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A critical review of the extraction methods of value-added products derived from rambutan (*Nephelium lappaceum*) seeds

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Abstract

Rambutan processing discards the peels and seeds as fruit by-products constituting almost 50 percent of the total fruit weight. In the Philippines, the country's total rambutan fruit production volume reached 8,312 metric tons in 2020 which usually ends up in landfills, contributing to municipal solid waste. Production of value-added products (VAPs) is an effective agro-waste management approach to valorize rambutan seeds for food and non-food applications. This paper aimed to assess the extraction methods of rambutan seed VAPs, the benefits and limitations of each extraction technique, and the strategies for optimizing the operational conditions to ensure maximum yield. Conventional extraction methods heavily rely on the use of large amounts of organic solvents which pose safety and environmental risks, typically requiring long extraction time with low yield. In this paper, digestion, solvent extraction, mechanical oil extraction, maceration, and Soxhlet extraction were the conventional methods assessed. Recently, development towards addressing these limitations is the focus of non-conventional extraction methods such as supercritical fluid, ultrasound-assisted, and enzyme-assisted extraction. These extraction methods are environment-friendly and can be employed to produce VAPs, often with higher extraction yield at shorter extraction time. Choosing the most suitable extraction method is a challenging task as it weighs multifarious considerations in terms of economic value, technological cost, extraction time, environmental issues, and feasibility of scaling up. Strategies to improve extraction yield include pretreatments and optimization studies through response surface methodology. The development of smart, low-cost, and greener extraction methods addresses the environmental issues and low extraction efficiencies of conventional methods.

Introduction

During the processing of rambutan fruit, where seeds and peels are discarded as by-products, the peeled rambutan is usually consumed fresh (Hernández-Hernández et al., 2019). By contrast, in southeast Asian countries, rambutan fruit is deseeded and processed into jams and jellies (Jahurul et al., 2020), canned in sweetened syrup (International Tropical Fruits Network, 2012), or dried into rambutan prunes (Irader et al., 2010). Depending on the variety, rambutan seeds and peels constitute almost 50% of the total fruit weight (Mahmod et al., 2018). In 2020 alone, the Philippine Statistics Authority noted the country's rambutan fruit production volume was 8,312 metric tons. When by-products are discarded, a significant amount of rambutan seeds and peels may end up in landfills. This contributes to municipal solid waste derived from other major tropical fruits (i.e. banana, citrus, etc.), worsening the situation of waste pollution (Pathak et al., 2017).

Production of value-added products (VAPs) is one of the agro-food waste management strategies to combat the worsening issue of waste accumulation. Extraction techniques have been widely used to produce VAPs which can be categorized into conventional methods (Garcia-Vaquero et al., 2020; Nde & Foncha, 2020; Rasul, 2018) and non-conventional methods (Bitwell et al., 2023; Cheng et al., 2015; Santos & Santana, 2022). In the context of rambutan seeds, conventional extraction methods are traditional techniques used to produce VAPs, including digestion (Evaristus et al., 2018), solvent extraction (Zurina et al., 2014), mechanical oil extraction (Khairy et al., 2018), Soxhlet extraction (Ghobakhlou et al., 2019), and maceration (Lourith et al., 2016). The use of toxic solvents and low extraction efficiencies are the drawbacks of these methods (Abubakar & Haque, 2020). Non-conventional extraction methods refer to modernized and advanced techniques such as supercritical fluid extraction (Eiamwat et al., 2016), ultrasound-assisted and enzyme-assisted extraction (Xuan et al., 2017) which are environment-friendly and characterized by improved extraction yield (Santos & Santana, 2022). These techniques vary in terms of operational conditions, yield values, product quality, extraction time, and environmental risks (Alexandre et al., 2017; Gharib-Bibalan, 2018). The objective of this review is to assess the extraction methods of rambutan seed VAPs, the benefits and limitations of each extraction technique, and the strategies for optimizing the operational conditions to ensure maximum yield.

Utilization of Rambutan Seeds into Value-added Products

In the context of food waste management, the processing of agricultural by-products into something valuable for various industries is called the production of value-added products. This approach helps to mitigate the environmental and health risks posed by traditional forms of waste disposal such as incineration and landfilling. Agricultural by-products generally consist of important components such as carbohydrates, lipids, proteins, and inorganic compounds. Characterization of these components can guide the user on which type of extraction method to employ to produce the VAP on top of the application of physical, chemical, or biochemical treatments (Gunjal, 2019; Matumoto-Pintro & Saraiva, 2023; Otles & Kartal, 2018). Examples of rambutan seed VAPs include mucilage, amylase inhibitor, industry-grade starch, bio-coagulant, specialty fat, biodiesel feedstock, seed fat, and oil. VAPs derived from rambutan seeds are important in many industrial applications such as food (Wanthong & Klinkesorn, 2020), renewable energy (Nguyen et al., 2016), cosmetics (Lourith et al., 2016), pharmaceutical (Fang & Ng, 2015) and wastewater treatment industries (Zurina et al., 2014). In this way, industrially important VAPs can also reduce the issue of high production cost (Mahendran et al., 2015). At the same time, it can align to consumer preference for product sustainability as found in the report of the First Insight and Baker Retailing Center (2021).

Proximate analysis of rambutan seed by Wahini et al. (2018) revealed the important macro- and micronutrient components of the fruit by-product. Specifically, rambutan seed contains macronutrients (64.19% carbohydrate, 11.38% protein, 6.01% lipid), micronutrients (0.33% Vitamin B, 62.50% minerals (calcium, iron, phosphorus)), and other components (14.20% moisture, 2.51% fiber, and 1.70% ash). The composition serves as the basis on which higher quantities of value-added products can be derived. Food and non-food applications of rambutan seed VAPs and their extraction methods are summarized in Table 1.

Extraction Methods

According to Zhang et al. (2018), extraction serves as the initial step to obtain the desired compounds from the raw materials. In the context of this paper, the raw materials or plant materials, referred to as the fruit by-products, are rambutan seeds. Zhang et al. (2018) also describe the extraction process, which first involves solvent penetration into the plant material. Then, the solute, which is the compound of interest, dissolves in the solvent. This is followed by the diffusion of solute from the plant material. Lastly, the extracted solute is collected. Factors that affect the extraction efficiency include the following: 1) type of solvent used (polarity of the desired compound), 2) particle size of plant material (affects surface contact with solvent), 3) temperature (heat-sensitivity of the compound), and 4) extraction period (Abubakar & Haque, 2020; Stéphane et al., 2021). All these factors ultimately affect the extraction yield of the desired compound.

The choice of solvent depends on the nature of the desired compound. Based on the law of immiscibility, solvents with the same polarity as the solute will result in better extraction (Popova & Bankova, 2023). Alcohols such as ethanol and methanol are polar in nature so these are used widely in the extraction of polar phytochemicals. Intermediate polar solvents are acetone and dichloromethane; meanwhile, nonpolar solvents include n-hexane, chloroform, and ether. Other considerations in choosing which type of solvent are solubility, cost, and safety (Abubakar & Haque, 2020).

Table 1. Summary of the extraction method for food and non-food value-added products derived from rambutan seeds.

Product	Extraction method	Operational conditions	Extraction time	Yield	Reference
Food					
Conventional extractions methods					
Cocoa butter alternative	Mechanical oil extraction	Screw oil expeller (60°C)	30 min	20% (RSF*)	Khairy et al., 2018
Bio-oil	Soxhlet extraction	Petroleum ether solvent (40-60°C boiling point)	8h	34.25-37.62%	Ghobakhlou et al., 2019
Non-conventional extraction methods					
Albumin	Enzyme-assisted extraction	Beta-glucanase Viscozyme L® (5 FBG* per g dry weight)	90 min	48.0%	Xuan et al., 2017
	Ultrasound-assisted extraction	20 Watts per g dry weight material (2nd stage ultrasonic power)	20 min (with 2 min sonication time)	45.90%	Xuan et al., 2017
Seed fat Seed oil	Supercritical fluid extraction	Carbon dioxide as supercritical fluid (35 MPa, 45°C)	44h	16-23% 5.0-7.6%	Eiamwat et al., 2016
Non-food					
Conventional extractions methods					
Biocoagulant	Solvent extraction	dH_2O (100 mL, room temperature)	Several minutes	100 $mg{\cdot}L^{-1}$	Zurina et al., 2014
Amylase Inhibitor	Digestion	Enzymatic in-vitro digestion (95°C)	30 min	12 and 14 $mg{\cdot}g^{-1}$	Evaristus et al., 2018
Industry-grade starch		0.1 M NaOH, 0.1 M HCl (26-27°C)	18 h	56.06%	Arollado et al., 2018
Mucilage		250 mL, dH_2O*	Boiled for 1 h under reflux	3.30%	Mahendran et al., 2015
Specialty fat	Maceration	N-hexane (industrial-grade, 1 h)	1 h	32.60%	Lourith et al., 2016

*FBG: fungal beta-glucanase unit, RSF: rambutan seed fat, dH_2O: distilled water

Low particle size has a significant effect on the extracted compounds due to better contact with the solvent. This effect stems from the heightened surface contact with the plant materials and extraction solvent, consequently leading to increased extraction yield.

This implies the importance of grinding as one of the initial steps to homogenize plant materials. Increasing the temperature will also increase solubility and diffusion; however, when the temperature is too high, it can degrade heat-sensitive compounds (Abubakar & Haque, 2020).

Conventional Extraction Methods

Digestion

Heat application reduces the solvent's viscosity, which facilitates an easier exclusion of plant components (Majekodunmi, 2015). The use of moderate heat is advantageous to maintain the integrity of the compound to be extracted, and is utilized in the digestion technique (Pandey & Tripathi, 2014). The extraction solvent is poured onto the plant material in a container, and the mixture is typically heated either over a water bath to sustain an elevated temperature or placed in an oven, ensuring that the heat does not exceed 50°C (Abubakar & Haque, 2020). However, the use of solvents requires a long extraction time and poses risks to the environment (Pandey & Tripathi, 2014; Stéphane et al., 2021).

Digestion methods were utilized by several studies to produce rambutan seed VAPs. In mucilage isolation by Mahendran et al. (2015), powdered rambutan seeds were soaked in distilled water for 24 h. At elevated temperatures, the seed was boiled under reflux. Isolation techniques such as filtration were done before the addition of ethanol in the filtrate. For effective settling of the desired polymer, the filtrate-ethanol mixture was kept refrigerated for one day. The final powdered product, after it was dried completely in an incubator, was stored inside a desiccator until it was used. Extracting the mucilage as a natural polymer has certain advantages over synthetic polymer, given the absence of toxic solvents, making it environmentally friendly and more cost-effective. However, the digestion method is labor intensive.

Evaristus et al. (2018) utilized sequential digestion to extract amylase inhibitor peptides from rambutan seeds. The rambutan seeds were first defatted before protein was extracted using a phosphate buffer. The extracted protein from rambutan seed samples weighed 12 $mg \cdot g^{-1}$. In the simulation of *in vitro* digestion, the study employed three digestion conditions using gastro-digestive enzymes. Enzymes included pepsin, chymotrypsin, and pepsin followed by chymotrypsin. Each of these was subjected to a buffer solution with varying pH. The samples were heated in a thermomixer at 95°C for 30 min, terminating the digestion process. At 3 kilodalton (kDa) molecular weight cutoff, membrane filtration screened protein hydrolysates extracted per digesting condition. Inhibitor peptides produced from rambutan seed proteins were found to be effective using chymotrypsin.

Arollado et al. (2018) isolated the starch from rambutan seeds by immersing the plant material in an 80% ethanol solution at 85°C. This was done to remove the mono- and oligosaccharides. Extraction of starch was done by first grounding the rambutan seeds in a food blender and then removing the supernatant. Using 0.1M NaOH, the residue was treated for 18h at room temperature. Next, the top layer was removed and treated with 0.1 M HCl to neutralize the alkalinity. Further mechanical methods such as filtering, drying, crushing, and sieving were done to produce the powdered final product.

Solvent Extraction

This method relies on differential solubility to selectively dissolve components from mixtures. Depending on the choice of solvent, the extraction method is versatile and capable of extracting products like essential oils, bioactive compounds, and metal ions. Advantages include high selectivity and adaptability, but drawbacks include the environmental and health risks posed by the use of hazardous solvents (Abubakar & Haque, 2020).

Zurina et al. (2014) extracted active ingredients such as a coagulant agent of rambutan seeds using solvent extraction. The skin of the seed material was first removed. Pulverization of seed material was carried out using an electric blender to produce a fine powder. Particle size ranged from 65-200 μm. The powdered seed material was then suspended in distilled water as the solvent and was filtered using a muslin cloth. The bio-coagulant property of rambutan seed was tested for turbidity removal in wastewater.

Mechanical Oil Extraction

A conventional method for extracting oil in plant samples uses mechanical pressers. A manual ram press, according to Baskar et al. (2019) can extract 60-65% oil whereas an engine-driven press is capable of extracting a higher percentage, ranging from 68-80%, from seed material. According to the authors, the design of the mechanical press affects the extraction yield, and requires subsequent filtering and degumming processes. Other considerations such as pretreatment and cooking process increase the extraction yield. This method gives instant high-quality crude oil without the use of any solvent, but entails low yield and low extraction efficiencies (Bhargavi et al., 2018). On an industrial scale, mechanical oil extraction is unprofitable due to the high oil content present in the residual fruit seeds (Bhargavi et al., 2018).

Khairy et al. (2018) extracted rambutan seed fat, a potential cocoa butter alternative, using a screw oil expeller. Pretreatment of the rambutan seeds included a six-day fermentation period, concluding with the roasting of the dried rambutan seeds at the end of the fermentation process. Filtration at an elevated temperature (60°C) separated the rambutan seed fat from its seed butter. The extracted rambutan seed fat was further analyzed.

Soxhlet Extraction

A traditional extraction method for high-value compounds from a solid sample employs Soxhlet extraction which combines the method of percolation and maceration (Kim et al., 2012; Malik & Mandal, 2022). Nafiu et al. (2017) explained that the sample is usually treated with a wide variety of solvents. The desired compound should have limited solubility to the chosen solvent. According to de Boer (2005), Soxhlet extraction may utilize the following solvents: dichloromethane and acetone or hexane in pure or mixed form. Although it is simple and effective, it has a long extraction time of around eight hours, and requires large amounts of solvent. Also, the method is prone to sample contamination with impurities (Azmin et al., 2016).

Ghobakhlou et al. (2019) utilized Soxhlet extraction to produce bio-oil from dried rambutan seeds from Malaysian cultivars, which exhibited high amounts of arachidic acid, demonstrating resistance to oxidation. Hajar et al. (2017) extracted edible fat from dried rambutan seed, using petroleum ether as the solvent. Soxhlet extraction methods were used as preliminary procedures in the production of non-hydrogenated fat (Wanthong & Klinkesorn, 2020) and biodiesel (Nguyen et al., 2016). Drying is a crucial step in this extraction method to ensure a high fat extraction efficiency from rambutan seed fat (Chimplee & Klinkesorn, 2015).

N-hexane Maceration

In this type of solvent extraction, n-hexane offers advantages for extracting essential compounds. n-hexane, according to Kumar et al. (2017), has the following attributes: 1) non-polar, 2) high selectivity to other solvents, and 3) low latent heat of vaporization of approximately 330 $kJ \cdot kg^{-1}$. This method is simple and inexpensive and can be used in a wide range of extracting thermolabile components. However, using n-hexane solvent involves environmental and toxicological issues primarily due to solvent use. This method is characterized by a lengthy extraction time, and its low efficiency places limitations on the extraction process (Cheng et al., 2015; Zhang et al., 2018).

Lourith et al. (2016) utilized this type of maceration in extracting specialty fat in the personal care industry. The surface area of the rambutan seed was increased by grounding, thus, achieving effective maceration. Removal of the solvent, according to their research, was done using a rotary evaporator to finally obtain the specialty fat.

Non-conventional Extraction Methods

Ultrasound-assisted Extraction

This extraction method uses ultrasound waves to generate mechanical energy. The sonication process involves the formation of voids and small bubbles that subsequently implode within the plant material. This force brought by cavitation action results in cell membrane destruction, sonolysis, and intracellular material extraction (Kumar et al., 2021). Ultrasound-assisted extraction (UAE) is relatively cheap in comparison to other nonconventional extraction methods. According to Herrero et al. (2012), other advantages of UAE are simple instrumental requirements, reduced extraction time, and the absence of toxic solvents. Also, UAE can be used to extract a wide variety of important compounds such as fats, isoflavones, lycopenes, and flavanone glycosides (Pingret et al., 2012). However, Gulzar and Benjakul (2019) emphasized that UAE may cause degradation to extracted lipid compounds due to oxidation and hydrolysis.

Xuan et al. (2017) extracted albumin from defatted rambutan seed using ultrasound-assisted extraction. The study compared the yield values of albumin with enzyme-assisted extraction. It was found that optimal extraction conditions were 20 Watts per g dry weight material, 2 min sonication time, and 20 min extraction time which produced a yield of 45.9% albumin. The authors noted that there was no change in the albumin yield when sonication time was increased above two minutes. Also, extracted albumin from rambutan using the UAE did not affect its functional properties which makes it suitable for food applications.

Enzyme-assisted Extraction

The principle of this extraction method involves the role of enzymes in plant cell wall disruption. This was carried out by enzyme hydrolysis of the cell wall performed under controlled conditions. The binding of the cell wall into the enzyme's active site causes the modification of enzyme shape. It leads to bond fragmentation releasing the intracellular components (Nadar et al., 2018). The process carried out in a controlled temperature condition can result in the effective removal of heat-sensitive biomolecules. The benefits of using the method include shorter extraction time and the reduced impact of the use of hazardous solvents (Cheng et al., 2015). However, implementing this method on an industrial scale is challenging due to incomplete breakdown of plant cell walls caused by the limitations of the available enzyme. Also, the high cost of the commercially available enzymes inhibits the potential of this method (Cheng et al., 2015; Zhang et al., 2021).

In the context of rambutan by-product utilization, Xuan et al. (2017) extracted albumin from defatted rambutan seed fat. Enzyme-assisted extraction yielded the maximum albumin content found in the extract, which was found to be 3.6% higher relative to ultrasound-assisted extraction. Important functional properties innate to the albumin were not altered with the two methods used. The study utilized a commercial broad-spectrum enzyme for the hydrolysis of plant tissue.

Supercritical (SC) Fluid Extraction

This is an environmentally friendly extraction technology suitable for heat-sensitive hydrophobic compounds (Yang & Hu, 2014). The principle behind this extraction method involves the use of a specialized solvent called supercritical solvent, which, under specific temperatures and pressures, surpasses its critical points. Specifically, at elevated pressure, carbon dioxide (CO_2) will liquefy which then acts as a solvent extracting the bioactive compound of interest (e.g. essential oil, etc.) from the plant material. As the pressure is lowered, the CO_2 returns to a gas state, extracting volatile oil (Ibáñez et al., 2016). The SC-CO_2 technology is environmentally friendly as it avoids the use of toxic solvents and, at the same time, produces high extraction efficiency. Also, the extraction method does not degrade the bioactive compounds. However, the method is capital-intensive in terms of equipment and the need for high temperatures and pressure. Mourtzinos and Goula (2019) emphasized that SC-CO_2 may not be efficient in extracting polar polyphenolic compounds due to the nonpolar nature of CO_2. To increase solvating power and selectivity for extracting the bioactive compound of interest, modifiers such as water, methanol, and ethanol can be used.

Eiamwat et al. (2016) extracted rambutan seed fat and oil using SC-CO_2 extraction at 35 MPa and 45°C. The results revealed that the seed fat yield varied between 16-23%, whereas the seed oil exhibited a yield ranging from 5.0-7.6%. The study also performed a 14-day acute oral toxicity test on rats at a 5000 $mg \cdot kg^{-1}$ bodyweight limit dose. No signs of intoxication were found, which means that rambutan seeds can be a potential fat and oil source for food applications.

Strategies to Improve Extraction Yield

Employ Pretreatment

Zhao et al. (2014) enumerated physical (sun drying, grounding, heating), chemical (defatting), and biological (enzyme hydrolysis, microorganism) techniques as potential pretreatment to obtain high extraction yield. Pretreatment increases the cell wall permeability of the rambutan seed (plant material). While important bioactive compounds remain within the plant cell wall, solvent diffusion is restricted. Nevertheless, employing pretreatment increases the extraction rate by increasing cell wall permeability.

Optimize Process Parameters

Extraction time and temperature are the operational parameters during the extraction process that directly impact the extraction yield. Response surface methodology (RSM) can be used to determine the optimal independent variables to acquire a maximum response. This statistical technique guides the user in understanding how each variable (extraction time, temperature, solvent concentration, etc.) interacts with each other. This process is more efficient relative to optimizing a single parameter alone as it consumes less time, raw materials, and reagents (Kim et al., 2014).

In fat extraction from rambutan seeds by Sirisompong et al. (2011), RSM technology was utilized to investigate the impacts of some parameters (i.e. particle size, extraction time, moisture content) on the fat yield. The study confirmed the optimum conditions necessary to obtain the maximum yield of the product, around 37.35 g per 100 g. This suggests the utilization of RSM to VAPs derived from rambutan peels and seeds to have an optimum condition for various extraction methods.

Summary and Conclusion

Rambutan seed is one of the by-products obtained from rambutan processing. To address the issue of waste accumulation, VAP production is an effective approach to valorize rambutan seeds for various industrial applications. This offers economic opportunities for potential technological adoption by micro-, small, and medium enterprises in rambutan producing regions. Since rambutan seeds do not compete in the food system as conventional food materials, they can be used as cost-effective raw materials for both food and non-food applications.

VAPs are processed using extraction methods that differ in complexity. There are advantages and disadvantages to each extraction method in terms of cost, efficiency, and degree of recovered product. For instance, conventional extraction methods are simple to perform; however, the extraction time is typically long with low extraction yield. In the case of solvent extraction, the process relies on the use of organic solvents which entails safety and environmental issues. Non-conventional extraction methods, on the other hand, address these limitations but these technologies are usually capital-intensive in terms of equipment cost and require advanced technical skills. Proper selection of the most suitable extraction method for the VAPs is multi-factorial. It considers the nature and value of the VAP to be extracted along with the industrial application, technological cost, extraction time, and environmental issues of the solvents. Every extraction technique has specific advantages and limitations, and the target user who will adopt the extraction technology must carefully examine each of these factors.

Moreover, optimization studies via response surface methodology (RSM) of the extraction process parameters (e.g. time, temperature, concentration) are essential to ensure high extraction yield and efficiency. However, there is a scarcity of studies incorporating RSM for value-added product (VAP) production using rambutan fruit by-products.

Future perspective

There are still gaps in fully maximizing value-added production using rambutan fruit by-products. For future directions, further optimization studies with process parameters need to be conducted among other VAPs to ensure a high extraction yield. Also, it is recommended to continuously develop smart, cost-effective, and greener extraction alternatives that solve the environmental issues and poor extraction efficiencies of conventional extraction methods. Other novel technologies can be explored such as pulse electric field-assisted extraction and high-pressure extraction methods to extract VAPs from rambutan seeds. These methods have better extraction yields and efficiency without the use of toxic solvents (Huang et al., 2013; Ranjha et al., 2021).

Policymakers also play a crucial role in crafting regulations for rambutan seed VAPs. While more research is needed, it is envisioned that private industries would use rambutan VAPs as non-conventional raw materials to replace synthetic and high-cost raw materials. Ultimately, this will open an avenue for international regulatory bodies to be inclusive of value-added products as sustainable raw materials for various industries.

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No potential conflict of interest was declared by the authors.

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