



Review Article

A Review: Extraction Methods of Phenolic Compounds from Rambutan Peel (*Nephelium lappaceum* L.)

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Abstract: Rambutan is a tropical fruit native to the ASEAN countries well-known for its refreshing flavour. However, due to the fresh consumption and its short shelf life, the peels are usually discarded as waste in a huge amount. Efforts have been made to reduce the amount of waste by optimally utilising to achieve sustainable development by utilising the peel for industrial applications. The rambutan peels contain a significant number of antioxidants due to the beneficial and nutritive phenolic compounds present. Rambutan peel extract possesses antioxidant, antidiabetic, anti-obesity, antiproliferative, antimicrobial and anticancer properties thus, can be used for application in the food, pharmaceutical and cosmetics industries. An extraction process is needed to isolate the phenolic compounds from the rambutan peel. Factors such as polarity of solvents, cost, extraction efficiency and extraction time need to be considered in the selected method as it will be implemented in industries. Nevertheless, no review paper has focused on the most suitable extraction method of rambutan peel that possibly can be adopted in the industry. This review paper summarises available extraction methods used to extract the phenolic compounds from rambutan peel and determines the most suitable extraction method that may be potentially used in industries. From the literature, the ultrasound-assisted extraction (UAE) method is the most potent method to be applied in the industry.

Keywords: rambutan; rambutan peel; phenolic compound, extraction methods; ultrasound-assisted extraction (UAE)

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1. Introduction

Rambutan (*Nephelium lappaceum L.*) is a tropical fruit belonging to the Sapindaceae family and it can be found massively in ASEAN countries such as Malaysia, Thailand, and Indonesia (Li *et al.*, 2018). Since rambutan is a seasonal fruit, an abundant amount has been produced during its season which falls in December (main season) and August (second season) resulted in the increase number of total polyphenol compounds in rambutan seed and peel will be different due to numerous factors such as the type of solvent, solvent polarity, extraction methods and parameters, liquid-solid ratio and sample particle size (Tingting *et al.*, 2022). Previous research has been conducted to utilise rambutan peel wastage in industrial applications as one of the best solutions to overcome the wastage issue. The utilisation of rambutan peel is not only limited to environmental conservation but most importantly the phenolic compounds extracted from the peel can be used by industries.

The extraction of phenolic compounds from various materials is gaining popularity because of their chemical and structural diversity and complexity, making the extracting, identifying, and characterisation of new phenolics from various plant-based materials difficult (Suleria et al., 2020). The yield of each phenolic component extracted from fruit byproducts is determined by the extraction method used. As a result, extraction is the most crucial stage in any study involving bioactive substances from plants, and it has a direct impact on the study's outcome (Phuong et al., 2020b). Over the last 50 years, nonconventional methods such as ultrasonic-assisted extraction (UAE), supercritical fluid extraction (SFE), and microwave-assisted extraction (MAE) have been developed to be more environmentally friendly with the reduced usage of synthetic and organic chemicals, and short extraction time thus improving the extraction yield and quality (Tingting *et al.*, 2022); López-Bascón & Luque de Castro, 2020). On the contrary, the conventional method requires a large volume of solvents and a long extraction time. The UAE and MAE are some of the greener extraction methods to extract phenolic compounds from rambutan peel compared to conventional methods such as maceration and Soxhlet extraction (López-Bascón & Luque de Castro, 2020; Mahmood et al., 2018; Phuong et al., 2020b).

The extraction method is used to obtain and separate the desired phenolic compounds in rambutan peel for further analysis. The rambutan peel will be pulverised using a miller, mortar, or sieve before extraction to achieve a smaller particle size and hence a better extraction result. The small particle size improves extraction efficiency by increasing solvent penetration and solute diffusion (Lavilla & Bendicho, 2017). However, too fine particle size will result in increased solute absorption in the solid. Consequently, it will result in difficulty

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during filtering. Thus, it is suggested that the particle size of rambutan peel be around 420 μ m 40 mesh sieve (Boyano-Orozco *et al.*, 2020). Several studies have reported that ellagic acid, corilagin, and gallic acid are the major phenolic compounds contained in rambutan peel extract and geraniin is the dominant compound (Rakariyatham *et al.*, 2020; Thitilertdecha *et al.*, 2010). Since phenolic compounds are known for their high antioxidant activities, thus this property can be applied to delay oxidation and longer the shelf life. In rubber products, antioxidants are required to be added to natural rubber to enhance its ageing properties as natural rubber is susceptible to oxidative degradation by extending the service life of natural rubber products by preventing oxidative ageing (Sukatta *et al.*, 2021). Moreover, rambutan peel also has the potential to replace the synthetic antioxidant used in rubber.

This review will provide insights into available extraction methods that have been used to extract phenolic compounds from rambutan peel, including the determination of the most suitable extraction method to be adopted in industries in terms of solvent polarity, low cost and less extraction time. Additionally, this review enlightens on the advantages and disadvantages of each extraction method mentioned above.

2. Available Extraction Methods for Rambutan Peel Phenolic Compounds

Phenolic compound extraction is a crucial process that depends on the solvent used, extraction time and temperature, and the chemical nature of the sample together with the extraction technique (Monrroy *et al.*, 2020). Various extraction techniques have been applied to explore these phenolic compounds in rambutan peel. The phenolic compounds found in rambutan peel extract based on the extraction method used are presented in Table 1.

Extraction method	Phenolic compounds found	Extraction condition	Detection technique used	References, country
Maceration (Conventional)	Geraniin (74.78 g)	Ethanol extraction (1:15; w/v) was carried out at room temperature for 24 h in an orbital shaker.	Reverse- phase HPLC and LC/MS	(Perera <i>et al.</i> , 2012), Malaysia
	Geraniin (568.0 mg/g) Corilagin (71.9 mg/g) Ellagic acid (53.5 mg/g)	Extracted with ethyl ether three times, the residue was then extracted with methanol three times and finally with water three times.	Column chromatograp hy $(4 \times 55$ cm) and HPLC	(Thitilertdech a <i>et al</i> ., 2010), Thailand

Table 1. Total phenolic compounds found in rambutan peel extract based on the extraction method.

Extraction

method

Phenolic compounds found

Extraction condition	Detection technique used	References, country
racted with nol 40%, (1:15;) at room perature for 12 h dark room.	Aluminium colourimetric method.	(Gusman & Tsai, 2015), Taiwan
tilled water. (1:5:	HPLC/ESI/M	(Hernández-

	used		
Quercetin (7.74±0.53 mg/g)	Extracted with ethanol 40%, (1:15; w/v) at room temperature for 12 h in a dark room.	Aluminium colourimetric method.	(Gusman & Tsai, 2015), Taiwan
Apigenin (269 m/z) Pelargonidin (270 m/z) Brevifolin carboxylic acid (291 m/z) Ellagic acid (301 m/z) p-Coumaroyl glucose (325 m/z) Vanillic acid hexoside (329 m/z) Ellagic acid pentoside (433 m/z) Vitisin A (560 m/z) Apigenin arabinoside- glucoside (563 m/z) Corilagin (633 m/z) Castalagin/Vescalagin (933 m/z) Galloyl-bis-HHDP- hexoside (Casuarinin) (935 m/z) Geraniin (951 m/z) Unknown (979 m/z)	Distilled water, (1:5; w/v) at extraction temperature of 60°C for 30 min.	HPLC/ESI/M S	(Hernández- Hernández <i>et</i> <i>al.</i> , 2017), Mexico
Ellagic acid (1.6±0.03 g/100 g) Gallic acid (17.3 g/100 g) Catechin (3.2 g/100 g)	50% ethanol, extraction temperature of 140°C for 35 min.	HPLC	(Venturini <i>et al.</i> , 2018), Spain
Geraniin (504.41mg/g) Corilagin (117.24 mg/g) Ellagic acid (19.50 mg/g) Gallic acid (2.28 mg/g)	Methanol, extraction time of 16 h.	HPLC	(Sukatta <i>et al.</i> , 2021), Thailand
Geraniin (146.3 mg/g) Corilagin (69.1 mg/g) Ellagic acid (25.91 mg/g) Gallic acid (3.2 mg/g)	Ethanol, mass/volume ratio of 1:5, extraction time of 1 h.	HPLC	(Chollakup <i>et al.</i> , 2020), Thailand
	mg/g) Apigenin (269 m/z) Pelargonidin (270 m/z) Brevifolin carboxylic acid (291 m/z) Ellagic acid (301 m/z) p-Coumaroyl glucose ($325 m/z$) Vanillic acid hexoside ($329 m/z$) Ellagic acid pentoside ($433 m/z$) Vitisin A ($560 m/z$) Apigenin arabinoside- glucoside ($563 m/z$) Corilagin ($633 m/z$) Castalagin/Vescalagin ($933 m/z$) Galloyl-bis-HHDP- hexoside (Casuarinin) ($935 m/z$) Geraniin ($951 m/z$) Unknown ($979 m/z$) Ellagic acid ($1.6\pm0.03 g/100 g$) Gallic acid ($17.3 g/100 g$) Catechin ($3.2 g/100 g$) Geraniin ($504.41mg/g$) Corilagin ($117.24 mg/g$) Ellagic acid ($19.50 mg/g$) Gallic acid ($12.28 mg/g$) Geraniin ($146.3 mg/g$) Corilagin ($69.1 mg/g$) Ellagic acid ($25.91 mg/g$)	mg/g)ethanol 40%, (1:15; w/v) at room temperature for 12 h in a dark room.Apigenin (269 m/z)Distilled water, (1:5; w/v) at extraction temperature of 60°C for 30 min.Brevifolin carboxylic acid (291 m/z)Distilled water, (1:5; w/v) at extraction temperature of 60°C for 30 min.Ellagic acid (291 m/z)Distilled water, (1:5; w/v) at extraction temperature of 60°C for 30 min.Ellagic acid (291 m/z)Distilled water, (1:5; w/v) at extraction temperature of 60°C for 30 min.Yanillic acid hexoside (329 m/z)Distilled water, (1:5; w/v) at extraction temperature of 60°C for 30 min.Ellagic acid (291 m/z)Vanillic acid hexoside (329 m/z)Ellagic acid pentoside glucoside (563 m/z)SoftCorilagin (633 m/z)Castalagin/Vescalagin (933 m/z)Castalagin/Vescalagin (933 m/z)SoftGeraniin (951 m/z)Unknown (979 m/z)Ellagic acid (1.6±0.03 g/100 g)Soft ethanol, extraction temperature of 140°C for 35 min.Gallic acid (17.3 g/100 g)Methanol, extraction time of 16 h.mg/g)Ellagic acid (19.50 mg/g) Gallic acid (2.28 mg/g)Geraniin (146.3 mg/g) Corilagin (69.1 mg/g)Ethanol, mass/volume ratio of 1:5, extraction time of 1 h.	Quercetin (7.74±0.53 mg/g)Extracted with ethanol 40%, (1:15; w/v) at room temperature for 12 h in a dark room.Aluminium colourimetric method.Apigenin (269 m/z) Pelargonidin (270 m/z) Brevifolin carboxylic acid (291 m/z)Distilled water, (1:5; w/v) at extraction temperature of 60°C for 30 min.HPLC/ESI/M SEllagic acid (301 m/z) p-Coumaroyl glucose (325 m/z)Distilled water, (1:5; wov) at extraction temperature of 60°C for 30 min.HPLC/ESI/M SEllagic acid (301 m/z) p-Coumaroyl glucose (325 m/z)Distilled water, (1:5; wov) at extraction temperature of 60°C for 30 min.HPLC/ESI/M SEllagic acid pentoside (433 m/z)Corilagin (633 m/z) Castalagin/Vescalagin (933 m/z) Galloyl-bis-HHDP- hexoside (Casuarinin) (935 m/z)So% ethanol, ethanol, ethanol, ethanol, ethanol, extraction temperature of 140°C for 35 min.HPLCEllagic acid (17.3 g/100 g)Methanol, extraction time of 16 h.HPLCGeraniin (504.41mg/g) Ellagic acid (19.50 mg/g)Methanol, extraction time of 16 h.HPLCGeraniin (146.3 mg/g) Ellagic acid (2.28 mg/g)Ethanol, mass/volume ratio of 1:5, extraction time of 1 h.HPLC

Extraction

method

Phenolic compounds found

Extraction	Detection	References,
condition	technique	country
	used	
anol 80.85%,	UPLC-QQQ-	(Li <i>et al</i> .,
action time of	MS	2018),
39 sec, and the		China
o of liquid to		
d of 24.51:1		

			usea	
Microwave- assisted extraction, MAE (Non- conventional)	Geraniin (140.02 mg/g) Corilagin (7.87 mg/g)	Ethanol 80.85%, extraction time of 58.39 sec, and the ratio of liquid to solid of 24.51:1	UPLC-QQQ- MS	(Li <i>et al.</i> , 2018), China
	Geranin (122.18 mg/g) Catechin (9.80 mg/g) Ellagic acid (9.31 mg/g) Corilagin (7.56 mg/g) Gallic acid (0.69 mg/g) Quercetin 3-O-b- glucoside (0.64 mg/g) Rutin (0.16 mg/g)	Ethanol 80.85%, extraction time of 58.39 sec, microwave frequency of 2450 MHz and the ratio of solid: liquid of 1:24.51	UPLC-Q- Orbitrap-MS ²	(Zhuang <i>et</i> <i>al.</i> , 2017), China
	p-Coumaric acid ($19.44\pm1.04 \text{ mg/g}$) Syringic acid ($16.86\pm0.98 \text{ mg/g}$) Rutin (9.29 ± 0.91 mg/g) Vanillin (5.18 ± 0.32 mg/g) Catechin (4.90 ± 0.41 mg/g) Chlorogenic acid (3.66 ± 0.06 mg/g) Protocatechuic acid (3.24 ± 0.13 mg/g), Gallic acid (3.03 ± 0.11 mg/g) Benzoic acid (1.80 ± 0.05 mg/g) Salicylic acid (1.74 ± 0.03 mg/g)	Ethanol 80.85%, extraction time of 58.39 sec, microwave frequency of 2450 MHz, and the ratio of solid to liquid of 1:24.51	Agilent 1200 HPLC system with a photodiode array detector equipped with an autoinjector and reversed- phase HPLC	(Sun <i>et al.</i> , 2012), China
	Catechin (118.26±4.57 mg/g)	Ethanol 60%, (1:20; w/v), extraction time of 3 min with a frequency of 2450 MHz	Modified colourimetric method.	(Chaiwarit <i>et al.</i> , 2021), Thailand
Ultrasound- assisted extraction, UAE	Geraniin (397.28±6.21 mg/g) Ellagic acid (176.99±0.39 mg/g)	Methanol 80%, room temperature for 20 min.	HPLC	(Phuong <i>et al.</i> , 2020a), Vietnam

Extraction method	Phenolic compounds found	Extraction condition	Detection technique used	References, country
(Non- conventional)	Quercetin (167.37±9.80 mg/g) Rutin (36.40±1.67 mg/g)			
	Geraniin $(397\pm9.5 \text{ mg/g})$ Ellagic acid $(177\pm0.4\text{mg/g})$ Quercetin $(167\pm9.8\text{mg/g})$ Rutin $(36.4\pm1.7 \text{ mg/g})$ Corilagin $(3.81\pm0.7 \text{ mg/g})$	Methanol 80%, room temperature for 20 min.	HPLC	(Phuong <i>et</i> <i>al.</i> , 2020b), Vietnam
	Gallic acid, Brevifolin carboxylic acid, Ellagic acid, Gallic acid 3-0- gallate, Isorhamnetin 3-0- glucoside 7-0- rhamnoside, Galloyl- HHDP-hexoside, Corilagin, Pedunculagi, Theaflavin 3,3'-0- digallate, Galloyl-bis- HHDP-hexoside, Geraniin	Ethanol 10%, mass/volume ratio 1:7, room temperature extraction time 10 min.	Negative ionisation as MS operating conditions by HPLC/ESI/M S	(Mendez- Flores <i>et al.</i> , 2018), Mexico
	Rutin (104±1.13 mg/100g) Cyanidin-3-O- glucoside (10.26±0.39 mg/100 g)	Distilled water, extraction temperature of 50°C, ultrasound power of 20 W, extraction time of 20 min and solid-liquid ratio of 1:18.6 g/mL.	Aluminium chloride assay method.	(Maran <i>et al.</i> , 2017), India

Extraction method	Phenolic compounds found	Extraction condition	Detection technique used	Reference country
	Quercetin (76±2 mg/g)	Ethanol 60%,	AlCl ₃	(Monrroy e
	Cyanidin-3-O-	mass/volume ratio	colourimetric	al., 2020)
	glucoside (0.57 ± 0.03	of 1:10, extraction	method, pH	Panama
	mg/g)	time of 20 min at	differential	
	5-Methylfuran-2-	room temperature.	method and	
	carbaldehyde (109		GC/MS	
	m/z),		analysis.	
	1,2-Benzenediol (64			
	m/z),			
	Catechin (52 m/z),			
	3-Hydroxybenzoic			
	acid (121 m/z),			
	Hexadecanoic acid or			
	palmitic acid (60 m/z),			
	Oleic acid methyl ester			
	(182 m/z),			
	(9Z,12Z)-Octadeca-			
	9,12-dienoic acid or			
	linoleic acid (81 m/z),			
	9-Octadecenoic acid or			
	elaidic acid (73 m/z),			
	Beta-tocopherol or			
	vitamin E (151 m/z)			
	2,4-Di-tert-			
	butylphenol (206 m/z)			
	Isobutyl octyl			
	phthalate (57 m/z),			
	Hexadecanoic acid,			
	methyl ester (87 m/z),			
	Hexadecanoic acid,			
	ethyl ester (101 m/z),			
	Octadecanoic acid,			
	methyl ester or methyl			
	stearate			
	(87 m/z),			
	Oleic acid, ethyl ester			
	(55 m/z),			
	Octadecanoic acid,			
	ethyl ester (101 m/z)			

HPLC, high-performance liquid chromatography; LC/MS, liquid chromatography-mass spectrophotometry; UPLC-QQQ-MS, ultra-high performance liquid chromatography coupled with triple, quadruple mass spectrometry; UPLC-Q-Orbitrap-MS², Ultimate 3000 Series UPLC system (Thermo Scientific)-Q-Exactive hybrid quadrupole-orbitrap mass spectrometer; HPLC/ESI/MS, high-performance liquid chromatography-electrospray ionisation-mass spectrometry; GC/MS, gas chromatography-mass spectrometry

3. Extraction Methods

3.1 Maceration

The simplest and basic extraction method requires the powder sample to be immersed in a solvent and allowed to be in a closed system for a certain amount of time. A separation method is used to separate the solid components from the solvent after the extraction. This procedure is generally carried out through filtration, decantation, or clarification.

For this type of extraction, the highest amount of geraniin was found with a total amount of 74.78 g from 362 g of ethanolic crude rambutan peel extract with optimised extraction conditions as follows; solid-solvent ratio 1:15, room temperature and, 24 h of extraction time (Perera et al., 2012). Reverse-phase chromatography was used to identify the geraniin compound in the extract obtained. This study has performed a large-scale purification of geraniin, and this explained the reason for the high amount of geraniin obtained. Furthermore, a study by Thitilertdecha et al. (2010) has reported, that for 1 g of methanolic extract of rambutan peel, 568.0 mg of geraniin, 71.9 mg of corilagin and 53.5 mg of ellagic acid were obtained. A quercetin compound also has been detected in a rambutan peel extract done by Gusman and Tsai (2015) with an amount of 7.74±0.53 mg/g by using 40% ethanol as extraction solvent with a mass/volume ratio of 1:15 at room temperature for 12 h in a dark room. A total of 13 phenolic compounds were found in the rambutan peel extract obtained at an optimised condition of distilled water (100 mL) as extraction solvent, w/v ratio 1:5, extraction temperature at 60°C and 30 min extraction time (Hernández-Hernández et al., 2017). This study also has proved the existence of geraniin as the major compound in rambutan peel extract from the Mexican rambutan variety using HPLC/ESI/MS analysis (Figure 1).

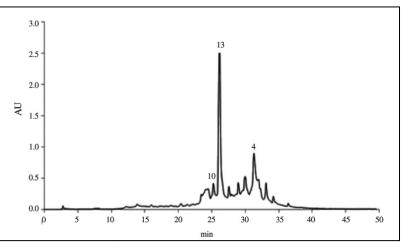


Figure 1. Main phenolic compounds found in rambutan peel using HPLC/ESI/MS analysis; Corilagin (10), Geraniin (13), and (4) Ellagic Acid (Hernández-Hernández *et al.*, 2017)

From all these previous studies, a shorter extraction time can be achieved by using the maceration techniques by increasing the extraction temperature due to increased solubility and diffusion of the solvents and rambutan peel. Methanol, ethanol, and distilled water are the types of polar solvents used as extraction solvents. A polar solvent is the best extraction solvent to extract phenolic compounds because of the high solubility of polyphenols in such solvents. A study by Nawaz *et al.* (2020) has shown that using a polar solvent instead of a non-polar solvent increased the extraction yield, the antioxidant activity and reduced the properties of phenolic compounds due to the high affinity of antioxidant compounds. Hydroethanolic was used instead of pure ethanol as the addition of water into ethanol increases the polarity and thus reduces the ratio of water in the mixture of water and ethanol, resulting in a higher yield of phenolic content (Chaiwarit *et al.*, 2021). Moreover, this type of solvent allows for the extraction of both polar and semipolar compounds (Monrroy *et al.*, 2020). The reason why the different concentrations of ethanol or methanol used in the studies are due to the variance of phenolic compounds' polarity found in rambutan peel such as geraniin (polar compound), ellagic acid (non-polar) and corilagin (polar and non-polar).

Although the maceration method is widely used and easy to handle because of its simplicity, maceration requires a long extraction time (30 min to 24 h), a prolonged process, and low extraction efficiency that limits its usage (Chaiwarit *et al.*, 2021). Besides that, from the previous study, more solvent (refer to Table 1 for maceration method) is needed when using this type of extraction, and this will incur more costs. This method will not be suitable for industrial applications as the industry requires a fast process because the industry needs to produce a large volume of products in a short time.

3.2 Soxhlet Extraction

Soxhlet extraction involves weighing a small amount of dry material in a thimble and then placing it in a distillation flask with the extraction solvent. The thimble-holder solution is aspirated with a syphon when it reaches an overflow level, and the solution is returned to the distillation flask via the syphon as illustrated in Figure 2. This solution is used to dissolve the extracted solutes in the bulk liquid (López-Bascón & Luque de Castro, 2020). The solute is kept in the flask while the solvent is returned to the solid bed of the plant, and this procedure is repeated until the extraction is completed.

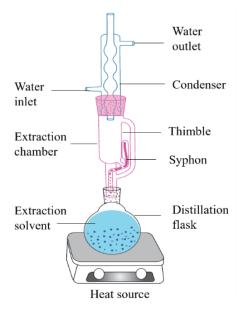


Figure 2. Illustration of Soxhlet Extraction method

For Soxhlet extraction, the highest amounts of gallic acid, catechin, and ellagic acid were recorded by Venturini et al. (2018) with a total of 17.3 g/100 g, 3.2 g/100 g and 1.6 ± 0.03 g/100 g, respectively. The rambutan peel was extracted under an optimised condition of 50% ethanol, and an extraction temperature of 140°C for 35 min. From this study, it is proven that increasing the water contents in ethanol resulted in higher contents of phenolic compounds as shown in Table 1. In other words, the more polar the solvent used, the higher the extraction yield achieved. However, a study done by Kamaludin et al. (2016) obtained the highest antioxidant activity with a yield of 77.21% from ethanol extraction followed by acetone extraction (74.635%), aqueous extraction (74.63%) and methanol extraction (55.56%). Although aqueous and methanol solvents have higher polarity compared to ethanol, ethanol extraction showed better extraction yields due to different types of phenolic compounds present having a different affinity towards the polar solvent. The solvent type, solvent polarity, and phenolic solubility in the particular solvent would influence the recovery of antioxidants from rambutan peels (Kamaludin et al., 2016). As mentioned before, not all phenolic compounds in rambutan peel are polar compounds which makes the solubility decrease when the polar solvent is used thus affecting the extraction. This study also included the effect of extraction temperature and time (Figure 3) on the antioxidant activity of rambutan peel extract.

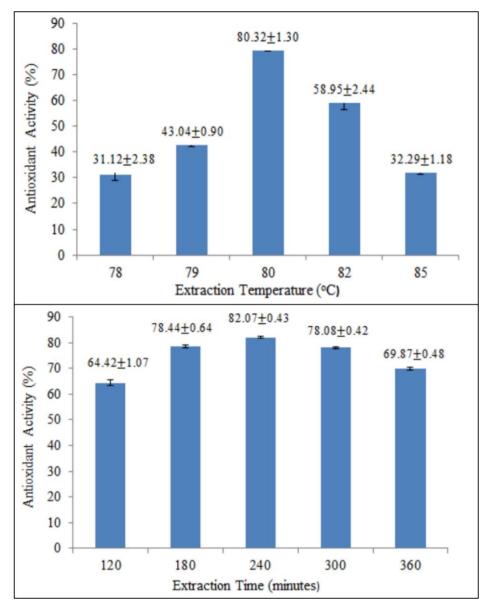


Figure 3. Effect of extraction temperature and time on antioxidant activity (Kamaludin et al., 2016)

From Figure 3, when the extraction temperature reaches 80°C, the antioxidant activity increases significantly to a maximum of 80.32±1.30% and starts to decrease drastically when the temperature exceeds 80°C. This indicated that a too-high temperature would affect the antioxidant activity due to the possible thermal decomposition of phenolic compounds. The same results were obtained for extraction time as the antioxidant activity decreased after 240 minutes. This indicated that excessive duration would also lead to the deterioration of phenolic compounds as they were more exposed to light and oxygen during the extraction process.

Sukatta *et al.* (2021) have recorded a total amount of geraniin and corilagin of 504.41 mg/g and 117.24 mg/g, respectively. In this study, the rambutan peel extract was obtained by

using methanol as an extraction solvent for 16 h and then, quantified using high-performance liquid chromatography (HPLC). Other than geraniin and corilagin compounds, ellagic acid and gallic acid were also detected in the Sukatta *et al.* (2021) study with a record of 19.50 mg/g and 2.28 mg/g, respectively. Since a longer extraction time was used in this study, the amounts of geraniin and corilagin recovered were higher compared to a study by Chollakup *et al.* (2020) that only recovered about 146.3 mg/g of geraniin and 69.1 mg/g of corilagin as one-hour extraction was applied to extract the phenolic compounds. Both studies showed that longer extraction time could increase the extraction yield of the phenolic compounds. However, in a Kamaludin *et al.* (2016) study, too long of an extraction time decreased the total phenolic content due to the deterioration of the phenolic compounds. These studies have shown that it is important to determine the optimum extraction condition to achieve the best result.

The advantages of Soxhlet extraction over the maceration method are lesser extraction time needed (4 h for the Soxhlet extraction method compared to 24 h for the maceration extraction method), high extraction efficiency and use of less solvent. However, Soxhlet extraction also has several serious drawbacks. The Soxhlet extraction method requires heating, and too high a temperature combined with a prolonged extraction duration increases the risk of thermal degradation (López-Bascón & Luque de Castro, 2020). Since the phenolic compounds are heat sensitive, therefore this type of extraction method is not suitable for industrial applications. It will affect the effectiveness of a product produced containing the phenolic compounds extracted from rambutan peel.

3.3 Microwave-Assisted Extraction (MAE)

MAE is one of the green methods that is currently used for recovering polyphenols and is popular due to the equipment being easily available at a low cost (Panja, 2018). MAE is a method of extraction that combines microwave energy with the conventional extraction method, maceration. Microwave energy will heat the solvents in contact with solid or liquid samples allowing the chemicals of interest from the sample to be partitioned into the solvent (Llompart *et al.*, 2018). In contrast to conventional heating which requires the container to be heated for a period of time before the heat is transferred to the solution, in MAE, microwaves would directly heat the solution instead (Llompart *et al.*, 2018).

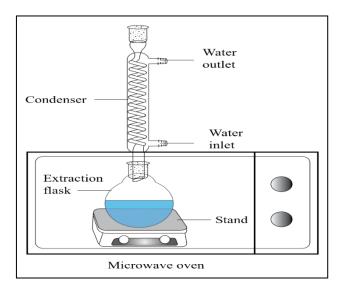


Figure 4. Illustration of Microwave-Assisted Extraction (MAE) method

For the MAE method, the highest amount of geraniin compound recorded was 122.18-140.02 mg/g (Li et al., 2018; Zhuang et al., 2017). The rambutan peels used in both studies were extracted under the same optimised extraction condition as follows: ethanol 80.85%, extraction time of 58.39 sec, and the ratio of liquid to solid of 24.51:1. However, Zhuang et al. (2017) had recovered phenolic compounds in the extract such as catechin (9.80 mg/g), ellagic acid (9.31 mg/g), corilagin (7.56 mg/g), gallic acid (0.69 mg/g), quercetin 3-O-b-glucoside (0.64 mg/g) and rutin (0.16 mg/g). Furthermore, Sun et al. (2012) were the ones who determined the optimum extraction condition as mentioned before. In the study, water, methanol, and ethanol were used but ethanol showed the highest yield of soluble phenolic compounds in rambutan peel extracts. This has shown that the yield of interested phenolic compounds is dependent on the solvent. In other words, the selection of solvent is based on the polarity of desired and undesirable phenolic compounds, safety concerns, and overall cost. From these studies, determining the optimum extraction condition saved time and it is useful for future studies to implement the optimised extraction condition. On the other hand, Chaiwarit et al. (2021) recovered a higher extraction yield of catechin with 118.26±4.57 mg/g at an optimised condition (ethanol 60%, (1:20; w/v), extraction time of 3 min with a frequency of 2450 MHz). According to Cuevas-Valenzuela et al. (2014), the higher the percentage of ethanol-water mixtures, the higher the solubility of catechin due to the dielectric constant of water-ethanol decreases, thus enhancing the solubility of catechin. On the other hand, findings in Chaiwarit et al. (2021) have shown that a much higher amount of catechin was achieved even though a lower concentration of ethanol was used, which was in contrast compared to the studies by Li et al. (2018); Sun et al. (2012); Zhuang

et al. (2017) that used a higher concentration of ethanol. However, considering that the extraction time in Chaiwarit *et al.* (2021) study was longer than those studies, this might explain that the result obtained was due to extraction time as it also influenced the result of extraction.

Through this MAE method, it reduced the usage of solvent, the extraction time also has been reduced by only 58.39 sec to 3 min thus, lowering the cost by reducing the energy usage and being more efficient compared to the conventional method. Nevertheless, MAE is not an appropriate method for a heat-sensitive compound, the equipment set-up is expensive and difficult to operate (de la Guardia & Armenta, 2011). Since most of the phenolic compounds are heat sensitive, the MAE method might not be the best extraction method to extract the phenolic compounds in rambutan peel.

3.4 Ultrasound-Assisted Extraction (UAE)

UAE, also known as sonication or ultrasonic extraction, is a type of extraction that employs ultrasonic wave energy. UAE waves are sound waves that are high in frequency beyond human hearing capability, making the extraction work of ultrasound devices in the range of 20 kHz to 2 MHz (Panja, 2018). Ultrasound in the solvent causes cavitation which speeds up the process of solute dissolution, diffusion, and heat transfer thus, enhancing extraction efficiency (Lavilla & Bendicho, 2017).

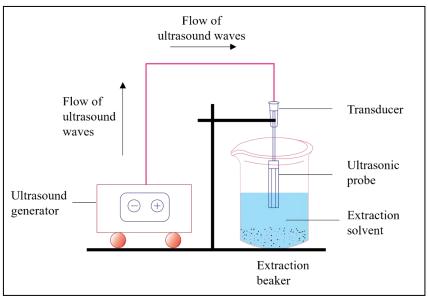


Figure 5. Illustration of Ultrasound-Assisted Extraction (UAE) method

Phuong *et al.* (2020b) achieved a high yield of geraniin, ellagic acid, quercetin, rutin and corilagin from methanolic extracts of rambutan peel with 397.28±9.5 mg/g,

177±0.4mg/g, 167.37±9.80, 36.4±1.7 mg/g and 3.81±0.7 mg/g, respectively. The extracts were obtained under the optimised conditions as follows: methanol 80%, room temperature for 20 min. The different proportion of water in methanol also influenced the extraction yield as it contributes to the production of a moderately polar medium that enhances phenolic component extraction (Phuong et al., 2020b) and this was shown when methanol 80% showed the highest extraction yield compared to the use of methanol 100% and methanol 50%. In this study, methanol showed higher efficiency in phenolic compound extraction compared to ethanol and water. Water showed the lowest amount of phenolic recovered, and this has proved that a very high polarity of solvent will not extract a high number of phenolic compounds. Other studies also have proved that using water alone produced a low amount of phenolic compounds (Maran et al. 2017). The rambutan peel was extracted using distilled water as the extraction solvent. Nevertheless, through this study, the optimised extraction condition has been determined as follows: extraction temperature of 50°C, ultrasound power of 20 W, extraction time of 20 min, and solid-liquid ratio of 1:18.6 g/mL. This finding should be certainly helpful for further studies to fully utilise the peels. Monrroy et al. (2020) have discovered that using only 60% ethanol could give a moderate amount of total phenolic content (340 \pm 4 mg/g), quercetin (76 \pm 2 mg/g) and cyanidin-3-O-glucoside (0.57 \pm 0.03 mg/g). A lower amount of total phenolic compounds was achieved by Mendez-Flores et al. (2018) from rambutan peel (Mexican variety) with only 487.67 mg/g under extraction condition of ethanol 10%, mass/volume ratio 1:7, room temperature extraction time 10 min. As low extraction time and low ethanol concentration were applied, this might explain the low concentration of phenolic compounds yielded. The rambutan origin country might be another factor that influenced the yield as higher extraction of phenolic compounds was recovered in rambutan peel extract from Asian origin (Mendez-Flores et al., 2018).

Relying on these studies, UAE seems to be less expensive due to the lower involvement of solvent volume (From Table 1, UAE method used range of solid to liquid ratio (sample: solvent) of 1:7-18.6 (Monrroy *et al.*, 2020; Mendez-Flores *et al.*, 2018; Maran *et al.*, 2017) as compared to other methods such as maceration which used 1:5-15 (Gusman *et al.*, 2015; Hernández-Hernández *et al.*, 2017), Soxhlet extraction method which used 1:5-10 (Chollakup *et al.*, 2020; Kamaludin *et al.*, 2016) and MAE method which used 1:20-24.51 (Chaiwarit *et al.*, 2021; Zhuang *et al.*, 2017). The usage of solvent reflects the involvement of cost since higher sample volume usage will incur more cost.), higher sample volume tested, and requires less extraction time thus, making it suitable for industrial application. Moreover, UAE could lower the cost as it requires low consumption of solvent and energy by reducing

the extraction time and temperature when compared to the conventional method such as maceration. The set-up cost in the UAE is relatively lower than MAE based on lower solvent usage (from solid: liquid ratio). According to Tiwari (2015), UAE also enhances the extraction yield, increasing extraction rates with the presence or absence of solvents allowing alternatives generally regarded as safe (GRAS) solvents such as ethanol and ethyl acetate to be used by improving their extraction performance, and increasing heat-sensitive components extraction whereby the yields would otherwise be low or unacceptable. All these advantages have made UAE one of the most suitable extraction methods for phenolic compounds extraction from rambutan peel.

4. The Most Suitable Extraction Methods to be Adopted in Industries

Industries are encouraged to extract their phenolic compounds from natural sources because the current concern about the impact of food on health has been influencing consumer choice of products. According to Martillanes *et al.* (2017), phenolic compounds are starting to substitute chemical additives in food because they are regarded to be safer and are supposed to reduce safety concerns. Therefore, the use of phenolic compounds from rambutan peel in food is a good opportunity for the application of their biological activities and allows the production of food without synthetic additives for consumers.

From the previous studies, UAE is the most frequently used extraction method to extract the phenolic compound from rambutan peel. The main reason is the extraction time is much faster due to the increased surface area between the solid (rambutan peel powder) and liquid phase (solvent) caused by the cell disruption and dispersion of particles compared to traditional methods (Carreira-Casais et al., 2021) which are maceration and Soxhlet extraction. According to Carreira-Casais et al. (2021), the major advantages of ultrasound are increased extraction performance, faster kinetics and simplicity in its operation, as well as more economical compared to other extraction techniques such as MAE (de la Guardia & Armenta, 2011). The UAE technique is inexpensive, simple, and efficient if the optimised condition is known. Moreover, when the optimum extraction conditions are already determined, it can minimise the setup times, so it creates more production and resulting more products being produced at one time. Since UAE needed a short extraction time, it can reduce the setup costs and increase production. Maran et al. (2017) achieved a total phenolic compound content of 552.64±1.57 mg gallic acid equivalent/ 100 g by UAE with the optimised condition of distilled water as a solvent, extraction temperature at 50°C, ultrasound power 20 W, extraction time of 20 min, and the solid-liquid ratio of 1:18.6. This finding also exceeded the predicted value which was 546.98 mg GAE/ 100 g. However, looking at the aspect of less extraction time, MAE provides a much lesser extraction time with 3 min under optimised conditions (Chaiwarit et al., 2021) compared to UAE which needs 20 min per mL (Maran et al., 2017; Monrroy et al., 2020; Phuong et al., 2020b). MAE tends to use much higher power compared to the UAE. This may increase the cost of the extraction process. Both methods involved simple pre-extraction methods; the peel just needed to be dried and pulverised at a specific particle size before extraction. Furthermore, UAE has recorded a higher number of phenolic compounds recovered with 250-300 mg/g compared to MAE with only 125 mg/g (Chaiwarit et al., 2021; Phuong et al., 2020b). In addition, UAE used a smaller amount of solvent that ranged from 10-18 mL (Maran et al., 2017; Monrroy et al., 2020; Phuong et al., 2020b) compared to MAE which needed a larger amount of solvent (Chaiwarit et al., 2021). Considering all these factors, it is therefore suggested that UAE is the most suitable method to be used in the industry for phenolic compounds extraction in terms of extraction time, cost reduction, and simple preparation needed. Maran et al. (2017) also mentioned that UAE is the most ideal extraction method due to its capability to produce high quantities of bioactive compounds in a short time. Moreover, there was an effort to apply UAE on an industrial scale through the first industrial-scale ultrasonic reactor designed to perform natural product extraction in a batch process (Vinatoru *et al.*, 2017). It is expected that the UAE will be used extensively on a large scale basis to extract the phenolic compounds soon.

The selection of solvent needs to be considered for the solute being extracted to accomplish other desirable characteristics such as low cost, low feed-phase solubility, and high recoverability. From Table 1, most of the solvents that have been used for the UAE extraction method are water, ethanol, and methanol because of their polarity properties. Polar solvents are frequently used for recovering polyphenols from plants and could give a high capacity of total extracts. Ethanol has long been recognised as an excellent solvent for polyphenol extraction that is also safe to consume, while methanol is more effective at extracting polyphenols with lower molecular weights (Morales-Olán *et al.*, 2020). Ethanol and methanol are polar solvents, which will result in a high extract yield. Since phenolic compounds are polar due to their hydroxy group (OH), they can be extracted easily with water, ethanol, and methanol. Besides that, ethanol and methanol are widely used as extraction solvents for phenolic compounds.

The concentration of ethanol and methanol greatly influences the total phenolic content of rambutan extract. Higher total phenolic compounds obtained from rambutan peel were extracted by using methanol 80% with 397 ± 9.5 mg GAE/g (Phuong et al., 2020b). Methanol could offer better results compared to ethanol as methanol has a lower boiling point than ethanol. By having a low boiling point, a lower temperature is needed to evaporate the solvent thus, protecting the extract from thermal degradation. Moreover, the mixture of water and ethanol or methanol (alcohols) has been revealed to be more efficient because it enhances the extraction of phenolic compounds compared to mono solvent as in findings (Chaiwarit et al. (2021); Gusman and Tsai (2015); Phuong et al. (2020b)) and are often used in extracting phenolic compounds in tropical fruit peel such as rambutan peel. However, most of the previous studies have used ethanol as the solvent (Gusman & Tsai, 2015; Kamaludin et al., 2016; Monrroy et al., 2020). Another study by Hernández-Hernández et al. (2020) also chose water as the best extraction condition over ethanol 70% with 307.57±20.27 mg/g extraction considering the aspect of the safety of the extraction, the amount of solvent used, the polarity of interest compounds, and the cost. Water is neither hazardous nor environmentally harmful, making it suitable for human consumption and industrial application. Other than solvents, extraction instruments are essential to ensure a better and more efficient yield of a phenolic compound extracted from rambutan peel. Advanced technologies nowadays have improved and eased the extraction process. Some of the available instruments that have been used for the UAE method are Power-Sonic, Korea (Maran et al., 2017), Hielscher UP 400S, Germany (Phuong et al., 2020b), Elmasonic P60H, Singen, Germany (Monrroy et al., 2020) and phase-power control microwave extraction (PC-MHG) system (Chaiwarit et al., 2021).

5. Conclusions

Rambutan peel is part of rambutan by-products that cannot be consumed directly due to its undesirable bitter taste causing the peel to always be discarded and this has contributed to the number of waste produced. To overcome this matter, this review describes the available extraction methods to extract the phenolic compounds from rambutan peel to transform it into a useful product since the peels contain abundant valuable phenolic compounds (geraniin, corilagin, ellagic acid, and rutin) and by utilising these phenolic compounds for industrial purposes could significantly reduce the number of rambutan peel waste. The presence of phenolic compounds in rambutan peel serves several biological activities such as antibacterial, antioxidant and anti-cancer that could be beneficial for health purposes and to prevent oxidation in rubber products. Therefore, the selection of an extraction method is crucial as industries need to produce a large number of products. UAE, a non-conventional extraction method has the best potential to be adopted in the industries because of its high

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extraction efficiency, short extraction time, lower cost and water alone can be used as the solvent. UAE also can be used to extract other bioactive compounds such as vitamins which may be used as an alternative method to extract vitamins from existing methods. Nonetheless, UAE needs to be operated in a large sample volume to determine its optimum condition to establish these phenolic compounds that can be utilised in food, cosmetics, pharmaceuticals, and rubber industry applications.

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