cm^{-1} represented backbone of pectin and those at 1149 cm^{-1} (b) indicated glycosidic linkages (C—O—C) of two monomers (c). Furthermore, the strong absorption at 1460 cm^{-1} (d) represented highly esterified pectin [11, 16]. The absorption area in FTIR spectra were consistent with the calculated %DE from titration method, as we could observe strong absorption at region (a) in pectin yield from autoclaved treatment and weak absorption in microwave and boiling treatments, respectively.

Our study was in agreement with the result from Reichembach and Petkowicz study, which yielded HMP from the coffee pulp when using 0.1 M of nitric acid [6]. The double extraction method performed by Andres and Belalcazar using hydrochloric acid also yielded HMP from coffee pectin [10]. The change in solvent from nitric acid or hydrochloric acid to citric acid in this study, not only reduced

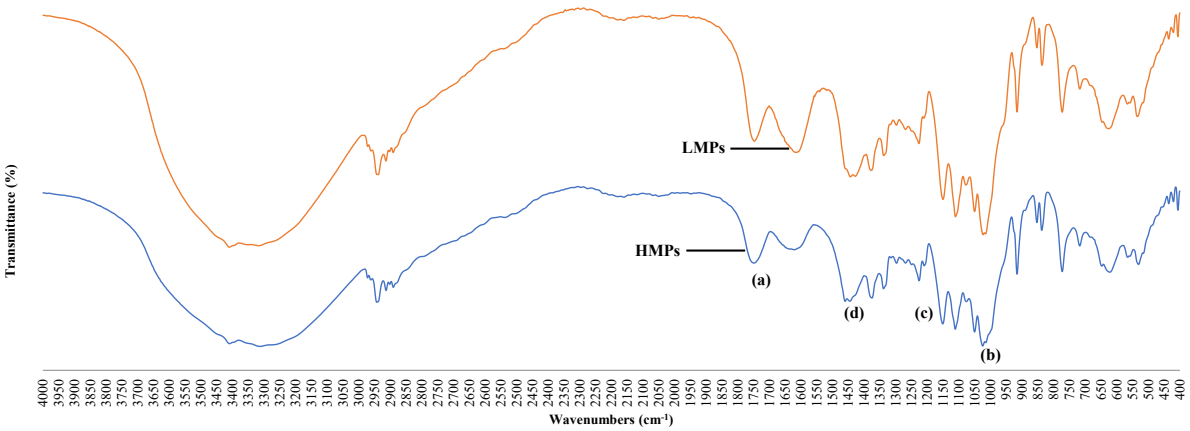

Fig. 2 FTIR spectra of commercial pectin of the HMP and LMP types.

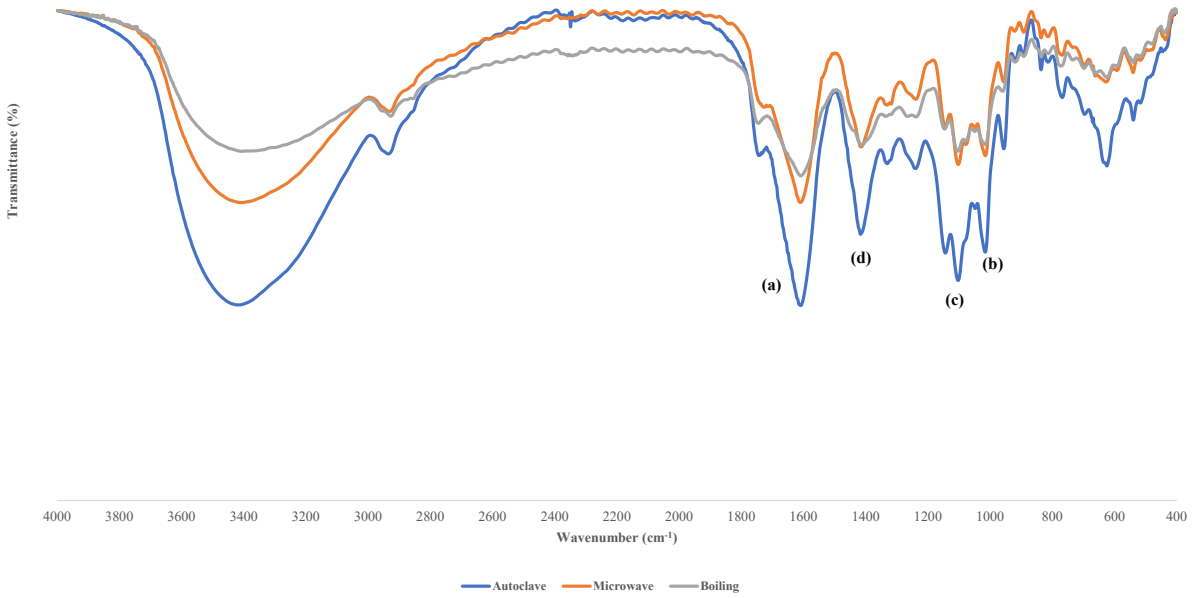

Fig. 3 FTIR spectra of coffee pectin extracted by different extraction methods.

the toxicity of the solvent used during extraction, but also gave 5% higher yield than that reported in previous studies.

CONCLUSION

In this experiment, the yield of pectin from coffee pulp was higher by 5% when using double extraction method than the conventional method. The optimal parameters in double extraction method for yielding LMP, were boiling coffee pulp in 0.1 N citric acid at 90 °C for 3 h. Given the best yield and type of pectin, the boiling condition can be applied at industrial extraction scale in the future. Citric acid used in this experiment is less toxic compared to nitric acid or hydrochloric acid, both of which are commonly used in the conventional extraction method. Finally, producing LMP from coffee pulp has the good potential to reduce post-harvest agricultural waste. Production of coffee pectin also provides value to agricultural waste and additional income to coffee growers. The extraction method in this article is under the petty patent "LMP extraction method from coffee using organic acid and base" registration number 2003002291.

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