

# Optimization of total monomeric anthocyanin (TMA) and total phenolic content (TPC) extractions from mangosteen (*Garcinia mangostana* Linn.) hull using ultrasonic treatments

C.Y. Cheok<sup>a</sup>, N.L. Chin<sup>a,\*</sup>, Y.A. Yusof<sup>a</sup>, R.A. Talib<sup>a</sup>, C.L. Law<sup>b</sup>

<sup>a</sup> Department of Process and Food Engineering, Faculty of Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

<sup>b</sup> Department of Chemical Engineering, Faculty of Engineering, University of Nottingham, Malaysia Campus, Jalan Broga, 43500 Semenyih, Selangor, Malaysia

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## ABSTRACT

The extraction yields of anthocyanins (TMA) and total phenolics (TPC) from mangosteen hull were optimized by varying the amplitude and time of ultrasonic treatment. The highest TMA recovery of 2.92 mg cy-3-glu/g hull powder was achieved using methanol aqueous solvent when direct ultrasonic treatment was applied for 15 min at 20% amplitude. For the TPC, 245.78 mg GAE/g hull powder was obtained in ethanol with sonication time of 25 min and at 80% amplitude. These TMA and TPC yields obtained are respectively 45.6% and 8.8% higher ( $p < 0.05$ ) when compared to those without ultrasonic treatment. The ultrasonic treatment is able to improve anthocyanin extraction more effectively than the total phenolics from mangosteen hull.

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## 1. Introduction

The application of ultrasound-assisted extraction for plant materials or herbs (Vinatoru, 2001) and its opportunities in the food processing industry (Vilkhu et al., 2008) has proved many advantages. Its effectiveness in increasing extraction yield is proven as the ultrasonic wave generates supersonic liquid microjet that has the

(Adjé et al., 2010) and *Garcinia indica* Choisy (Nayak and Rastogi, 2011), and the extraction of total phenolics from coconut shell powder (Rodrigues and Pinto, 2007) and rice bran (Tabaraki and Nateghi, 2011). The anti-cancer property of anthocyanins (Wang and Stoner, 2008) and the remarkable relationship of total phenolics with antioxidant capacity (De Oliveira et al., 2009) have intensified the needs of these extraction studies. The efficacy of

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Sakagami et al., 2005; Tewtrakul et al., 2009) have grown important and expanding due to the climbing need of pharmaceutical products in combating various kinds of diseases especially cancer (Itoh et al., 2008; Nakagawa et al., 2007). The biological active constituents of mangosteen have been reviewed and highlighted by Dembitsky et al. (2011).

The use of ultrasonic is widely explored recently in the extraction of bioactive compound from plant materials. Examples are the extraction of anthocyanins from *Delonix regia* tree flowers

in the extraction of salvianolic acid B from *Salvia miltiorrhiza* root (Dong et al., 2010), and the extraction of phenolic acid from *Citrus unshiu* Marc peels (Ma et al., 2009). The ultrasound-assisted extraction was revealed to have the ability of extracting low molecular weight semivolatiles makers from two unifloral honeys from *Robinia pseudoacacia* L. and *Castanea sativa* L. that were unable to be extracted using hydrodistillation (Jerković et al., 2007). Even though Aspé and Fernández (2011) reported that the yield of total phenols and tannins extracted using ultrasound-assisted extraction from *Pinus radiata* bark were comparatively less superior when compared with the Soxhlet method, they, however, observed a remarkable reduction of 98.3% in the extraction time (Aspé and Fernández, 2011).

Apart from its advantage of a single direct extraction process, ultrasound has also found its coupling use with other extraction methods. When coupled with a vacuum distillation process for the extraction of flavor compounds from varieties

\* Corresponding author. Tel.: +60 3 89466353; fax: +60 3 89464440.

E-mail address: [chinnl@eng.upm.edu.my](mailto:chinnl@eng.upm.edu.my) (N.L. Chin).

of *Mentha spicata*, it produced extracts with higher flavoring strength compared with hydro-distillation (Porto and Decorti, 2009). In combining ultrasound with agitated bed extraction, it has resulted higher total phenolic yields from *Jabuticaba* compared to sole ultrasound-assisted extraction or agitated bed extraction for a same extraction time of 2 h (Santos et al., 2010).

The improvement of bioactive compound extraction yield in the ultrasound-assisted extraction depends on the collapse of cavitation bubbles which produces microjets that disrupt plant cell membrane and subsequently provoke the release of phenolic compounds. The degree of yield enhancement depends largely on the cavitation bubbles formed within the solvent. The formation or threshold of cavitation bubble is influenced by the ultrasound frequency and intensity, sonication exposure time on the solvent, physical properties of solvent which include its surface tension, viscosity, and the presence of gas or particulate matters in the solvent (Mason and Lorimer, 2002). Sun et al. (2011) conducted studies to evaluate the effects of various extraction factors, including particle size, extraction solvent, solid/solvent ratio, temperature, extraction time, electrical acoustic intensity, liquid height and duty cycle of ultrasound exposure on the extraction yield of all-*trans*- $\beta$ -carotene from citrus peels.

Although many published studies on the extraction of phenolic compounds from mangosteen using ultrasound-assisted methods are available, i.e., including quantitative and qualitative determination of xanthenes (Ji et al., 2007), fast screening and fractionation of major xanthenes (Destandau et al., 2009), and the effects of drying methods on assay and antioxidant activity of xanthenes (Suvarnakuta et al., 2011), the effect of ultrasound sonication time and amplitude on total phenolic and anthocyanin yield of mangosteen has not been reported. The objective of this study was to optimize the extraction yield of total monomeric anthocyanin (TMA) and total phenolic content (TPC) from mangosteen hull by varying the amplitude and sonication time of a direct ultrasonic treatment using a probe. The best ultrasonic probe conditions were compared with an indirect application of ultrasonic treatment using a bath system.

## 2. Materials and methods

### 2.1. Sample preparation

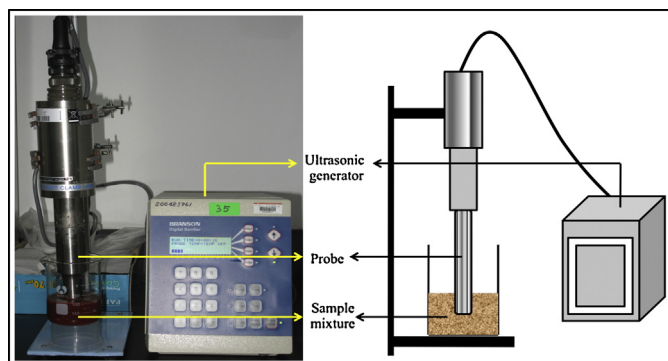
Mangosteen hulls were collected from a private orchard in Sungai Gadut, Negeri Sembilan, Malaysia (2°43' North, 101°53' East) in mid December 2011 and dried under a fan at room temperature following the procedure of Cheok et al. (2012a). One kilogram of dried hull was crushed into powder of particle size lower than 4 mm using a dry crusher (SM100 Comfort, Retsch, Germany). The crushed hull was stored in a freezer (ACF15F, Acson, Malaysia) at –10 °C until the extraction process.

### 2.2. Full factorial design for solvent extraction coupled with ultrasonic treatment

A three-level and two-factor full factorial experiment (Table 1) was designed and created using MINITAB statistical software (Release 14, Minitab Inc., State College, PA, USA) to cover the range

**Table 1**  
Levels in full factorial experiments for direct ultrasonic pre-treatment extraction investigation using probe.

Factor	Level 1	Level 2	Level 3
Sonication time (min)	5	15	25
Amplitude (%)	20	50	80



**Fig. 1.** Experimental setup for ultrasound probe treatment for sample mixture.

of investigated ultrasonic treatment time and amplitude. Sonication time and amplitude were chosen as independent variables and the total monomeric anthocyanins (TMA) and total phenolic content (TPC) were the responses of the design. The effects of ultrasound were tested using two types of extraction solvents, i.e., methanol and ethanol aqueous solvents acidified with 0.20% HCl, because they were found yielding anthocyanins effectively from mangosteen hull in an earlier study (Cheok, 2012). The acidified methanol or ethanol aqueous solvents were prepared by transferring 10 ml of HCl solution with concentration of 0.02 (ml/ml) into a 100-ml volumetric flask which contained 70 ml of methanol or ethanol absolute, that was later topped-up with distilled water until 100 ml.

In preparing mangosteen sample mixture for the extraction process, 5.00 g ( $\pm 0.01$  g) of mangosteen hull powder was mixed with a 100 ml of acidified methanol solvent in a 250 ml-beaker. The sample mixture was pre-treated with ultrasonic waves (Digital Sonifier 450, Branson, CT, USA) using an ultrasonic probe with a 2.54 cm diameter cylindrical titanium alloy head operated at 20 kHz and 100 W (Fig. 1). The tip of the probe was placed at 2 cm depth in the sample mixture and treated randomly following each condition set in the full factorial design. After the ultrasonic pre-treatment, the sample mixture was stirred for 1 h on a stirring hot plate at room temperature using a magnetic stirrer (Cheok et al., 2012b), centrifuged (Centrifuge 5430, Eppendorf, Hamburg, Germany) at 5000 rpm for 10 min, and filtered through a filter paper (Advantec, Toyo Roshi Kaisha Ltd., Japan) with a vacuum pump aspirator (DOA-P604-BN, Cast Manufacturing Inc., USA). The filtrate of each solvent was collected as crude extracts and quantified in triplicate for its total monomeric anthocyanin (TMA) and total phenolic content (TPC).

The full factorial experimental design for each response with each extraction solvent was analyzed using Minitab and expressed in a first order model:

$$Y = \beta_0 + \beta_1 T + \beta_2 A + \beta_{12} TA \quad (1)$$

where  $Y$  is the response variable (TMA or TPC);  $\beta_0$  is an intercept term;  $\beta_1$  and  $\beta_2$  are linear coefficients;  $\beta_{12}$  is interaction coefficients;  $T$  and  $A$  are the independent variables representing the sonication time and amplitude. The coefficient of determination ( $R^2$ ), response surface and contour plots were to examine and evaluate the models.

### 2.3. Total monomeric anthocyanin (TMA) determination

The TMA of the crude mangosteen extract was determined using the pH differential method (Giusti and Wrolstad, 2005). A single beam UV–vis spectrophotometer (DR2800, Hach, USA) was used for spectral measurements at wavelength at 510 and 700 nm with

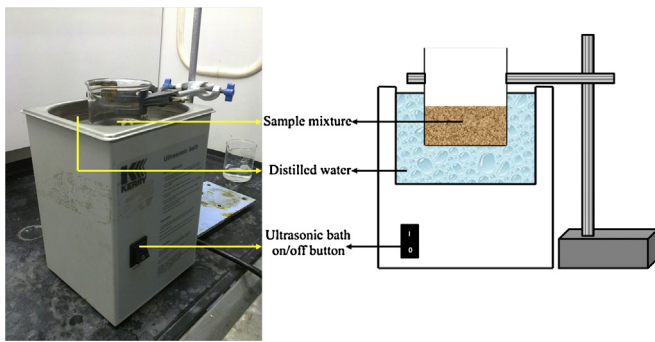


Fig. 2. Experimental setup for ultrasound bath treatment for sample mixture.

distilled water as the blank. The TMA at each full factorial design condition was calculated using following equation:

$$\text{TMA (mg/L)} = \frac{A \times \text{MW} \times \text{DF} \times 1000}{\epsilon \times l} \quad (2)$$

where  $A$  (absorbance) =  $(A_{510} - A_{700})_{\text{pH } 1.0} - (A_{510} - A_{700})_{\text{pH } 4.5}$ ; MW (molecular weight) = 449.2 g/mol for cyanidin-3-glucoside; DF (dilution factor) = 200; 1000 = conversion from g to mg;  $\epsilon$  (molar absorptivity coefficient in L/mol/cm for cyanidin-3-glucoside) = 26,900;  $l$  = path length in cm. The TMA results in mg cy-3-glu/L were then expressed as mg cy-3-glu/g of hull powder by dividing it with solid to solvent ratio 0.05 g/ml.

#### 2.4. Total phenolic content (TPC) determination

The TPC determination from crude extract was measured colorimetrically using the Folin-Ciocalteu (FC) method (Cheok et al., 2012a). The crude extract obtained was diluted with a dilution factor of 200. Then, 1.0 ml of the extract aliquot in triplicates was transferred using a 1 ml transfer pipet into a test tube and then mixed thoroughly with 5.0 ml of FC reagent priorly diluted 1:10 with distilled water. After shaking for 3 min, 4.0 ml of sodium carbonate (7.5%, w/v) was added and mixed thoroughly. The mixtures were then allowed to stand for 30 min in the dark before measuring its absorbance in a single beam UV-vis spectrophotometer (DR2800, Hach, U.S.A) at 765 nm (Singleton and Rossi, 1965) against the blank of methanol or ethanol pure solvents. The TPC values were expressed in mg gallic acid equivalent (GAE)/g hull powder by dividing with solid to solvent ratio of 0.05 g/ml.

#### 2.5. Comparison between direct and indirect ultrasonic pre-treatment

The optimized direct ultrasonic probe conditions in giving the highest extracted TMA and TPC yields with respect to ultrasonic time and amplitude were compared to an indirect ultrasonic bath system. Fig. 2 shows the set-up of the bath system (KC2, Kerry Ultrasonic Bath, England) where the ultrasonic generator is powered at 75 W with operating frequency of 38 kHz. The mangosteen samples were pre-treated following the optimized ultrasonic exposure durations obtained from the direct ultrasonic probe method. The sonication times for extracting TMA using methanol and ethanol were 15 and 5 min respectively, while for TPC extraction, the samples were exposed for duration of 25 min in the ultrasonic bath for both the extraction solvents to obtain the TMA and TPC. The controls are extraction process with conventional stirring process on magnetic stirring hot plate for 1 h. All the experiments including the control were repeated and the measurement of TMA and TPC of each experiment run was conducted in triplicates. The results

are expressed as mean value  $\pm$  standard deviation ( $n=6$ ) and statistically analyzed using Student- $t$ 's distribution and for significant difference if  $p < 0.05$ .

### 3. Results and discussion

#### 3.1. Ultrasonic sonication time and amplitude on TMA yield

The result shows that the ultrasonic sonication time of 15 min and 20% amplitude gave a maximum TMA recovery of  $2.92 \pm 0.04$  mg using methanol, while using the ethanol, maximum TMA recovery of  $2.14 \pm 0.07$  mg was from sonication time of 5 min and 20% amplitude (Table 2). The maximum TMA obtained using methanol was 36.50% significantly higher ( $p < 0.05$ ) than using ethanol at the same amplitude of 20%. This result indicated that methanol is better than ethanol in the recovery of anthocyanins from mangosteen hull at the same amplitude but with a longer sonication time.

The ultrasonic sonication time and amplitude required to achieve a maximum bioactive compound recovery from the plant material vary with factors such as the extraction solvent used, bioactive compound interest, plant material, ultrasonic extraction procedure, etc. A previous study done by Nayak and Rastogi (2011) reported that the optimum ultrasonic probe conditions of the sonication time of 35 min and an amplitude of 10–14% gave a higher anthocyanins recovery from *G. indica* Choisy using the ultrasonic probe extraction. Although the extraction solvent used was not mentioned, the slight difference in amplitude with this study may be due to the different ultrasonic instrument used, extraction procedure, and/or plant material studied. Meanwhile, Chen et al. (2007) found that the optimum ultrasonic probe conditions for extracting anthocyanins from red raspberries were at sonication time of 200 s and ultrasonic power of 400 w. Golmohamadi et al. (2013) observed that the extracted total anthocyanins from red raspberry puree increased by 12.6% at 20 kHz after 10 min sonication, but no further significant increase was recorded after 20 min. Other than anthocyanins, Chua et al. (2009) discovered that ultrasound of 20% amplitude and sonication time of 30 min gave the highest phospholipids extracted from palm-pressed fiber.

From the response surface analysis obtained (Fig. 3), the models are well fitted with linear regression equations for both methanol ( $\text{TMA}_m$ ) and ethanol ( $\text{TMA}_e$ ) with high values of coefficient of determinations ( $R^2$ ):

$$\text{TMA}_m = 1.430 - 0.431T - 0.816A - 0.301TA, R^2 = 0.911 \quad (3)$$

$$\text{TMA}_e = 0.413 - 0.531T - 0.590A + 0.156TA, R^2 = 0.987 \quad (4)$$

Table 2

Effect of sonication time and amplitude on TMA yield from mangosteen hull with full factorial design.

Run order	Sonication time (min)	Amplitude (%)	TMA (mg cy-3-glu/g hull powder)	
			Methanol	Ethanol
1	5	80	$1.67 \pm 0.00$	$0.78 \pm 0.04$
2 <sup>a</sup>	15	20	$2.92 \pm 0.04$	$1.47 \pm 0.07$
3 <sup>b</sup>	5	20	$2.47 \pm 0.07$	$2.14 \pm 0.07$
4	15	50	$1.11 \pm 0.08$	$0.42 \pm 0.04$
5	25	80	$0.45 \pm 0.04$	$0.00 \pm 0.00$
6	5	50	$2.38 \pm 0.04$	$1.07 \pm 0.07$
7	15	80	$0.82 \pm 0.04$	$0.02 \pm 0.04$
8	25	50	$1.05 \pm 0.08$	$0.07 \pm 0.07$
9	25	20	$2.45 \pm 0.10$	$0.74 \pm 0.07$

<sup>a</sup> Optimum conditions yielded maximum TMA using acidified methanol aqueous solvent.

<sup>b</sup> Optimum conditions yielded maximum TMA using acidified ethanol aqueous solvent.

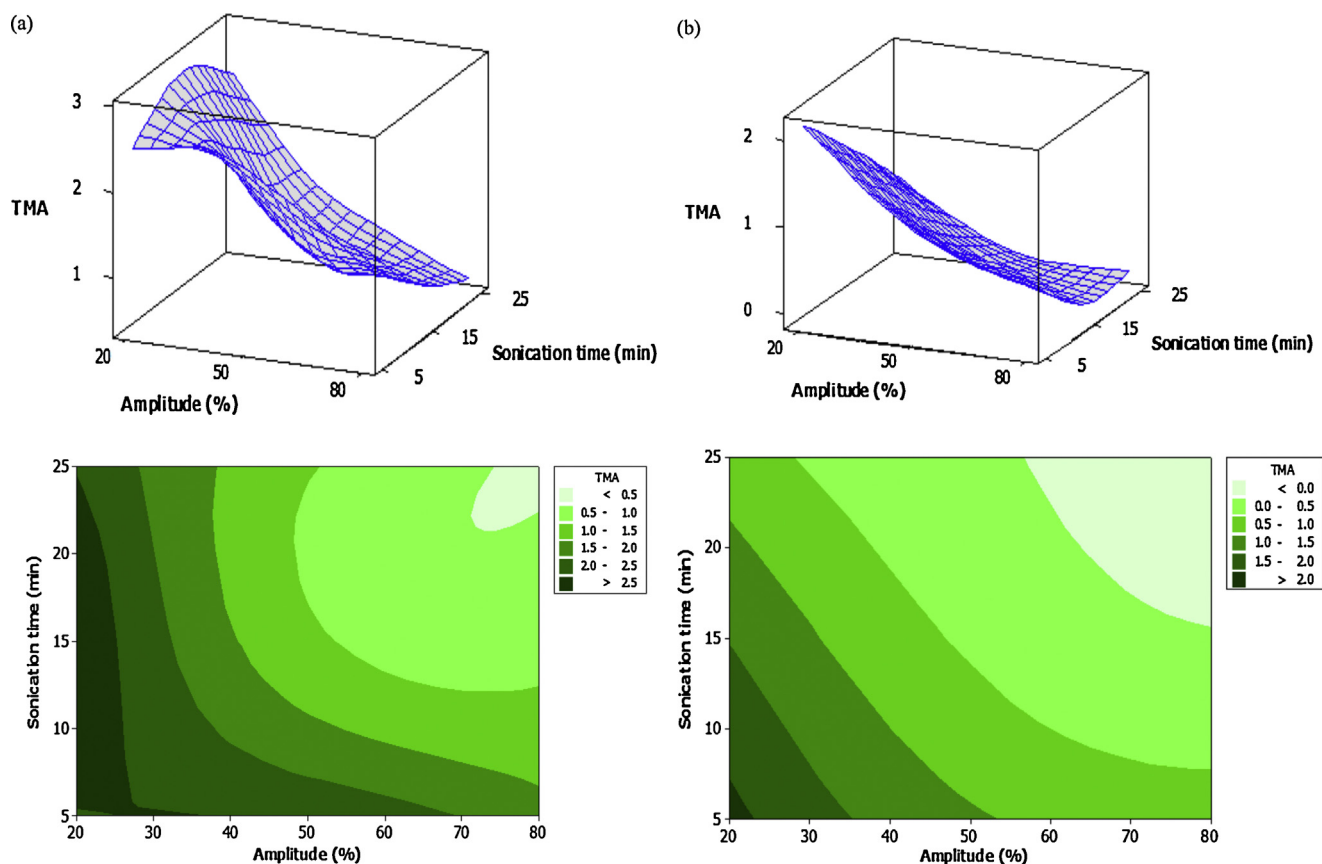


Fig. 3. Response surface and contour plots showing the effect of sonication time (min) and amplitude on TMA (mg cy-3-glu/g hull powder) extracted from mangosteen hull using acidified (a) methanol and (b) ethanol, aqueous solvents.

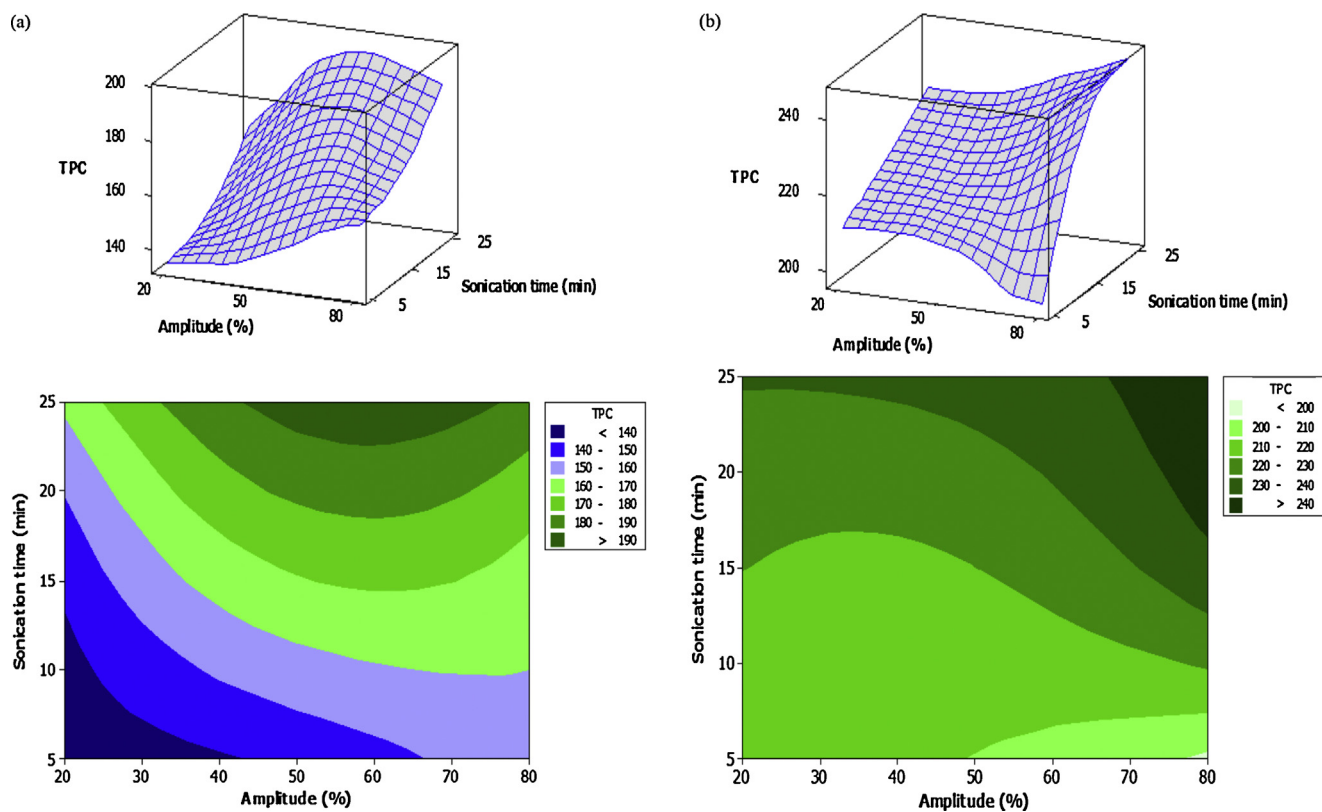


Fig. 4. Response surface and contour plots showing the effect of sonication time (min) and amplitude on TPC (mg GAE/g hull powder) yield extracted from mangosteen hull using acidified (a) methanol and (b) ethanol, aqueous solvents.

Both the regression equations show that ultrasonic amplitude has a significant ( $p < 0.05$ ) effect on the TMA. No significant effect ( $p > 0.05$ ) of sonication time on TMA when using methanol while there was significant effect ( $p < 0.05$ ) using ethanol. The interaction between amplitude and sonication time did not have a significant effect ( $p > 0.05$ ) for both equations.

### 3.2. Ultrasonic sonication time and amplitude on TPC yield

Table 3 shows that the maximum TPC of  $193.91 \pm 7.52$  mg was achieved at 50% amplitude using methanol, which was 26.75% significantly lower ( $p < 0.05$ ) than the maximum TPC recovery of  $245.78 \pm 9.86$  mg using ethanol at 80% amplitude with a same sonication time of 25 min. In contrast with the TMA yield, ethanol was found more preferable in the recovery of TPC than methanol although a higher amplitude was required.

The response surface analysis show that the TPC yield obtained from both methanol (TPC<sub>m</sub>) and ethanol (TPC<sub>e</sub>) were fitted well ( $p < 0.05$ ) into first order linear regression equations with a high coefficient of determinations (Fig. 4):

$$\text{TPC}_m = 165.779 + 18.002T + 11.937A + 0.461TA, R^2 = 0.936 \quad (5)$$

$$\text{TPC}_e = 224.028 + 15.348T + 3.108A + 6.960TA, R^2 = 0.922 \quad (6)$$

For both models, sonication time has a significant ( $p < 0.05$ ) effect on TPC yields, while interaction between sonication time and amplitude was not significant ( $p > 0.05$ ). Ultrasonic amplitude displayed a significant effect ( $p < 0.05$ ) on the TPC using methanol while there was no significant effect ( $p > 0.05$ ) when using ethanol.

In comparison with the TMA yield, the extraction of TPC from mangosteen hull required longer sonication time and higher amplitude regardless of the extraction solvent used. This observation is in agreement with Golmohamadi et al. (2013) where they discovered that TPC and TMA were significantly increased by 11.97% and 12.6% after 30 and 20 min sonication at 20 kHz and 400 W ultrasonication conditions. However, Portto et al. (2013) obtained a maximum yield of TPC and TMA after 15 min ultrasonication at 20 kHz and 150 W in their extraction of grape seeds. Anthocyanins is preferably extracted at a lower amplitude and this is most probably due to its sensitivity to ultrasonic amplitude power. This observation is in agreement with the previous finding by Nayak and Rastogi (2011) where they found that an amplitude of 10–14% was optimum for extracting anthocyanins from *G. indica* Choisy although a longer extraction time was needed. Carrera et al. (2011), however reported contradicting results of a higher amplitude resulting in a higher total phenolics and anthocyanins recovery from grapes where they were evaluated using a single-factor experimental design.

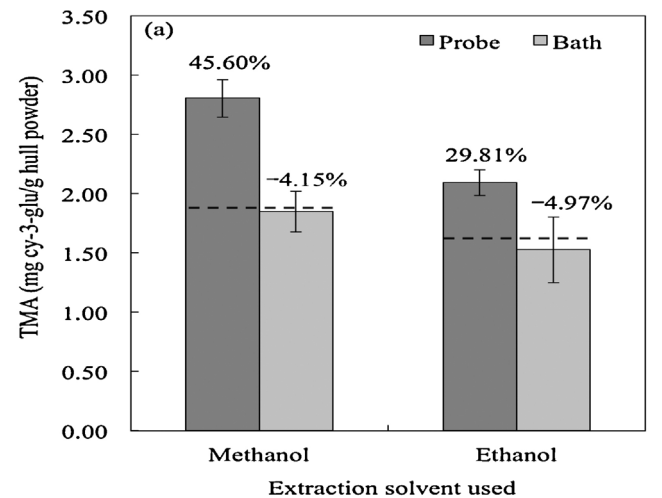
**Table 3**

Effect of sonication time and amplitude on TPC yield from mangosteen hull with full factorial design.

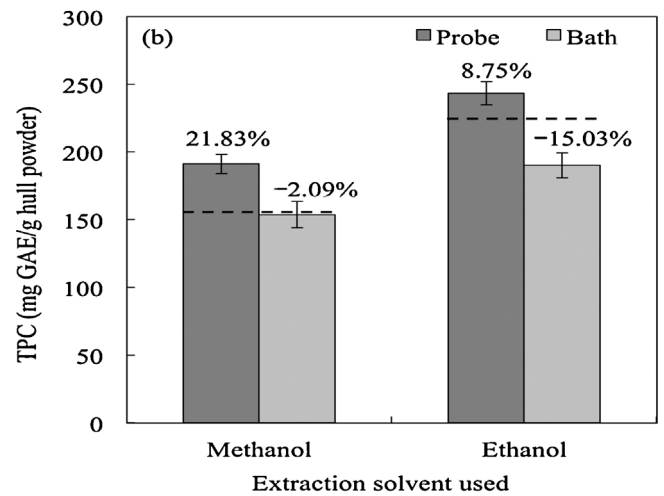
Run order	Sonication time (min)	Amplitude (%)	TPC (mg GAE/g hull powder)	
			Methanol	Ethanol
1	5	80	$157.87 \pm 11.08$	$198.16 \pm 2.28$
2	15	20	$142.10 \pm 4.60$	$220.21 \pm 13.63$
3	5	20	$134.73 \pm 4.19$	$211.01 \pm 5.51$
4	15	50	$169.16 \pm 10.90$	$219.96 \pm 3.61$
5 <sup>b</sup>	25	80	$187.01 \pm 5.75$	$245.78 \pm 9.86$
6	5	50	$142.33 \pm 4.25$	$209.75 \pm 6.88$
7	15	80	$165.69 \pm 6.48$	$236.71 \pm 2.18$
8 <sup>a</sup>	25	50	$193.91 \pm 7.52$	$234.44 \pm 9.72$
9	25	20	$162.02 \pm 2.83$	$230.79 \pm 4.35$

<sup>a</sup> Optimum conditions yielded maximum TPC using acidified methanol aqueous solvent.

<sup>b</sup> Optimum conditions yielded maximum TPC using acidified ethanol aqueous solvent.



Dotted lines represent controls for:  
Methanol:  $1.93 \pm 0.10$  mg cy-3-glu/g hull powder  
Ethanol:  $1.61 \pm 0.22$  mg cy-3-glu/g hull powder



Dotted lines represent controls for:  
Methanol:  $156.95 \pm 8.65$  mg GAE/g hull powder  
Ethanol:  $223.80 \pm 8.40$  mg GAE/g hull powder

**Fig. 5.** Comparison of ultrasonic probe and bath on (a) TMA and (b) TPC yield.

### 3.3. Comparison of ultrasonic probe and bath with conventional magnetic stirring extraction on TMA and TPC yields from mangosteen hull powder

The optimum direct ultrasonic pre-treatment conditions using the probe for maximum TMA and TPC yields were compared with an in-direct pre-treatment using a bath and the conventional magnetic stirring extraction (control). The result shows that TMA extracted with direct ultrasonic was significantly higher ( $p < 0.05$ ) than both the bath and the control methods in methanol and ethanol solvents (Fig. 5a). For TMA, it was a significant increase by 45.60% ( $p < 0.05$ ) from control of  $1.93 \pm 0.10$  mg while for ethanol, it was 29.81% ( $p < 0.05$ ) from control of  $1.61 \pm 0.22$  mg. Considering the higher percentage of the TMA yield enhanced using methanol than ethanol, the choice of solvent could be the determinant factor when it comes to extraction yield of bioactive compound. In comparing both controls alone, methanol also gave a higher TMA yield by 19.88% ( $p < 0.05$ ) compared to ethanol. This indicates that the methanol is a preferable solvent for TMA recovery from mangosteen hull. The indirect pre-treatment using the ultrasonic bath did

not give improvement in the extraction of TMA for both solvents used.

For extracted TPC, the direct ultrasonic pre-treatment using probe resulted significant higher ( $p < 0.05$ ) yields by 21.83% ( $p < 0.05$ ) for the methanol and 8.75% for ethanol from controls of  $156.95 \pm 8.65$  mg and  $223.80 \pm 8.40$  mg, respectively. Similarly, the indirect ultrasonic pre-treatment using the bath did not provide any positive improvements such that the controls even had better TPC yields and ethanol was more efficient by 42.59% than methanol.

The improvement of TMA and TPC extractions from mangosteen hull powder with direct ultrasonic pre-treatment over the indirect method of using bath has proven. The advantage of using the probe compared to the bath has also been reported in the extraction of oleuropein from olive fruit (Jerman et al., 2010). They revealed that the optimized ultrasonic probe conditions of the three extraction steps of 20 min, 44 °C and pure methanol was more efficient in comparison to the ultrasonic bath and agitation, with up to 33% and 80% enhancement in the extraction of oleuropein from olive fruit.

The efficacy of ultrasonic-assisted extraction is not only shown through the improved bioactive compound yield recovery but also helps in reducing the extraction time. In this mangosteen study, the extraction time has been shortened to 1 h from 20 h (Cheok et al., 2012a) and 2 h (Cheok et al., 2012b). A supporting study on this is on the extraction of total phenolics from *D. regia* tree flowers (Adjé et al., 2010) where although the total phenolics yields obtained by the two extraction methods of ultrasonic-assisted bath extraction with the mechanical stirring method have no significant difference, the ultrasonic bath did tremendously reduce the extraction time from 3 to 1 h.

#### 4. Conclusions

With the aid of full factorial design, the optimized sonication time of 15 min and 20% amplitude was found giving the highest TMA recovery, while 25 min and 80% amplitude gave the highest TPC recovery. These optimized conditions using direct probe ultrasonic treatment have enhanced TMA and TPC yields by 45.6% and 8.8% respectively compared to the non-sonicated control. The ultrasonic treatment is more effective in improving anthocyanins extraction than the phenolics contents and it is thus recommended for use in extraction of anthocyanins from mangosteen hull powder in the industry.

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