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EXTRACTION OF NATURAL DYE FROM BLACK TEA WASTE FOR COTTON DYEING APPLICATION

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Abstract

An investigation has been made to predict the efficiency of ultrasound-assisted extraction methods in natural dye recovery from black tea waste for cotton dying applications. The black tea waste was obtained from Universiti Malaysia Kelantan (UMK) cafeteria. The ultrasonic-assisted extraction was done for different volume of solvents (100 ml, 200 ml, 300 ml, and 400 ml). The solvent used in this research work is petroleum ether. Result shows that 300 ml of solvent volume gives highest yield of extract. Then, the extraction is further proceeding with different extraction time (2 hours, 4 hours, 6 hours and 8 hours) before followed to extraction without ultrasonic. The result shows that 8 hours of extraction time is more effective in extracting black tea waste. For cotton dyeing application, it has demonstrated an excellent dyeing result at 40℃ -50℃ with a minimal amount of metal mordant (0.5 %). The experimental data were analysed using the colorimeter to determine the specification of colours, and Fourier transform infrared spectroscopy (FT-IR) to confirm the components in black tea waste. Results from this work mainly can be applied in the textile industry.

Keywords: Black tea waste, Cotton dyeing, Natural dyes, Theaflavins, Ultrasonic-assisted extraction.

1. Introduction

Tea is one of the world's most essential beverages and it is produced in at least 30 countries worldwide [1]. Most of these end up landfill sites. Waste derived- natural dyeing from tea waste will give additional value and economically beneficial for the application in the textile dyeing industry [2]. From that, we can control our environment from pollution by using natural dye for coloration of textiles replacing chemical colorants.

The application of tea (Camelia sinensis) as a natural dye on natural fibers has been reported in the literature [3]. Tea contains theaflavins and thearubigins as the chief colouring components. Both components have hydroxyl groups in their structures in a position favourable for the formation of complexes with suitable metal [4]. The pigments of theaflavins are the yellowish-brown while the pigments of thearubigins are reddish-brown.

Diverse techniques are available in extracting dye such as aqueous, alkaline, acid, microwave, ultrasonic and super critical fluid extraction methods [5]. Different extraction methods affect the yield, reduce the quantity of the natural colorant and also the stability of the dye itself. Considerable research is being done around the world on the application of natural dyes [5, 6]. An exhaustive review on the subject of natural dyes in textile applications has been published by Taylor [7]. There are different techniques for the extraction of colouring materials such as aqueous, alkali, acid, microwave, ultrasonic, enzymatic, supercritical fluid, solvent extractions [2], and fermentation methods [8].

According to Bydoon [9], different shades of colour can be obtained using different chemical and natural mordants using tea as a natural dye source. Although literature shows that numerous techniques have been done in extraction of natural colorant from tea waste, there are some technical limitations occur (i.e., low yield, longer dyeing time, high cost of mordants and larger dyestuff). Additionally, some limitation of the listed extraction methods involves labour intensive for Soxhlet extraction, high analytical cost for supercritical fluid extraction, subjected to interferences of microwave energy absorbing materials for microwave assisted extraction and safety issues for pressurized fluid extraction. The mechanism behind the ultrasound-assisted extraction methods is as shown in Fig. 1. Hence, this study aims to use ultrasound methods to improve the efficiency of colour extraction. Thus, the rate of the extraction can be completed in a shorter time with better yield.

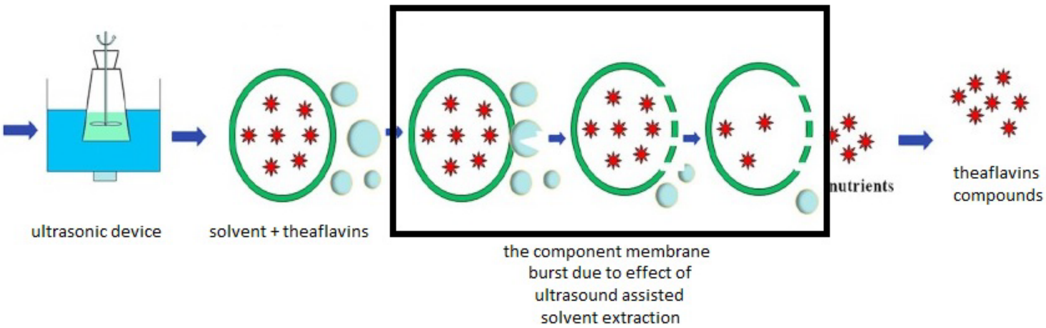


Fig. 1. Mechanism of ultrasound-assisted extraction method.

2. Methods

2.1. Raw material preparation

The black tea waste was obtained from UMK Jeli cafeteria. Then, the raw black tea waste was washed with tap water for 2 to 3 times to remove the impurities (i.e., dust, caffeine and catechins). After that, the raw black tea waste was dried for 2 days under the sunlight and oven dried for 30 minutes at 105°C. The raw black tea waste was crushed using milling machine. The spindle is driven by a motor and therefore rotates the arbor. During milling, the cutter rotates along a horizontal axis. The cutter's side removes the raw black tea waste from the workpiece until it passed through 350 μm mesh size to increase the surface area of the samples in contact to the solvent.

2.2. Ultrasound - assisted extraction

A total of 480 g of black tea waste were soaked in 1000ml of distilled water. The ultrasound – assisted extraction process was carried out in a water bath where the sample was sonicated at 20 – 25 kHz for 30 minutes with amplitude of 60℃. For each different parameter, about 20 g of sonicated dried raw black tea waste was extracted using a Soxhlet extractor wool. The extraction was done at different extraction time: 2 hours, 4 hours, 6 hours and 8 hours at difference solvents (petroleum ether) volume (i.e. 100, 200, 300 and 400 ml). The steps were repeated without using the ultrasound-assisted extraction method of black tea waste under the optimum condition.

2.3. Mordanting

Cotton was added and rotated often for the first 15-20 minutes and fully submerged in the copper solution. Then, the copper sulphate mordant bath was heated at 50℃ and hold for 30 minutes. After mordanting, the samples were rinsed in cold water to remove the excess mordant and then dye with dye extract from ethanol. The steps repeated using black tea waste without sonicated.

2.4. Functional group analysis

The extract solution of raw black tea waste and fresh tea waste powder from organic solvents which are petroleum ether were analysed by FTIR in the range of 400-4000 cm^{-1} with a resolution of 4 cm^{-1}.

2.5. Colour analysis

The characterization of colour was determined using a colorimeter. The colour was analysed based on the L*, a* and b* values. L* values express darkness to the lightness that ranges from 0 to 100, while for positive or negative a* value, it is related to the (+) redness or (-) greenness of the samples. The b* values present the colour range from positive b* value (yellow) to negative b* value (b). The measurement of colour explains that it enhances the intensity of colour effects that covers the difference of lightness and saturation.

3. Result and Discussions

3.1. Effect of solvent volume

Table 1 shows that 400 ml of solvent was more effective than others for extracting black tea waste samples. 400 ml of solvent gives the highest average yield of 15 ml, followed by a 13 ml average yield of the extract, using 300 ml of solvent. However, the concentration of theaflavins extracted based on the calibration curve (Fig. 2) shows that the solvent of 300 ml gives high concentrations of theaflavins compared to 400 ml. The best conditions of theaflavins extraction were at 300 ml of solvent. These conditions were applied to prepared solvents for further extraction.

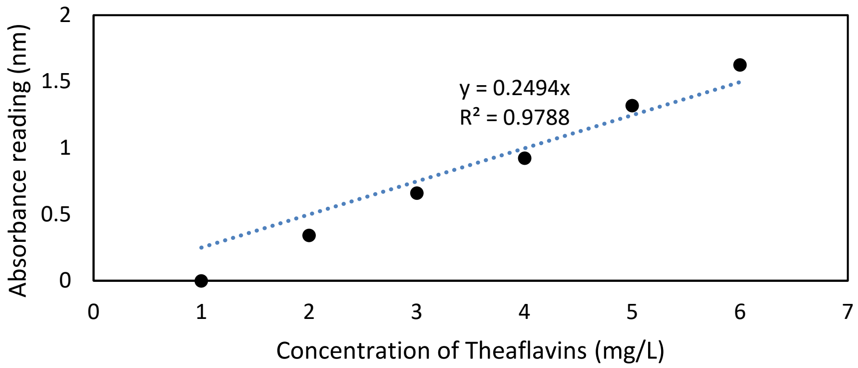


Fig. 2. The standard curve of theaflavins.

Table 1. The concentration of theaflavins with different solvent volume.

Volume of solvent (ml)	Time of extraction (hours)	Average volume of extract (ml)	Concentration of theaflavins (mg/L)
100	4	8.84	0.54
200	4	10.50	3.20
300	4	13.11	4.00
400	4	15.00	3.84

3.2. Effect of extraction time of black tea waste

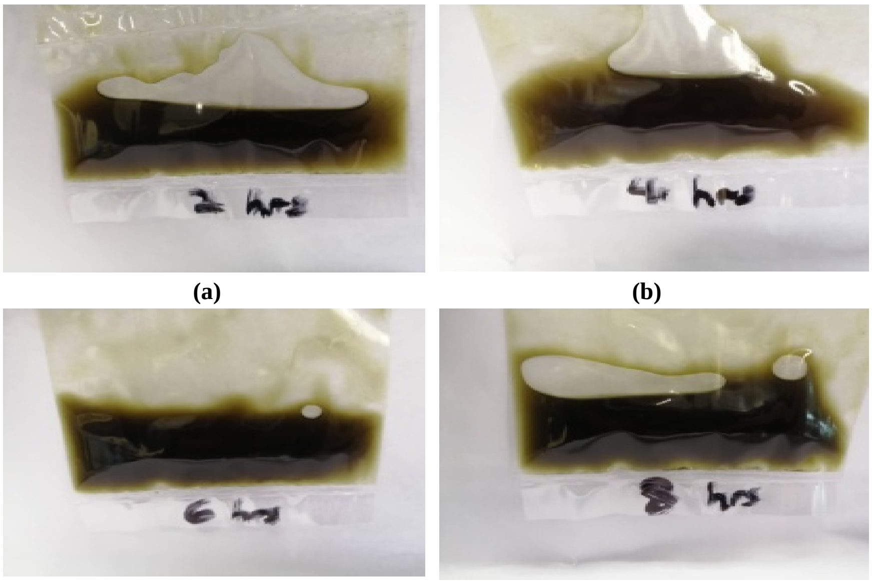
From Table 2, the average yield of extract after 4 hours extraction was decreased to 11.78 ml at 6 hours and 10.67 ml at 8 hours. It also observed that the yield of extract decrease may be due to evaporation during extraction. The result obtained the highest average yield of extract was at 4 hours which is 13.11 ml. It shows that 4 hours of extraction give the optimum time for maximum production extract from 20 g of black tea waste. Other study supported it by proving that 4 to 5 hours give optimum time for extraction [1]. However, these results showed that the concentration of theaflavins was higher at 8 hours of extraction, which 5.22 mg/L.

Table 2. The concentration of theaflavins with different time of extraction.

Volume of solvent (ml)	Time of extraction (hours)	Average volume of extract (ml)	Concentration of theaflavins (mg/L)
300	2	8.99	0.42
300	4	13.11	4.00
300	6	11.78	4.63
300	8	10.67	5.22

3.3. Colour analysis

Colour Using Conica Minolta Colorimeter, the L*, a* and b* measurements were used as colour classification. It was found that the colour of the black tea waste extracted by 2 hours was lighter than the others time extraction based on their L* value, as shown in Fig. 3. Based on the results, increase time of extraction was indicated the darkest colour appearance of extracted. Result also showed that the positive value of a* for all samples. Therefore, more redness was observed for the extracted black tea waste. However, the redness was observed for extracted was not affected by the solvent used. All samples showed a positive value in b* indication and thus are more yellow. The time extraction of 8 hours yield gives the highest value in b* among three extraction times studied.



(a) **(b)**

(c) **(d)**

Fig. 3. Colour analysis of black tea waste at different extraction time (a) 2 hours, (b) 4 hours, (c) 6 hours and (d) 8 hours.

3.4. Effect of different extraction time on dyeing application

From Table 3, the results indicated that the cotton dyed with used black tea waste using 0.5% of copper sulphate yield a variety of greenish colour shades according to different parameters while those cotton dyed with a non-mordant produce lightness of the brown shades on the cotton fabrics. It was observed that, from the difference used for extraction, the cotton fabric using 8 hours showed a higher depth of shade and colour values. Compared to the test of the cotton fabrics using other parameters of extraction. On the other hand, the cotton fabric without a mordant was observed to have the lightest colour shades and was influenced little by mordant use. It shows that the mordant leads to creating a diversity of greenish shades compared to those without mordant. It also explains that different types of dyed fabrics tend to be influenced by mordant types, due to a reaction with oxygen in the air [10]. Therefore, the extraction of used tea, represents a potential natural dye resource for creating various shades of brown colours on fabrics. This process involved low-temperature dyeing at 40 -50℃ and used of low metal mordant percentage, which is advantageous for industrial natural dyeing as well as energy-saving and eco-friendly. Thus the net enhancement of dye uptake due to metal mordanting has been reported [11] to be 64 -67% in cotton.

Table 3. The effect of different extraction time on fabrics using extracted natural dye.

Time extraction	Control	With mordant (copper sulphate)
2 hours		
4 hours		
6 hours		
8 hours		

3.5. Effect of ultrasound-assisted extraction

Table 4 showed that the effect of extraction method of theaflavins from black tea waste. Result shows that the concentration of sonicated theaflavins (4 mg/ml) is higher than the concentration of theaflavins without sonicated which is only 3.2 mg/ml. The results as reported in Table 4, revealed that the ultrasound- assisted extraction was much better than the extraction without ultrasound.

The results showed the significant difference colour between ultrasound-assisted extraction methods with the extraction without ultrasonic. The ultrasonic-assisted extraction method revealed the depth shade of cotton much better compared to extraction without ultrasonic. It could be proved that the technique of ultrasonic-assisted extraction of theaflavins from black tea waste was indeed useful and feasible in this study. From this research, black tea waste has a very good substantivity for cotton. In the case of mordanting method using ultrasonic and without ultrasonic, some of the dye was lost because of the formation of an insoluble complex in the dyebath itself. In contrast, while dye without using ultrasound some of the mordant was stripped out in the dyebath, which subsequently forms an insoluble compound with dye molecules in solution. Thus, without ultrasound decrease in the effective dye concentration in dye bath.

Table 4. The effect of extraction method of theaflavins from black tea waste.

Sample	Volume of extracted (ml)	Concentration of theaflavins (mg/ml)	Colour shade	
			Without mordant	Mordant
Sonicated	10.67	5.22		
Without sonicated	9.50	4.02		

3.6. Functional group analysis

Based on the Theaflavins are the responsible compounds to determine the colour of black tea infusion as conjugates of epicatechin (EC), epigallotechin (EGC) and gallic acid. The primary product ions of theaflavins 3-0-gallate in Fig. 4 was at peak of 1379.85 cm^{-1} and 1380.97 cm^{-1} respectively. The result corresponds with the previous research which was bands at 3416 cm^{-1} [12]. The peak at 1380.97 cm^{-1} is indicated the presence of alkane and aldehyde which is C-H on the without ultrasound spectrum and the peak was a shift to right at 379.85 cm^{-1} using ultrasound extraction. The previous research shows that the bands 2924 cm^{-1} were due to C-H stretching and bending vibrations [13]. The peak at 518.34 cm^{-1} indicates (C-O-C) of polysaccharide at spectrum without ultrasound. The peak then shifted to the left wavenumber of 602.98 cm^{-1} when using the ultrasound extraction. Thus, black tea waste contains protein-like, aliphatic-like, cellulose-like substances with hydroxyl, carboxyl and amine groups [14]. Based on Fig. 4, it showed the Fourier-transform infrared spectroscopy (FT-IR) result for the extraction of black tea waste using ultrasound-assisted extraction method have the highest peak of the functional group compared to the extraction without ultrasound.

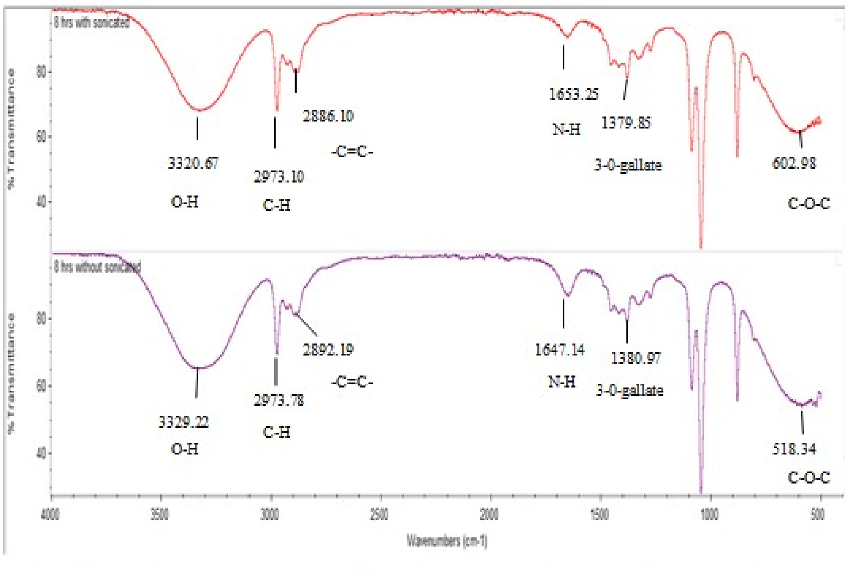


Fig. 4 . FTIR spectra of extraction black tea waste without using ultrasound-assisted extraction and ultrasound assisted extraction method using optimum parameters.

4. Conclusions

In conclusion, theaflavins successfully extracted from black tea waste by using ultrasound-assisted extraction with different volumes of solvents (100 ml, 200 ml, 300 ml and 400 ml) and different time (2 hours, 4 hours, 6 hours and 8 hours). The optimum volume of solvent to extracted 20 g of black tea waste was 300 ml and the optimum extraction time was 8 hours. Extraction time also gives effect to the extraction yield. It provides enough time for the chemical bond of phenolic compounds to break and diffuse into the solvent. Furthermore, using ultrasound-assisted extraction which is a sonicator bath give the different effects of extraction. FTIR characterized the presence of functional groups such as primary aliphatic alcohol and aliphatic hydrocarbon. For the cotton dyeing application, black tea waste has been shown to have good dyeing prospects. It can be explored for commercial natural dyeing, particularly the waste generated in the Tea leaves processing. 0.5 % of metal mordant was used at low-temperature dyeing (40 ˚C-50 ˚C), and dye effluents are less contaminated with metal salts, which is advantageous for natural industrial dyeing, being energy-saving and eco-friendly.

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