Recent trends in pre-processing and extraction of watermelon rind extract: A comprehensive review

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Abstract: Watermelon rind contributes 30 % (w/w) of overall fruit mass, mainly carbohydrates, fibre, and wax. The rind is often discarded due to its unappealing flavour. Several studies proposed that watermelon rind waste can be utilized as high fibre flour, dietary supplement, food additive and bio-sorbent material. However, before the offered product is developed, a proper technique of pre-processing and extraction methods are vitally important to be considered as they will determine the extraction yield quality. Thus, this review aims to provide an extensive overview of pre-processing and extraction techniques. A comparison of different procedures applied in the pre-extraction and extraction of watermelon rind is emphasized. Pre-processing parameters affecting extraction yields such as sample condition, drying condition, and grinding and grading technique are discussed. Several extraction techniques, including infusion, maceration, digestion, reflux, ultrasound-assisted extraction (UAE) and microwave-assisted extraction (MAE), are correlated to identify their efficiency in extracting the phytochemical from watermelon rind. Factors that influence the extraction yield such as extraction temperature, solvent-to-solid ratio, extraction duration, and type of solvents are elaborated.

Keywords: watermelon rind, phytochemical compound, extraction technique, extraction yield.

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1.0 Introduction

Watermelon (*Citrullus lanatus*) is one of the most cultivated fruits globally and is valuable for its sweet and juicy fruit. It is often consumed in hot weather in tropical and subtropical climates, especially in Africa, Asia, the United States, Russia, and the Mediterranean (Neglo et al., 2021). Watermelon belongs to the family Cucurbitaceae, including several other fruits such as winter melon, muskmelon, squash, cucumber, bottle ground and pumpkin (Ridwan, Razak, Adenan, & Saad, 2018). Watermelon comprises three main parts: flesh, rind, and seed. According to Ramakrishnan et al. (2020), watermelon flesh contributed to 68 % (w/w) of overall fruit mass, 30 % (w/w) is watermelon rind and the other 2 % (w/w) is leftover. Watermelon rind contains carbohydrates, fibre and wax (Petchsomrit, McDermott, Chanroj, & Choksawangkarn, 2020). Ramakrishnan et al (2020) also reported that watermelon rind consists of 13 % (w/w) pectin, 10 % (w/w) lignin, 23 % (w/w) hemicellulose and 20 % (w/w) cellulose. However, the composition percentage may vary depending on the watermelon genotype. Every part of the fruit, either flesh, rind, peel, or seeds contains a nutritional value. However, most people avoid eating the rind due to its unappealing flavour. Watermelon rind waste is often utilized in food products such as pickles, stir-fried and stewed. On the other hand, watermelon rind can also be converted into flour by drying and milling it to fine form. This flour can nutritionally enhance baked foods such as cookies and cakes, containing high fibre (Adegunwa, Oloyede, Adebanjo, & Alamu, 2019).

Watermelon rind is rich in vitamin C, fibre, potassium and a small amount of vitamin B (Adegunwa et al., 2019). The rind is also enriched with antioxidant compounds called citrulline, which can give a therapeutic effect, increase vasodilation in many body tissues, and reduce the risk of several cancers (Hartman, Wehner, Ma, & Perkins-Veazie, 2019). In the pharmaceutical industry, watermelon rind extract in the form of L-citrulline has been utilized as a dietary supplement to treat certain urea cycle disorders (UCD) (Johnson, 2017). The watermelon rind extract containing citrulline also acts as anti-ageing properties in the cosmetics industry by regulating collagen and improving skin suppleness (Raikou, Varvaresou, Panderi, & Papageorgiou, 2017; Schagen, 2017). The International Name Cosmetic Ingredient (INCI) recognized the cometic grade citrulline as Tripeptide-10 Citrulline. Watermelon rind could be applied in various applications such as an additive in food industries, anti-ageing in cosmetic products and bio-sorbent material in wastewater treatment. Traditionally, Korea has used cold watermelon rind thin slices as a sheet mask and blended the cold rind as a face mist. Besides, the watermelon rind can be utilized as a pectin source, as Lee & Choo (2020) reported. Watermelon rind pectin is widely used as a food additive in the food industry and as a biopolymer in pharmaceutical industries. Watermelon rind can also be converted into a bio-sorbent material to remove heavy metals from wastewater (Lee & Choo, 2020; Ramakrishnan et al., 2020).

Obtaining compounds in the rind like pectin needs several steps starting from sample preparation. Sample preparation can be described as a process in which a representative piece of a compound is extracted from a more significant number of sources and adequately prepared for further analysis (Dulski, 2016). The extraction technique used during processing depends on the desired constituent to be isolated from the watermelon rind. Thus, there is an urge in searching for the proper conditions of watermelon rind pre-processing and extraction methods that can provide the optimum conditions for the extraction yield of the watermelon rind. Therefore, this review intends to provide an extensive overview of different procedures that could be used in the pre-extraction and extraction of the watermelon rind to be applied in various industries.

2.0 Pre-extraction preparation of watermelon rind

2.1 Sample condition of watermelon rind

Sample condition plays an essential role in the extraction process. The rind condition, either fresh or dried, is crucial as it affects extraction yield. As previous studies reported, both situations have been applied in the extraction process depending on a specific phytochemical compound to be isolated. A survey conducted by Akshaya et al. (2018) on the determination of antioxidant activity and total phenolic content of fresh watermelon rind samples using different thermal treatments (steam blanching and water blanching) showed a significant difference in the amount of compound obtained. The total phenolic content obtained using steam blanching is 0.2875 mg/g, higher than water blanching 0.1412 mg/g. A similar result was obtained for antioxidant activity. Watermelon rind treated with steam blanching also showed a significant high tannins content (0.81 mg/g), alkaloids (0.6 mg/g), and saponin (1.52 mg/g) as compared to the water blanching (0.47, 0.31, and 0.87 mg/g respectively) (Akshaya et al., 2018). The situations proved that most compounds are sensitive to high temperature or thermal treatment.

However, fresh samples tend to deteriorate faster during the experiment as they contain high moisture levels that are prone to microbial growth and enhance enzymatic activity (Azmin et al., 2016; Babu, Kumaresan, Raj, & Velraj, 2018; Ho, Ramli, Tan, Muhamad, & Haron, 2018). Thus, in most cases, dried plant samples are always preferable to increase the sample shelf life by considering the time of experimental work. According to Ho et al. (2018), drying samples by reducing the moisture content by less than 15 % from the fresh sample can help in minimizing chances of bacteria growth and its proliferation, thus increasing the sample shelf-life. Extraction done by Augustia et al. (2020) using a fresh watermelon rind sample showed a significantly lower value of total flavonoid content (0.71 - 1.63 mg/L) as compared

to the dried sample conducted by Nurdalilah et al. (2018) (1.2175 mg/g) and Ho et al. (2018) (13.95 - 123.31 mg/g). The results were compatible with Baeeri et al. (2018), which the main objective is to compare three different conditions of the watermelon rind sample (fresh, dried and frozen). The result found that the extract from the dried sample contains higher total phenolic content and total amino acid content than the fresh and frozen sample. Meanwhile, there was no significant difference in anti-tyrosinase activity of all sample conditions. The distinction between results may be due to the degradation of the compound as fresh samples are fragile and tend to deteriorate faster than dried samples with a maximum of 3 hours to maintain their freshness.

2.2 Drying technique

Drying is the best method to preserve watermelon rind for a longer duration. Drying is the process of moisture removal by using heat which affects the microbial growth, enzymatic activity and sensory properties of plant material (Calín-Sánchez et al., 2020). Generally, drying temperature and time are varied by the plant type and plant part, the thickness of spread and volume of air. Previously, Hoque & Iqbal (2015) experimented on the determination of watermelon rind drying rate by using a similar drying method (cabinet dryer), constant sample thickness (8 mm), constant air velocity (0.6 m/sec) and different drying temperatures (55, 60, and 65 °C). The study confirmed that the higher the temperature and the longer the drying period will decrease the moisture ratio, thus resulting in a faster drying rate of the watermelon rind sample. This study found that the moisture content of fresh watermelon rind decreased from 94.62 % to 10.72 % after the drying process (Hoque & Iqbal, 2015). Hence, lowering the watermelon rind moisture content is essential to avoid spoilage and increase shelf life without deterioration in the nutrient levels. The ideal drying temperature (40 - 65 °C) of the watermelon rind sample is presented in Table 1 as suggested by several studies. This perfect temperature is crucial for retaining the original plant compositions such as antioxidant compounds, aromatic compounds, sugar, pectin, cellulose, and lipid composition. Babu et al. (2018) claimed that drying watermelon rind at a lower temperature can prevent colour quality deterioration, the chemical constituents' degradation, and preserve the organizational structure in plant samples.

2.2.1 Sun drying

Sun drying is a traditional method applied to dry watermelon rind by exposing directly to solar radiation. The optimal conditions for sun drying are at an average atmospheric air temperature of 28 – 40 °C with relative humidity below 60 % (Babu et al., 2018). This drying process is very time-consuming as it usually takes over a week to remove moisture from plant samples to the desired level, which is less than 10

% by weight (Babu et al., 2018; Sluiter, Amie, Justin Sluiter and Edward J, 2017). Egbuonu (2015b) studied on watermelon rind sample where it was sundried to 4 % by weight with wet weight of 1900.7 g to 82.6 g dry weight before further analysis. Another major drawback of this technique is that it may not be effective for some plant samples because it may cause degradation of colour and aromatic compounds. Egbuonu (2015b) determined that sun-drying does not affect mineral and vitamin composition in watermelon rind; however, the amino acid composition cannot be preserved (0.00/100 g). Egbuonu (2015b) reported that the mineral composition contained in sundried watermelon rind are calcium (28 ± 0.01), phosphorous (129.7 ± 0.01), sodium (11.4 ± 0.04), potassium (21.7 ± 0.00), magnesium (30.4 ± 0.01), manganese (1.30 ± 0.01), iron (4.63 ± 0.00), copper (0.4 ± 0.01) and zinc (1.25 ± 0.01). Meanwhile, the value of vitamin compositions in the sundried watermelon rind is retinol (50.15 ± 1.41), niacin (0.04 ± 0.1), ascorbic acid (7.23 ± 0.02), thiamine (0.03 ± 0.01), riboflavin (0.02 ± 0.1) and pyridoxine (0.04 ± 0.00) (Egbuonu, 2015).

2.2.2 Shade Drying

Shade drying is the effective drying method for preserving watermelon rind's primary nutrients and chemical constituents. Primary nutrients of watermelon rind such as carbon (26.13 – 29.09 %), hydrogen (2.56 – 4.32 %) and nitrogen (7.11 – 7.92 %) were able to preserve by shade drying pre-processing (Latif et al., 2019). The drying process involves plant material exposure at ambient temperature in a shaded place with plenty of air circulation. The drying process typically takes 3 - 7 days to months and up to a year, depending on the plant part and size of the plant sample. Despite that, the natural and non-thermal shade drying process can preserve most constituents, especially the heat-labile and light-sensitive compounds (Babu et al., 2018). A study published in 2019 by Latif et al. confirmed that compounds including amines, alcohol, carboxylic acid, hydroxyl groups, phenol, alkanes, amino acids, alkyl halide, and aromatic compounds of watermelon rind using shade drying pre-processing were still preserved and detected.

2.2.3 Oven Drying

Oven drying is considered the most simple and rapid thermal processing technique. Its mechanism involves the moisture exchanges between the plant sample and the hot air transfer by convection and radiation through the drying chamber (Babu et al., 2018). Temperature and drying time depend on the sample amount and plant part: leaves, roots, bark, fruit, or seed. Baeeri et al. (2018) proposed a small size cut of 2 x 1 cm using 60 ˚C hot air-drying mode for the thickness of the watermelon rind sample. Lee & Choo (2020) had a similar opinion by suggesting a small cutting size of 3 × 3 cm for 60 ˚C oven drying. As

for watermelon rind drying conditions, Petchsomrit et al. (2020) used hot air oven drying at 60 °C until a constant weight was obtained for lipid extraction, while Lee & Choo (2020) applied a temperature of 60 ˚C for 24 hours for pectin extraction. Higher temperature and longer drying time were used as watermelon rind contain high water content. However, a shorter period of drying time can preserve the phytochemical constituent in the plant from degradation.

Oven drying is commonly used as it is easy to conduct and low cost. However, the generation of high temperature during the drying process may cause crust formation and hardening on the watermelon rind surface, leading to the deterioration of sample quality and freshness (Calín-Sánchez et al., 2020; Hoque & Iqbal, 2015). A study conducted by Ho et al. (2018) on watermelon rind sample using different oven drying temperatures (40 ˚C and 60 °C) found that the dried sample at 60 °C showed a relatively higher DPPH value than the dried sample at 40 °C. Heating the plant sample at high-temperature results in breaking down free radicals. Ho et al. (2018) also opined that the oven temperature at 40 °C is not suitable for samples rich in moisture content such as watermelon rind because it still comprises a high level of enzymatic activity that will cause the degradation of antioxidant compounds.

2.2.4 Freeze-Drying

Freeze-drying or lyophilization is a low-temperature drying method involving two steps: freezing and drying. The first step in freeze-drying is the raw material reduction temperature until the moisture inside the plant sample forms a solid state. The frozen water is placed under vacuum pressure and removed from the sample by sublimation process as primary drying. Sublimation is the main principle in lyophilization which can achieve at pressure and temperature below triple point (0.01 °C, 0.00603 atm) (Babu et al., 2018; Gaidhani, Harwalkar, Bhambere, & Nirgude, 2015). Under the principle of sublimation, the frozen water is evaporated by heating, changing directly from the solid state to the gaseous state without passing through a liquid phase, as shown in Figure 1. After removing the water vapour in a separate chamber, the sample undergoes secondary drying by slowly warming to room temperature to obtain the final dried sample.

Generally, several studies have suggested freeze-drying as it shows better preservation of plant samples' colour, aroma, and other bioactive compounds (Babu et al., 2018; Thamkaew et al., 2021). A freeze dryer heats a sample at a lower temperature resulting in no thermal degradation of a heat-labile compound and fragrant components (Babu et al., 2018). However, lower phenolic compounds are reported in freeze-drying as compared to hot-air oven drying. A study conducted by Ho et al. (2018) confirmed that the freeze-dried watermelon rind sample presented a significantly lower TPC value (127.93 - 180.58 mg GAE/100 g) than hot-air oven drying (162.33 - 218.39 mg GAE/100 g). Similarly, in the case of DPPH

value, freeze dryer showed lower DPPH values (25.81 - 55.85%) of watermelon rind sample than those obtained using hot-air oven drying (23.49 - 84.88%). These results might be because the high vacuuming process may remove volatile compounds in plant samples (Gaidhani et al., 2015). Besides, the major drawback of freeze-drying is its expensive operation unit.

2.3 Grinding and grading technique

Grinding and sieving are part of the pre-processing involved in the experimental extraction process. The grinding step yields a minor surface contact between the watermelon rind sample and the extraction medium, thus resulting in more efficient extraction. The ideal particle size is smaller than 0.5 mm to achieve an efficient extraction. In the grinding procedure, conventional mortar and pestle, analytical blade mill and electrical blenders are commonly utilized based on preliminary sample conditions. As mentioned in the experimental study by Baeeri et al. (2018), fresh and frozen watermelon rind samples were crushed with a blender to form a fine paste or slurry, while dried watermelon rind was milled into powdery form. Table 2 exhibits previous studies' grinding and grading methods applied to the watermelon rind sample.

A powdered sample undergoes a sieving process to achieve a homogeneous particle size. As the mesh size increases, the size of the opening decrease resulting in the more refined the particles. Homogenized and finer particle size significantly results in a better contact area with extraction solvents, thus optimizing the extraction process (Petchsomrit et al., 2020). Figure 2 shows the effect of increasing surface area in the extraction yield. The grinding process could break the plant cell wall and expose the hidden surface, as shown in Figure 2(b). A large surface area will contact the extraction solvent to enhance the mass transfer efficiency to release active compounds into the solvent.

3.0 Extraction Process

Both conventional and modern extraction techniques have been used to extract watermelon rind samples, depending on the compounds to be extracted. The most common extraction techniques applied by previous studies on watermelon rind samples are maceration, infusion, digestion, reflux, ultrasound-assisted extraction, and microwave-assisted extraction. Table 3 shows the different extraction conditions utilized in different extraction techniques to extract compounds from the watermelon rind sample. In the next section, each watermelon rind extraction method will be elaborated.

3.1 Extraction Method

3.1.1 Maceration

The maceration technique is widely used in plant research since it is the simplest, easiest, and most affordable. In this method, the coarsely powdered plant materials are placed in a container with a solvent. The content can stand at room temperature for at least three days and could be up to months and years based on the sample type with periodic stirring to soften and break the plant's cell wall. The mixture must be strained by filtration or decantation at the end of extraction. The choice of solvents utilized in the soaking process plays a critical role as different solvents extract the different types of compound (Mohammad Azmin, Mustaffa, Wan Alwi, Manan, & Chua, 2014; M S M Nor, Manan, Mustaffa, & Lee, 2017; Mohd Shukri Mat Nor, Abd Manan, Mustaffa, & Suan, 2015). In the watermelon rind sample, both polar and non-polar solvents are used as extractants depending on desired isolated compounds (Mohd Shukri Mat Nor, Abd Manan, Mustaffa, & Suan, 2016).

Maceration is the best technique to extract the thermolabile compound when applied at low temperatures (Abubakar & Haque, 2020; Zhang et al., 2018). Nurdalilah et al. (2018) found that the DPPH value of watermelon rind extracted using the maceration technique was 35.89 %, while the DPPH value was 34.48 % using the infusion extraction method done by Neglo et al. (2021). Both extraction techniques have proven excellent preservation of antioxidant compounds in the extract. Both also resulted in the range (12.90 % to 85.28 %) of another study reported by Ho et al. (2018) using the infusion extraction method. However, considering the major issue of longer extraction time and larger solvent volume waste in maceration, infusion technique is preferable than maceration to extract antioxidant compound in watermelon rind sample.

3.1.2 Infusion Extraction

The infusion technique involved storing samples at room temperature over a short period. The process is suitable for extracting fresh samples with readily soluble bioactive components (Abubakar & Haque, 2020). A study by Petchsomrit et al. (2020) evaluated the effect of different extraction durations (2, 6, 12 and 24 hours) on the watermelon rind sample using hexane (1:5 w/v). The finding showed that the

extraction yield increases as the longer extraction time are applied. The extraction time for 24 hours was the most efficient to extract fatty acids compound in watermelon rind sample with 1.155 % yield compared to 0.187 % at 2 hours (Petchsomrit et al., 2020).

The solvent choice utilized as extractant also plays an essential role in determining the type of compound extracted and the extraction efficiency. Ho et al. (2018) soaked the watermelon rind sample for 24 hours at room temperature using four different solvents: distilled water, methanol, ethanol, and acetone. The outcome showed that water was the most effective solvent to extract phenolic compounds compared to methanol, ethanol and acetone due to its highest polarity (Ho et al., 2018). Furthermore, the infusion method showed better preservation of anthocyanin pigment. 0.02 mg/L of total anthocyanin content (TAC) from the watermelon rind sample was observed by Augustia et al. (2020). The anthocyanin compound is sensitive to high temperature and high light exposure; thus, extraction at low temperature using the infusion technique can retain the pigment compound (Augustia et al., 2020).

3.1.3 Ultrasound-Assisted Extraction (UAE)

Ultrasound-assisted extraction (UAE) or sonication is an advanced technique that uses ultrasound frequencies ranging from 20 kHz to 20 MHz at room temperature or under heat to disrupt plant cell walls for solvent penetration (Abubakar & Haque, 2020). However, the precaution particularly avoiding using a high amount of ultrasound energy as it may cause the degradation of plant active constituents through the formation of free radicals (Abubakar & Haque, 2020)

UAE has been considered the optimal extraction technique for small samples due to the excellent performance in reducing the extraction duration and amount of solvent used but increasing the extraction yield (Abubakar & Haque, 2020). The UAE was found to extract significantly higher amounts (2032 mg/g) of phenolic compounds in a dried watermelon rind sample that was examined by Baeeri et al. (2018) as compared with only 218.39 mg/g of phenolic compounds investigated using infusion technique by Ho et al. (2018). Both extraction methods used water as the extraction solvent. The vast difference in phenolic compound extraction using UAE occurred because of the plant tissue disruption by sound energy, which can increase solvent permeability into inner cell material even without the requirement of high-temperature treatment (Abubakar & Haque, 2020; Arshadi et al., 2016). Most importantly, the extraction process can retain the heat-sensitive compound from thermal degradation.

On the other hand, the solvent choice, either water, aqueous or non-aqueous solvent, and plant operating characteristics also play an essential role in this technique (Arshadi et al., 2016). In the case of

different extraction techniques of watermelon rind sample, the yield of pectin extracted using UAE was only 8.38 % (Lee & Choo, 2020) as compared to 19.3 % of pectin extracted using the maceration technique (Petkowicz, Vriesmann, & Williams, 2016) and 13.4 % of pectin using digestion extraction (Mendez, Fabra, Gomez-mascaraque, Lopez-rubio, & Martinez-abad, 2021). The results might be because, during the recovery process of UAE, smaller pectin molecules may not precipitate and might be eluted out with the solvent (Lee & Choo, 2020).

3.1.4 Microwave-Assisted Extraction (MAE)

Microwave-assisted extraction (MAE) is an advanced extraction technique used to extract plant constituents such as phenolics, pectin, essential oils and other organic compounds (Arshadi et al., 2016). Generally, two types of MAE methods are solvent-free extraction to extract volatile compounds, and solvent extraction for non-volatile compounds (Zhang et al., 2018). This technique is considered a highly selective method, favouring polar solvents only. Prakash Maran et al. (2014) used water as a polar solvent to extract pectin from the watermelon rind sample. Water solvent with high dielectric constant induced dipole rotation and ionic conduction, thus accelerating solvent penetration and increasing pectin extraction yield.

Ramakrishnan et al. (2020) recommended the MAE conditions at 190 °C for 30 min with operating of 230 V and 1200 W were able to increase the production yield of the watermelon rind sample 23 times than standard yield. For pectin extraction, Prakash Maran et al. (2014) determined the optimum MAE conditions at 477 W power, 128 s irradiation time, 1.52 pH condition, and 1:20.3 g/ml solid-to-liquid ratio were able to extract the highest yield of watermelon rind pectin (25.79 %). Increasing the microwave power could increase the extraction yield as the heat transferred by ionic conduction enhances the migration of analytes from the plant matrix into solvent through molecular interaction (Abubakar & Haque, 2020; Zhang et al., 2018). Sudden rise in temperature and internal pressure can also accelerate the plant cell rupture, promoting the exudation of pectin into the solvent (Prakash Maran et al., 2014).

Prakash Maran et al. (2014) investigated the effect of different irradiation times ranging from 60 s to 180 s on pectin yield. The result proved that the pectin yield rapidly increased to the maximum yield at 128 s before gradually dropping. This study confirmed that the extended irradiation time might destroy the pectin chain molecule.

A previous study by Ramakrishnan et al. (2020) reported that the microwave pre-treated watermelon rind substrate showed 23 times increase in itaconic acid production compared to the raw

watermelon rind sample. Pre-treatment using microwave radiation can assist the conversion of complex compounds in a watermelon rind sample into a simple compound (Ramakrishnan et al., 2020).

4.0 Factors that influence the extraction of watermelon rind

Several factors enhance the extraction yield of watermelon rind samples, such as the extraction temperature, type of solvents, solvent-to-solid ratio, extraction duration, solvent pH and particle size of the raw materials (Lee & Choo, 2020; Zhang et al., 2018), could be taken into account before starting the isolation process.

4.1 Extraction temperature

High extraction temperature can increase the solubility and diffusion between particle and solvent, but it may cause loss of solvents through evaporation and the degradation of thermo-labile components. In the case of watermelon rind extract, thermo-labile compounds such as phenolic and flavonoid are best to be extracted using maceration, infusion and ultrasound-assisted extraction at room temperature as the techniques do not involve high extraction temperature. On the other hand, heat-stable materials such as polysaccharides (30 °C to 70 °C) could be extracted using digestion. In comparison, pectin (70 °C to 95 °C) could be extracted using digestion, reflux, ultrasound-assisted and microwave-assisted extraction.

4.2 Solvent-to-solid ratio

As for the solid-to-solvent ratio, the higher the solid ratio leads to a high extraction yield. A protocol developed by Prakash Maran et al. (2014) using a different solid-to-solvent ratio of 1:10 to 1:30 (water as solvent extractant) were used to identify the optimum ratio of watermelon rind pectin extraction to solvent. The author found that the yield of pectin significantly increased with an increasing solid-to-liquid ratio up to 1:20.3 before gradually dropping with a further increased ratio. The finding could be used to reveal that polar solvent is favourable to accelerate the heat transfer through molecular interaction between solvent and material. However, too high solvent polarity could decrease the microwave adsorption of material because of more energy absorbed by the solvent (Prakash Maran et al., 2014). In contrast, Lee & Choo (2020), in their study, stated that the solvent-to-solid ratio did not show a significant effect ($p > 0.05$) on the yield of pectin extraction from watermelon rind sample. The author illustrated a detailed study using Design Expert 7.0 software for pectin extraction optimization.

4.3 Extraction duration

The conventional extraction methods usually cause a longer extraction duration (Mohammad et al., 2019). Various modern extraction techniques such as reflux, ultrasound-assisted, and microwave-assisted extraction have been introduced and implemented in watermelon rind extraction. The extraction of watermelon rind by using ultrasound-assisted extraction for 90 minutes by Baeeri et al. (2018) showed higher total phenolic content (691.4 mg/g) as compared to those obtained using the maceration technique for three days (2.9330 g/mg) by Nurdalilah et al. (2018). A shorter extraction duration is more favourable for retaining the bioactive constituent and considering the experimental design time.

4.4 Solvent type

Solvent type is crucial in plant extraction because the target compounds are dependent on the solvent used and its polarity. Various factors such as solvent selectivity, safety, cost, reactivity, recovery, viscosity, and boiling point, as shown in Figure 3 (Abubakar & Haque, 2020; Zhang et al., 2018), should be considered in selecting an extracting solvent.

Table 4 exhibits the solvent polarity chart classified according to their relative polarity. Augustia et al. (2020) reported that ethanol solvents with a low boiling point of 78.37 °C are capable of retaining anthocyanin compound (0.02 – 0.05 mg/L) from watermelon rind is a susceptible compound to high temperature and high light exposure. On the other hand, Baeeri et al. (2018) reported that extraction using methanol: water: acetic acid (70: 29: 1) is the most efficient extraction solvent to extract phenolic and amino acid content in watermelon rind sample as compared to methanol: water (70: 30) and water (100 %). The case studies proved that using several extraction solvents of different polarities and viscosity could increase the extraction efficiency. Solvent polarity plays a crucial role in increasing compounds' solubility and selectivity. Co-extraction between methanol, water and acetic acid can efficiently extract both polar and less polar compounds. Extracting solvents is more efficient when their relative polarity value is near the solute's polarity (Zhang et al., 2018).

In addition, ethanol and methanol are known as universal solvents as it is widely used in phytochemical extraction (Zhang et al., 2018). The alcohol solvents play an essential role in extracting certain phytochemicals from plant parts due to their polarity. The usage of ethanol and methanol as

extracting solvents to extract phenolic compounds in watermelon rind were observed in several studies. Nurdalilah et al. (2018) and Neglo et al. (2021) utilized methanol to extract phenolic compounds in watermelon rind resulting in 2.9330 g/mg and 0.026 g/mg, respectively. Ho et al. (2018) further expanded in their study by comparing methanol and ethanol solvent resulting in ethanol (147.58 – 166.68 mg GAE/100 g dry matter) was less efficient than methanol (169.15 – 198.56 mg GAE/100 g dry matter) in the extraction of total phenolic contents of the watermelon rind. It might be because methanol solvent has higher extraction polarity than ethanol. Both methanol and ethanol are proton donors. However, methanol has better solvation of phenolic molecules due to shorter methyl radical than ethyl radical in ethanol (Ho et al., 2018).

5.0 Conclusion

Watermelon rind is readily available and discarded due to its unappealing flavour despite its beneficial properties in the food, cosmetics, and wastewater treatment industry. The benefit of watermelon rind waste can be fully utilized by proper selection of pre-processing and processing techniques. This article could help determine the factors affecting the extraction yield of the watermelon rind. Optimal drying is essential for preserving phytochemical constituents and preventing the sample from deterioration. The polar extracting solvent was found to be more efficient in extracting watermelon rind bioactive compounds, mainly antioxidant compounds, as it possessed more hydrophilic behaviour. Conventional and modern extraction techniques are relevant and can efficiently extract watermelon rind based on availability, experimental design and desired constituent of the end product. In future endeavours, more research and review are required to analyze watermelon rind samples further using proper and detailed analysis techniques such as antioxidant analysis, chromatography analysis, or morphological image. Without a doubt, watermelon rind waste deserves more attention for its potential applicability in various industries.

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Table 1. Analyzed properties for different drying methods of watermelon rind sample

Drying Condition	Antioxidant	Aromatic	Sugar	Pectin	Cellulose	Lipid	References
Sun drying	/		/		/	/	(Egbuonu 2015a; Shivapour et al. 2020; Neglo et al. 2021)
Shade Drying	/	/			/	/	(Latif et al., 2019)
Oven Drying 40 °C	/			/	/	/	(Ho et al. 2018; Sanwiriya and Suleiman 2019; Gomaa 2020)
Oven Drying 60 °C	/			/	/	/	(Prakash Maran et al. 2014; Naknaen et al. 2016; Baeeri et al. 2018; Ho et al. 2018; Nurdalilah et al. 2018; Adegunwa et al. 2019; Sanwiriya and Suleiman 2019; Lee and Choo 2020; Petchsomrit et al. 2020)
Vacuum drying 60 °C			/				(Kumar et al. 2012)
Freeze Drying	/						(Ho et al. 2018)

Drying Condition	Antioxidant	Aromatic	Sugar	Pectin	Cellulose	Lipid	References
Ventilated dryer 50 °C	/		/		/	/	(Ho and Che Dahri 2016)
Cabinet dryer (55/60/65 °C)					/	/	(Hoque and Iqbal 2015)

Table 2. Grinding and grading methods applied to watermelon rind sample

Grinding	Equipment	Grading	End Product	Source
Ground		Fine powder	Lotion emulsion	(Petchsomrit et al. 2020)
• Ground • Milled	Blender, analytical mill IKA A-11		Pectin	(Petkowicz et al. 2016)
Ground 2 mins using high-speed spin	Blender			(Augustia et al. 2020)
• Milled (oven drying sample)	Analytical mill	• Fine powder		(Sanwiriya and Suleiman 2019)

Grinding	Equipment	Grading	End Product	Source
• Ground (foam mat drying sample)		• 400 mesh		
Blend				(Neglo et al. 2021)
Ground 30 mins	Grinder (Remi Make)	100 µm	As substrate for itaconic acid production	(Ramakrishnan et al. 2020)
Ground	Blade mill			(Lee and Choo 2020)
Ground		125 µm		(Nurdalilah et al. 2018)
• Ground (fresh and frozen samples) • Milled (dried sample)	Blender, mill			(Baeeri et al. 2018)
Ground	Blender	Powdered by freeze dryer	Lotion	(Alamsyah et al. 2016)
Blend			Production of invertase	(Arise et al. 2020)

Grinding	Equipment	Grading	End Product	Source
			enzyme	
Grated				(Akshaya et al. 2018)
Milled		Powdery form	Watermelon rind flour	(Adegunwa et al. 2019)
Ground	Stainless steel Grinder	Powder		(Ho et al. 2018)
Milled	Arthur Thomas Laboratory Mill	Powder		(Egbuonu 2015a, b)
		Powder		(Kumar et al. 2012)
Ground	Grinder	Powder	Watermelon rind flour	(Hoque and Iqbal 2015)
Ground	Laboratory mill	Fine powder (100 mesh)	Watermelon rind flour	(Naknaen et al. 2016)
Ground	Laboratory disk mill (Mulinex Depose-Brevete SGCG,France)	Fine powders (212 µm)	Watermelon rind powder	(Shivapour et al. 2020)

Grinding	Equipment	Grading	End Product	Source
Ground	Laboratory mill	Fine powder (250 µm)		(Ho and Che Dahri 2016)
Pulverized		40 Mesh	Pectin	(Prakash Maran et al. 2014)

Table 3. Extraction condition for different extraction techniques of watermelon rind sample.

Method	Solvent	Temperature	Pressure	Extraction time	Solvent-to-sample ratio	Extraction yield, % yield	Product extracted	Source
Maceration	80 % methanol	Room temperature	Atmospheric	3 days	10 ml:1 g		Phenolic compound Flavonoid compound Tannin compound	Nurdalilah et al. (2018)
Infusion	80 % ethanol contained with 0.1 % hydrochloric acid	Room temperature	Atmospheric		35 ml: 30 g		Anthocyanin compound Flavonoid compound Polyphenol compound	Augustia et al. (2020)

Method	Solvent	Temperature	Pressure	Extraction time	Solvent-to-sample ratio	Extraction yield, % yield	Product extracted	Source
	Hexane	Room temperature	Atmospheric	2 h 6h 12h 24 h	5 ml: 1 g	0.187 % 0.585 % 1.155 % 1.422 %	Lipid extraction	Petchsomrit et al. (2020)
	• Distilled water • Methanol • Ethanol • Acetone	Room temperature	Atmospheric	24 h	50 ml: 0.5 g		Phenolic compound	Ho et al. (2018)
	80 % methanol	Room temperature	Atmospheric		100 ml: 10 g		Flavonoid compound	(Egbuonu 2015a)
	Distilled water	Room temperature	Atmospheric	24 h	50 ml: 5 g		Cyanide compound	(Egbuonu 2015a)

Method	Solvent	Temperature	Pressure	Extraction time	Solvent-to-sample ratio	Extraction yield, % yield	Product extracted	Source
	Methanol	Room temperature	Atmospheric	24 h	400 ml: 1116 g	1.79 %	Phenolic compound Saponin compound Alkaloid compound Free reducing sugar	Neglo et al. (2021)
	80 % ethanol	Room temperature	Atmospheric	3 h	20 ml: 1 g		Phenolic compound	(Naknaen et al. 2016)
	70 % methanol	Room temperature	Atmospheric	24 h	100 ml: 1 g		Phenolic compound	(Ho and Che Dahri 2016)
Digestion	Distilled water	30 °C to 70 °C	Atmospheric	50–100 min	5 ml: 50 g		Polysaccharides extraction Monosaccharides extraction	Romdhane et al. (2017)
	Acidic aqueous solutions	95 °C	Atmospheric	90 min	20 ml: 1 g		Pectin extraction	Mendez et al. (2021)

Method	Solvent	Temperature	Pressure	Extraction time	Solvent-to-sample ratio	Extraction yield, % yield	Product extracted	Source
Liquid-liquid extraction	Methanol: dichloromethane: water (0.3:4:1 v/v/v)	Room temperature	Atmospheric				Sugar extraction	(Kumar et al. 2012)
Reflux	0.1 M nitric acid	Boiled	Atmospheric	20 min		13.4 %	Pectin extraction	Petkowicz et al (2016)
Ultrasound-assisted extraction	85 % ethanol	70 ˚c	Atmospheric	20 min			Pectin extraction	Lee & Choo (2020)
	• Water 100% • Methanol: water (70:30) • Methanol: water: acetic acid (70:29:1)	Room temperature	Atmospheric	90 min			Phenolic compound Amino acid compound	Baeeri et al. (2018)
Microwave-assisted technique (MAE)		190 °c	Atmospheric	30 min			As substrate for itaconic acid production.	Ramakrishnan et al. (2020)

Method	Solvent	Temperature	Pressure	Extraction time	Solvent-to-sample ratio	Extraction yield, % yield	Product extracted	Source
	Distilled water		Atmospheric		10-30 ml: 1 g	25.79 %	Pectin extraction	(Prakash Maran et al. 2014)

Table 4. Solvent polarity chart of watermelon rind extractant solvent

Polarity	Solvent	Relative Polarity	Functional Group	Watermelon rind extract	Source
More polar	Water	1.000	Water	Phenolic, flavonoid, galactose, arabinose, glucose, galacturonic acid, rhamnose, mannose, xylose, glucuronic acid, fucose, fructose, anhydrouronic acid, amino acid, sucrose, cyanide	(Kumar et al. 2012; Egbuonu 2015a; M. B. et al. 2017; Baeeri et al. 2018; Ho et al. 2018; Mendez et al. 2021)
	Glycerine	0.812	Hydroxy		
	Methanol	0.762	Alcohol	Phenolic, flavonoid, tannin, saponin, alkaloid, free reducing sugar, amino acid, rhamnose, sucrose, glucose, mannose	(Kumar et al. 2012; Egbuonu 2015a; Ho and Che Dahri 2016; Baeeri et al. 2018; Ho et al. 2018; Nurdalilah et al. 2018; Neglo et al. 2021)
	Ethanol	0.654	Alcohol	Anthocyanin, flavonoid, phenolic, pectin	(Naknaen et al. 2016; Ho et al. 2018; Augustia et al. 2020; Lee and Choo 2020)
	Acetic acid	0.648	Carboxylic acid	Phenolic, amino acid	(Baeeri et al. 2018)
	Acetonitrile	0.460	Nitrile		
	Ethyl ether	0.433	Ether		
	Acetone	0.355	Aldehyde	Phenolic, flavonoid	(Ho et al. 2018)
	Pyridine	0.302	Amine		
	Chloroform	0.259	Alkyl halide		
	Tetrahydrofura	0.207	Heterocycli		

Polarity	Solvent	Relative Polarity	Functional Group	Watermelon rind extract	Source
	n		c ether		
	Carbon tetrachloride	0.052	Alkyl halide		
	Toluene	0.009	Aromatic		
	Hexane	0.009	Alkane	Myristic acid, palmitic acid, stearic acid, oleic acid, linoleic acid, arachidic acid	(Petchsomrit et al. 2020)
Less polar	Cyclohexane	0.006	Alkane		

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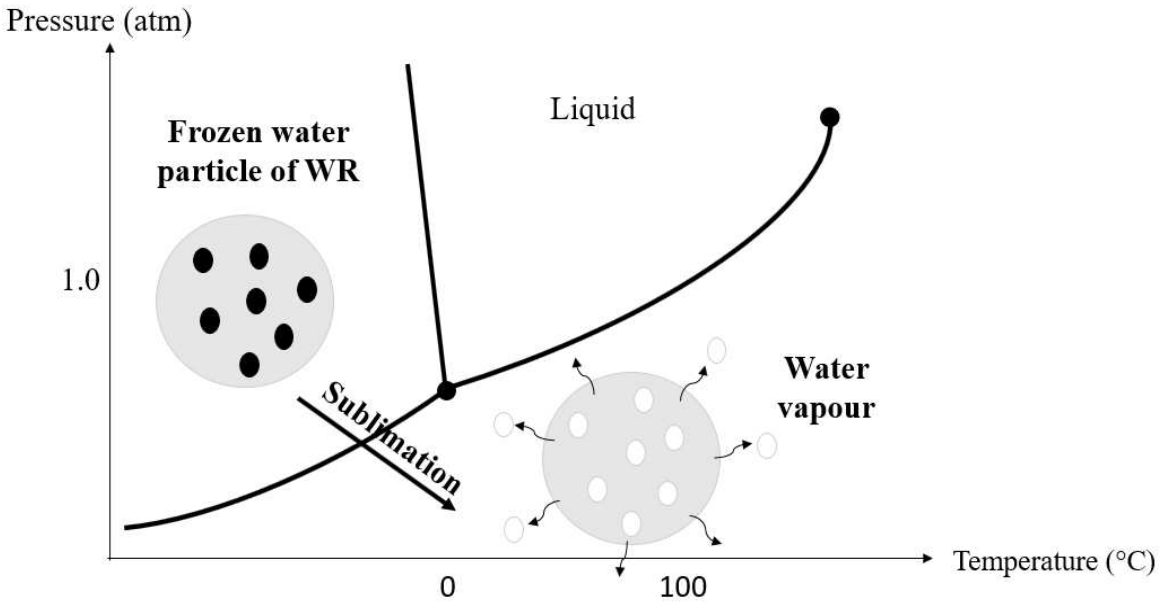

Fig. 1. The heat transfer in watermelon rind (WR) sample according to the principle of sublimation

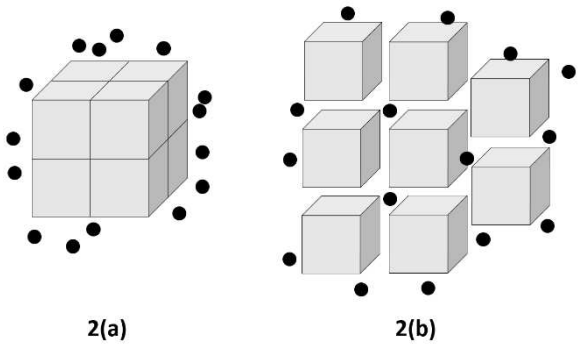

Fig. 2. Increasing surface area affecting reaction rates.

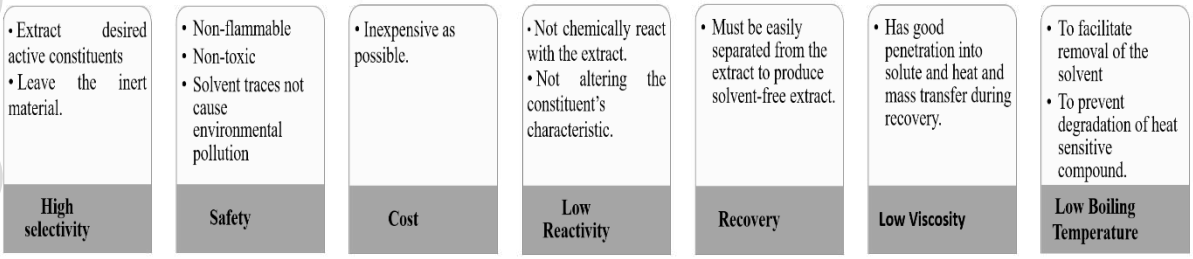

Fig. 3. Factors to be considered when selecting an extracting solvent