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Research Article

Antioxidant Stability of Moringa Leaves Extract Powders Obtained by Cocrystallization, Vacuum Drying, and Plating

Bambang Nurhadi,[1] **Fatsyarien Citra Angiputri**,[1] **Robi Andoyo**,[1] **Riksfardini Annisa Ermawar**,[2] **and Rudy Adi Saputra**[1]

[1]*Department of Food Industrial Technology, Universitas Padjadjaran, Jl. Raya Bandung Sumedang KM.21, Hegarmanah, Kec. Jatinangor, Kabupaten Sumedang, Jawa Barat 45363, Indonesia*
[2]*Research Center for Biomass and Bioproducts, National Research and Innovation Agency, Jalan Raya Jakarta Bogor KM. 46, Cibinong, Jawa Barat 16911, Indonesia*

Correspondence should be addressed to Bambang Nurhadi; bnhnur@gmail.com

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Cocrystallization, vacuum drying, and plating are three potential applications to preserve the antioxidant activity of moringa leaves. Moringa leaves extract was incorporated with sucrose at the same concentration (7 : 100, solid : solid) for all applications and stored for 30 days. This study aims to examine the effects of each application on the antioxidant stability of moringa leaves extract powders. Morphological properties by SEM showed that cocrystallized powders exhibited porous, agglomerated crystals, vacuum dried powders exhibited agglomerated crystals, and plated powders exhibited layered crystals. Based on XRD and hygroscopicity results, cocrystallization produced powders with the highest crystallinity, i.e., 69.11%, and the lowest hygroscopicity, i.e., $0.26 \times 10^{-4} \pm 0.02 \times 10^{-4}$ g H_2O/g solid/minute due to the slow water intake of the crystalline structure. Powders with the strongest initial antioxidant activity were obtained from cocrystallization, i.e., 3647.96 ± 20.29 ppm and followed by vacuum drying, i.e., 4378.51 ± 26.29 ppm. The least antioxidant activity was obtained from plating, i.e., 4733.46 ± 31.91 ppm. During 30 days of storage, powders obtained by cocrystallization maintained the most stable antioxidant activity (91.81–91.12%). The results indicated that the high temperature used in the process was likely to impact crystalline structure through the pore formation, which entrapped bioactive compounds and resulted in strong antioxidant activity. While, vacuum drying resulted in powders with a lower but increased antioxidant activity (84.06%–86.43%). In contrast to the other two applications, plating resulted in a decreased antioxidant activity (83.77–82.25%). This study suggests that application of cocrystallization produced moringa leaves extract powders with the strongest and most stable antioxidant activity during storage. Preserving the antioxidant stability of plant extract has been one of the major drives in the development of food encapsulation technology. Cocrystallization and vacuum drying are two relatively novel, less common techniques offering a simpler and more cost-effective method, but their effect on the antioxidant stability of moringa leaves extract has not yet been studied. This study discloses the effects of cocrystallization, vacuum drying, and plating (alternative extract incorporation method) on the antioxidant stability of moringa leaves extract powders. The results indicated that the three methods produced powders with high crystallinity and stable antioxidant stability during storage. Among the three methods, cocrystallization was the method that resulted in powders with the strongest and most stable antioxidant activity.

1. Introduction

Moringa leaves are leaves derived from *Moringa oleifera* plant, which has been cultivated and consumed worldwide because of its rich nutrient content[1]. Several bioactive compounds such as niazimicin and niazinin [2], quercetin, kaempferol, thiamine, O-coumaric acid, caffeic acid, gallic acid, and chlorogenic acid [3] possessed by the leaves are known for their antioxidant activity. Antioxidants from the compounds could act as neutralizers for free radicals in

many degenerative diseases such as hypercholesterolemia [4], hypertension [5], diabetes [6], liver disease [7], cancer [8], and inflammation [9].

The emerging challenge in utilizing moringa leaves as a functional food is its instability and unpleasant flavour when extracted in an aqueous form [10, 11]. Several studies have suggested encapsulation technology, such as spray-drying and freeze-drying [12], as a potential solution since it enables the protection of bioactive compounds from external light, moisture, and oxygen [13]. However, common encapsulation techniques like spray-drying and freeze-drying are often less accessible for the general public due to their high-end investments. Thus, finding a simpler but still effective technique to preserve moringa leaves' antioxidant compounds is imperative.

Recently, a simpler preservation method has been introduced and developed in order to preserve antioxidant compounds, such as cocrystallization, which utilizes sucrose to provide protection for antioxidant compounds [14]. Cocrystallization offers a flexible, simple, and low-cost process, since it can be performed through various methods, including traditionally because its principle relies on continuous agitation and sufficient heating, which can be provided by hand agitation without sophisticated equipment [15]. Cocrystallization of marjoram extract was reported to perform higher stability towards phenolic compounds (80–90% TPC retention) compared to the common spray-drying method (70–80% TPC retention) [16]. Several studies also reported satisfactory storage antioxidant stability on cocrystallization carried out to yerba mate tea which shows changes less than 50% in extreme 4 months storage condition of 75% RH, 40°C [15], and chokeberries which exhibits antioxidant activity of 80–90% during 30 days of storage [17].

Although the cocrystallization method is able to preserve antioxidant compounds in a simpler technique compared to the common spray-drying method, the technique itself requires high temperature, which is a possible drawback. The usage of high heating above 100°C might impair the antioxidant activity [14]. Lower temperatures can be provided with another encapsulation method known as vacuum drying [18]. Due to vacuum pressure, this process is conducted at a temperature less than 100°C (the boiling point of water at atmospheric pressure), followed by minimal air flow [18]. Vacuum drying of sea buckthorn juice has demonstrated an improved antioxidant activity of 50% during 6 months storage [19].

In terms of extract incorporation with minimal heating, plating served as an alternative to produce powders containing natural antioxidants with a very simple, cost-efficient process. However, plating methods may have a tendency to provide low protection for antioxidant compounds during storage [20]. Previous examination of the plating technique effects towards antioxidant stability of extracts has not been reported. To the best of our knowledge, there has not been any study reported on the antioxidant stability of moringa leaves extract obtained by cocrystallization, vacuum drying, and plating. Therefore, this study is being conducted to examine the effects of cocrystallization, vacuum drying, and plating on the antioxidant stability of moringa leaves extract powders.

2. Materials and Methods

2.1. Materials. Moringa leaves extract powder were prepared using moringa leaves extract and commercial sucrose. Moringa leaves extract was prepared using dry moringa leaves (moisture content 11.90% wb) purchased from Blora Regency, Central Java, Indonesia, and distilled water. Supersaturated natrium chloride (NaCl) (Merck, Germany) was used for hygroscopicity analysis. Folin-Ciocalteu (Merck, Germany), gallic acid (Sigma-Aldrich, USA), and natrium carbonate (Na_2CO_3) (Merck, Germany) were used for total phenolic analysis. Aluminium chloride ($AlCl_3$) (Sigma-Aldrich, USA), potassium acetate (CH_3COOK) (Merck, Germany), and quercetin (Sigma-Aldrich, USA) were used for total flavonoid analysis. DPPH (2,2-diphenyl-1-picrylhydrazyl) (Tokyo Chemical Industry Co., Ltd., Japan) was used for antioxidant activity analysis. Methanol (Merck, Germany) was used in total flavonoid and antioxidant activity analysis.

2.2. Preparation of Moringa Leaves Extract. The aqueous extract was prepared by following the modification method reported by López-Córdoba et al. [15]. Grounded dry moringa leaves (200 g) were mixed with distilled water (1 : 10) and boiled at 90 ± 2°C for 40 minutes. The extract was filtered with vacuum filter and its filtrate was contained in a dark bottle. The extract was stored at refrigerated temperature (2.3 ± 1°C) until use.

2.3. The Preparation of Cocrystallized Moringa Leaves Extract Powders. Aqueous extract (100 mL) and sucrose (71.4 g) were prepared in a ratio of extract's total solid to sucrose (7 : 100, solid : solid). The extract was mixed with sucrose on a wok with continuous stirring until the temperature reached 105°C–90° Brix solution. The heat was immediately turned off, while the stirring was maintained until cocrystals were formed. The formed agglomerate was dried with cabinet oven (Shel Lab, USA) at 50°C until constant weight, grounded with a grinder (Getra Herb Grinder IC-06B, China). It was packed in a sealed aluminium foil packaging and stored in a desiccator until further analysis.

2.4. The Preparation of Vacuum-Dried Moringa Leaves Extract Powders. The aqueous extract (100 mL) and sucrose (71.4 g) were prepared in a ratio of extract's total solid to sucrose (7 : 100, solid : solid). The mixture was dissolved and dried with cabinet oven (Shel Lab, USA) at 50°C until the mixture became thick and rubbery. The mixture was dried at a vacuum oven at 70°C (B-One VOV-50 Vacuum Drying Oven, China) and grounded with grinder (Getra Herb Grinder IC-06B, China). It was packed in a sealed aluminium foil packaging and sorted in a desiccator until further analysis.

2.5. The Preparation of Plated Moringa Leaves Extract Powders. The aqueous extract (600 mL) was evaporated with rotary evaporator (Buchi R-210 Rotavapor, Switzerland) until it was reduced to 37 mL and a thick extract was formed. Total solid of thick extract was calculated in °Brix. Sucrose was weighed in a ratio of thick extract's solid to sucrose (7 : 100, solid : solid). The thick extract (7.54 g) was then poured into sucrose (71.42 g) and grounded with Getra Herb Grinder IC-06B, China. The resulting powders were dried with cabinet oven (Shel Lab, USA) at 50°C until constant weight. It was packed in a sealed aluminium foil packaging and sorted in a desiccator until analysis.

2.6. Scanning Electron Microscopy (SEM). Scanning Electron Microscopy was performed to characterize powders morphologically. Powder samples on a sample grid was coated with carbon tape and analysed with FE-SEM (Thermo Scientific-Quattro S, USA) on 5000x magnification with high vacuum method and 3.00 kV voltage.

2.7. X-Ray Diffraction Analysis (XRD). Powder's crystalline patterns were studied with X-ray diffraction analysis. The analysis used X-ray diffractometer (X-ray Diffraction Shimadzu 7000s, Kyoto Japan) at 40 kV and current of 30 mA. Data were collected between 0 and 80 at 2θ angle. The X-ray diffraction patterns of powdered sucrose were used as the control and patterns of powdered moringa leaves extract were then determined.

2.8. Fourier Transformed Infrared Spectroscopy (FTIR). Sucrose powder with and without moringa leaves extracts were evaluated for their chemical structure using attenuated total reflectance (ATR)-Fourier transform infrared (FTIR) spectrometer (Spectrum two, Perkin Elmer, USA). Force gauge (FG) was pressed manually onto diamond plate containing the samples until spectra was obtained. 16 scans at 4000–400 cm^1 were taken per experiment with a resolution of 4.0 cm^{-1} and data interval 1 cm^{-1}.

2.9. DPPH Scavenging Activity and Antioxidant Stability. Antioxidant activity is expressed through free radical scavenging activity shown towards DPPH (1,1-diphenly-2-picrylhydrazly) reagent. DPPH reagent was prepared in a methanol solution (160 ppm). Powder samples of 300 g were diluted in 25 mL of methanol. Diluted samples were mixed with 0.5 mL of DPPH reagent and incubated for 30 minutes. Its absorbances were measured using spectrophotometer UV VIS-92000 (Beijing Rayleigh Analytical Instrument Co., Ltd., China). Initial antioxidant activity was expressed with IC_{50} value, whilst DPPH scavenging activity after 30 days of storage is expressed as inhibition percentage (%) of DPPH free radical calculated with the following equation:

$$I(\%) = \frac{Abs_b - Abs_s}{Abs_b}. \quad (1)$$

2.10. Colour Attributes. Powder samples were placed on a Petri dish until all glass surfaces were covered. Petri dish containing samples was placed on chromameter (Spectrophotometer CM-5, Konica Minolta, Tokyo, Japan). Sample's colour attributes were captured by the machine using Spektra Magic NX software. Values of L^*, a^*, and b^* were recorded and used to characterize the powder's colour.

2.11. Total Phenolic Content. Samples were weighed (500 mg) and diluted in 25 mL distilled water. An aliquot of 1 mL of samples was mixed with 0.5 mL of Folin-Ciocalteau reagent and 2.5 mL of 20% Na_2CO_3. After 40 minutes of incubation, the absorbances were measured at 725 nm using a spectrophotometer, the UV VIS-92000 (Beijing Rayleigh Analytical Instrument Co., Ltd., China). Gallic acid was used as standard.

2.12. Total Flavonoid Content. Samples were weighed (200 mg) and diluted in 10 mL distilled water. An aliquot of 1 mL of samples were mixed with 3 mL of methanol, 0,2 mL of 10% $AlCl_3$, and 0,1 mL of CH_3COOK 1 M. After 30 minutes of incubation, the absorbances were measured at 425 nm using a spectrophotometer UV VIS-92000 (Beijing Rayleigh Analytical Instrument Co., Ltd., China). Quercetin was used as standard.

2.13. Hygroscopicity Rate. The hygroscopicity rate was determined by the rate of moisture absorbed by the powders when exposed at 75% RH and room temperature. An amount of the sample (0.5 g) was placed in a desiccator containing supersaturated NaCl (75 ± 2% RH). The sample was weighed every 60 minutes for 5 hours. Hygroscopicity rate was calculated from the slope of moisture content on a dry basis against time (in minutes).

2.14. Angle of Repose. The angle of repose was conducted to express the flowability of the powders. Powder samples (10 mg) were poured through a 75 mm funnel (Pyrex, USA). The angle of the powder sample's pile was measured by the following calculation:

$$\tan \alpha = \frac{x}{y}, \qquad \alpha = arc \tan \frac{x}{y}. \quad (2)$$

3. Results and Discussion

3.1. Morphological Properties. Figure 1 displays the comparative variation in the morphology of moringa leaves extract powders obtained from three applications. Sharp edges were observed in the three powders, which indicates that the crystalline nature of sucrose was unchanged after extract addition. Figure 1(a) shows that cocrystallization resulted in irregular, agglomerated crystals. Voids and agglomeration observed in the structure increased the surface

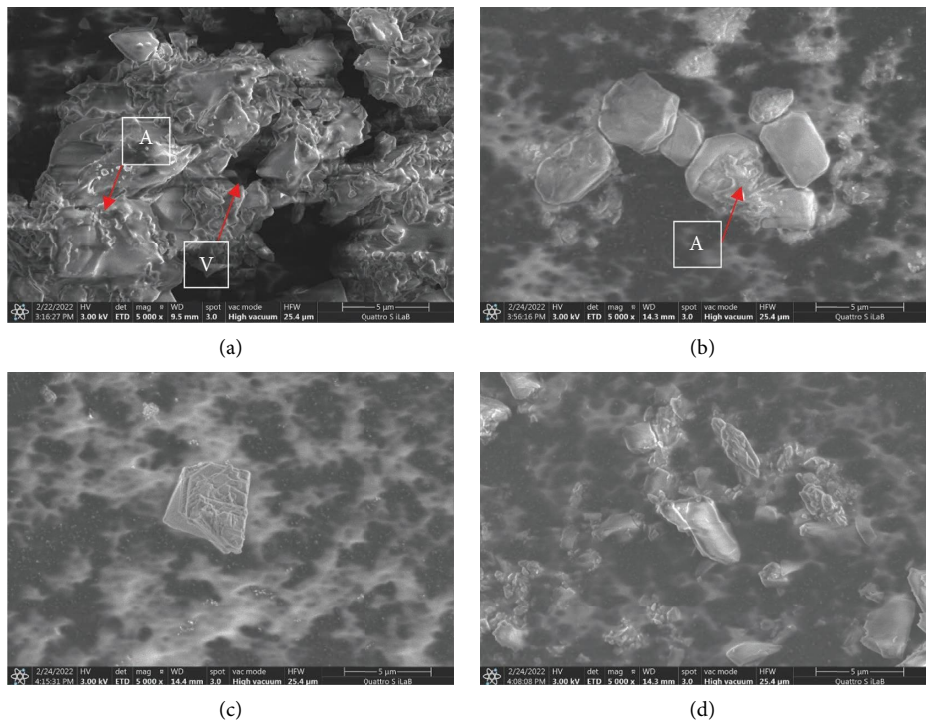

Figure 1: Scanning electron microscopy (SEM) images with 5000x magnification of cocrystallized moringa leaves extract powders (a). Vacuum-dried moringa leaves extract powders (b). Plated moringa leaves extract powders (c). Sucrose (d), V = void, and A = agglomeration.

area for incorporation of bioactive compounds [21]. Voids also represented the entrapment location of active compounds in moringa leaves extract. The results are in agreement with morphological characterizations shown by cocrystallized *Basella rubra* extract [22] and cocrystallized *Aronia* extract [17].

Figures 1(b) and 1(c) show that powders obtained from vacuum drying and plating had an uneven surface due to the deposition of moringa leaves extract on the sucrose crystals. Voids were not present in the microscopic structure of vacuum-dried and plated powders, which indicates that compounds from moringa leaves extract were not entrapped into sucrose structure but rather created a layer on the surface of sucrose as carrier material. However, partial agglomeration in vacuum-dried powders was found in Figure 1(b), which meant vacuum-dried powders had an increased surface area for extract incorporation. According to literature, sucrose used as a control has a perfect cubical structure with neat edges [23]. However, Figure 1(d) showed small-sized sucrose crystals with uneven edges. This difference was likely caused by the grinding process, which has impaired and reorientated the alignment of sucrose crystal molecules.

3.2. XRD (X-Ray Diffraction) Analysis. The crystallinity pattern of moringa leaves extract powders and sucrose as a control was presented in Figure 2. It was observed that all treatments did not change the crystallinity pattern of sucrose, as supported by other studies [22–24]. This signifies that the extract is not crystallized during the process and can be well incorporated in sucrose agglomerates.

Degree of crystallinity obtained from XRD analysis resulted in sucrose having the highest crystallinity (76.86%), followed by cocrystallization (69.11%), vacuum drying (67.59%), and plating with the least crystallinity (66.41%). Even though slight decreases were observed in extract powders compared to sucrose, all samples are still identified as strong crystalline according to the classification done by Bhandari and Hartel [25], which grouped molecules with a crystallinity value above 50% as strong crystalline, molecules with a crystallinity value of 20–50% as medium crystalline, and molecules with a crystallinity value less than 20% as weak crystalline.

High crystallinity of cocrystallized powders was to be expected since it introduces re-crystallization of sucrose crystals through heating and continuous agitation [14]. The crystallinity of vacuum-dried powders, however, differs with another study, which claims that the quick process of vacuum drying resulted in lower crystallinity because of inadequate time for sucrose molecules alignment [26]. High crystallinity from vacuum-dried powders might be caused by pretreatment of cabinet-drying which gives longer time for sucrose molecules to create crystals. This result also matches the micrograph of the crystal structure of vacuum-dried powders shown by SEM. Least crystallinity observed on plated powders might be caused by intensive grinding which is known to decrease crystallinity through reorientation of crystals by repeated fracturing and smealting [27, 28].

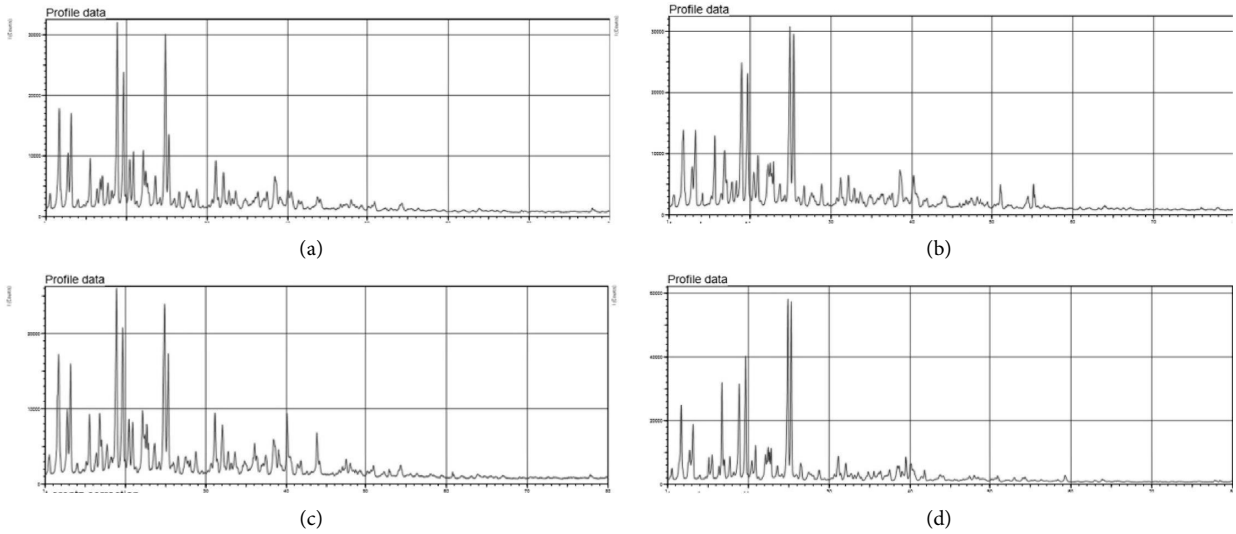

Figure 2: The XRD pattern of cocrystallized moringa leaves extract powders (a). Vacuum-dried moringa leaves extract powders (b). Plated moringa leaves extract powders and sucrose (d).

3.3. Fourier Transform Infrared Spectroscopy (FTIR) Analysis. Figure 3 displays the spectrum provided by FTIR analysis of the moringa leaves extract powders. FTIR results of sucrose (*D*) and powdered moringa leaves extract from cocrystallization (*A*), vacuum drying (*B*), and plating (*C*) showed strong peaks at the ranges of 3320–3400 cm^{-1}, 1000–1300 cm^{-1}, and 987–988 cm^{-1} which represented stretching of the O-H group due to strong intermolecular bonds, the C-O group, and the C-C bending bond, respectively. The powdered moringa leaves extract exhibited typical sucrose molecule bonds, in agreement with other researchers studying extracts powdered with sucrose [15, 29, 30].

Moringa leaves extract (*E*) exhibited a strong, broad, peak observed at 3307.20 cm^{-1} from moringa leaves extract, which depicted the presence of –OH group. The extract also exhibited a medium peak at 1637.99 cm^{-1}, which represented vibration from stretching of the C=C group. Meanwhile, weak peak at 1416 cm^{-1} signified the presence of carboxylic group [31]. The result is in agreement with moringa leaves extract FTIR analysis report [32]. The powders exhibited stronger resemblance to the spectra of sucrose because of the small amount of extract incorporated into sucrose. The similarity indicated that all moringa leaves extract powders still possessed advantageous physicochemical traits of sucrose such as solubility. However, powdered extracts showed very weak peaks at the range of 1600 cm^{-1} similar to the spectra of moringa leaves extract, representing C=C carboxylic group possibly originated by phenolic compounds.

3.4. Initial Antioxidant Activity. Antioxidant activity in form of DPPH radical scavenging activity in this study is expressed by IC_{50} value (half maximal inhibitory concentration) defined as concentration of sample needed to scavenge 50% of DPPH free radical. Less IC_{50} value suggests

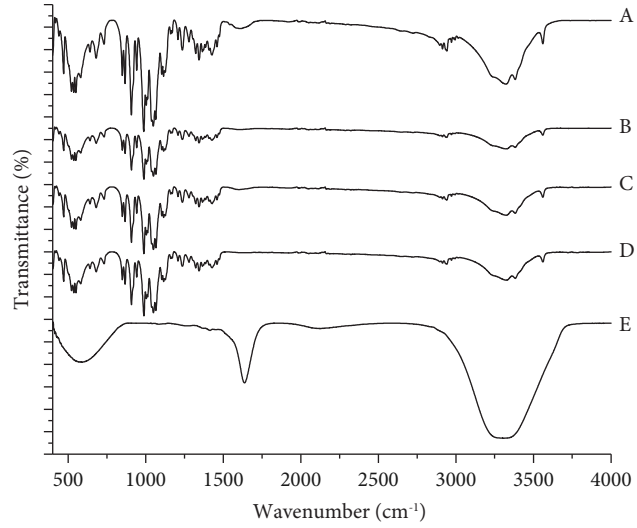

Figure 3: FTIR-ATR spectra of cocrystallized powders (*A*), vacuum-dried powders (*B*), plated powders (*C*), sucrose (*D*), and moringa leaves extract (*E*).

less amount of sample is needed to scavenge free radicals due to the presence of antioxidants [33]. The antioxidant activity of moringa leaves powders shown in Table 1 presents cocrystallization as a method, which resulted in lowest IC_{50} value and in turn strongest antioxidant activity. Porous agglomerates structures observed in the cocrystals may serve as a protection to the entrapped bioactive compounds [34]. The heat used in the process may degrade natural antioxidant compounds such as phenolics, but increased antioxidant activity can be achieved through degradative products and synergistic reactions between molecules caused by this high temperature [35, 36]. Heat treatment above 100°C can transform luteolin-7-O-glycoside to luteolin which possesses higher antioxidant activity than luteolin-7-O-glycoside

Table 1: The antioxidant activity of moringa leaves extract powders.

Samples	IC_{50} (ppm)
Cocrystallized powders	3647.96 ± 20.29
Vacuum-dried powders	4378.51 ± 26.29
Plated powders	4733.46 ± 31.91

whilst mesquitol can be degraded to a product with very high antioxidant activity [35, 37].

Another possible factor affecting the stronger antioxidant activity is the contribution of Maillard reaction products formed during heating. Maillard reaction in itself is a nonenzymatic browning reaction originated with the conjugation of amino and carbonyl groups during heating [38]. Several studies reported that Maillard reaction, primarily in intermediate and final stages, is able to generate antioxidant products with high DPPH scavenging activity [39, 40].

Weaker antioxidant activity exhibited by vacuum-dried powders might be caused by longer exposure time compared to cocrystallization. Long exposure time may cause more intense bioactive compound's impairment. Therefore, minimalization of drying time becomes an important step to avoid loss in the antioxidant activity [41, 42]. It is also possible that lower temperature of 70°C applied in vacuum drying may not be high enough to form antioxidant Maillard reaction products, which resulted in a lower antioxidant activity. This is supported by a study which states that formation of Maillard reaction products with antioxidant activity-increasing ability occurs at temperature above 100°C [43].

The plating treatment is observed to produce powders with the weakest antioxidant activity. The low antioxidant activity is caused by the impairment of antioxidant compounds due to additional light, oxygen, and heat exposures during the extract concentration step. The impairment could be more intense because the extract is not yet protected inside sucrose matrix. Plating also does not involve sugar heating so its antioxidant activity is not supported by antioxidant Maillard reaction products.

3.5. The Antioxidant Stability of Moringa Leaves Extract Powders. The antioxidant stability of moringa leaves extract powders is concluded from changes on DPPH radical scavenging activity during 30 days of storage and expressed through inhibition percentage in which higher inhibition percentage indicates a stronger antioxidant activity. Figure 4 shows that cocrystallized moringa leaves powders exhibit a stable antioxidant activity and maintain higher values than other two treatments throughout 30 days of storage. The tendency of stable and even increased antioxidant activity is also reported in *B. rubra* cocrystals [22] and yerba mate cocrystals [15]. The stability provided by this treatment may be caused by the entrapment of antioxidant compounds inside the porous crystal's structure, which limits the interaction of antioxidant compounds from external factors such as light, oxygen, and moisture during the storage [14].

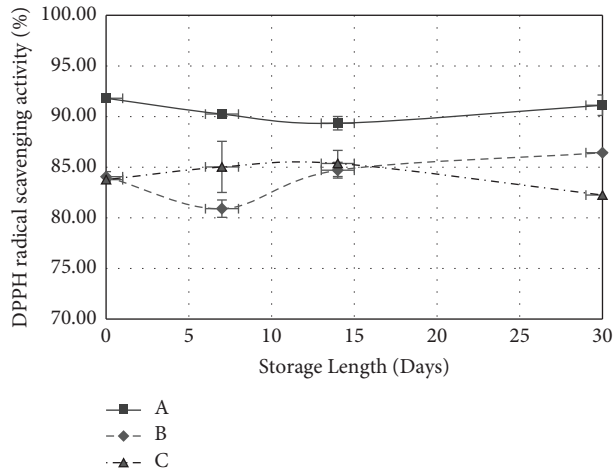

Figure 4: The antioxidant stability of moringa leaves extract powders during 30 days of storage.

The antioxidant activity of moringa leaves produced from vacuum drying declines on the seventh day but increases on the rest of storage duration as shown in Figure 4. This stability is promoted by the ability of vacuum-dried powders in protecting antioxidant compounds through agglomerated crystals. Lower antioxidant values compared to cocrystallization may be caused by less crystalline region and more amorphous region, as shown in the XRD analysis, since literature claims that the crystalline structure is able to protect entrapped active compounds better than the amorphous structure [16].

The antioxidant activity of plated powders inclines to decrease throughout the storage length. This behavior is possibly caused by the inability of carrier material to encapsulate and protect bioactive compounds from external oxidising factors during storage [44]. This is supported by the morphological result which shows no pores and only layers of the extract deposited on the plated powder's surface. However, the antioxidant activity decrease is not shown to be drastic and tends to be stable where in the final day of storage plated powders still exhibit a high antioxidant activity above 80%. This gives an interesting perspective towards plating method as extract powdering method for its stable antioxidant activity event despite of the modest and low-energy process.

Relatively fluctuative pattern obtained in this study is also observed in yerba mate cocrystals [15] and chokeberries cocrystals [17]. The longer storage time might enable the decomposition of antioxidant compounds through higher exposure of light and oxygen [45, 46] but increased antioxidant activity is also found in several studies [47, 48]. Several studies disclose that the formation of new phenolic compounds with a similar or higher antioxidant activity may occur during the storage due to the conjugation of phenolic compounds [49–51]. Structural changes in the phenolic compounds can also occur which results in increase of the antioxidant activity [17].

Table 2: Colour attributes of moringa leaves extract powders.

Samples	L^* (lightness)	a^* (redness)	b^* (yellowness)
Cocrystallized powders	79.94 ± 0.17	3.67 ± 0.09	27.38 ± 0.19
Vacuum-dried powders	86.84 ± 0.03	0.91 ± 0.01	22.15 ± 0.09
Plated powders	87.74 ± 0.54	0.57 ± 0.02	19.95 ± 0.60

3.6. Colour Attributes. Table 2 displays the colour attributes of moringa leaves extract powders consisted of L^* (lightness), a^* (redness), and b^* (yellowness). Cocrystallization shows lowest L^* value and highest a^* value. This is caused by the formation of brownish pigment which can be marked by the loss of lightness (lower L^* value) and shift towards reddish colour (+a^* value) [52]. Brown colour is a result of Maillard reaction. Higher temperature will promote more intense brown colour in Maillard reaction [53]. Aside from the increased reaction rate, denaturation of proteins occurs at a temperature above 100°C which makes more proteins available for Maillard reaction [54]. This can explain stronger brown colour in cocrystallized powders compared to vacuum-dried powders because the latter occurs at a lower temperature less than 100°C. As for plated powders, low L^* and a^* values indicate less brown pigment formation because the treatment occurs without sugar heating.

3.7. Total Phenolic and Flavonoid Contents. According to Table 3, moringa leaves extracted by the boiling method resulted in higher total phenolic and flavonoid contents compared to moringa leaves without boiling. This indicates that the boiling method used was able to extract phenolic compounds in the leaves' matrix. The high temperature applied on the water solvent increased the mobility of water molecules and decreased electrostatic interaction between the molecules which enables the dissolvement of less polar compounds contained in moringa leaves [55]. The powders had lower total phenolic and flavonoid contents than the moringa leaves extract which was expected due to the low concentration of moringa leaves extract incorporated into the sucrose. When comparing the powders, it was observed that the three processes were able to produce powders with similar phenolic and flavonoid contents. This indicates that despite of the high temperature used, cocrystallization resulted in powders with similar phenolic and flavonoid contents as vacuum drying and plating which operated at lower temperatures.

Distinct relations between total phenolic and flavonoid contents and antioxidant activity were also not found. This phenomenon is also observed by other studies [15, 35, 56]. Disproportionate results between these three parameters are likely caused by different qualitative profiles of phenolic and flavonoid compounds [57]. Other antioxidant nonphenolic compounds found in moringa leaves such as niazimicine, glucosinolates, isothiocyannates, vitamin A, vitamin C, riboflavin, and α-tocopherol [58] possibly influence the differing results.

Table 3: Total phenolic and flavonoid contents of moringa leaves extract, moringa leaves, and moringa leaves extract powders.

Samples	Total phenolic (mg GAE/g solid)	Total flavonoid (mg QE/g solid)
Moringa leaves	9.08 ± 0.59	5.62 ± 0.01
Moringa leaves boiled extract	21.32 ± 1.09	7.12 ± 0.05
Cocrystallized powders	1.34 ± 0.04	0.53 ± 0.05
Vacuum-dried powders	1.44 ± 0.04	0.48 ± 0.01
Plated powders	1.35 ± 0.05	0.54 ± 0.03

3.8. Physicochemical Properties. Physicochemical properties determined in this study are hygroscopicity rate and angle of repose. Hygroscopicity rate presented in Table 4 shows that cocrystallized powders exhibited the lowest hygroscopicity rate ($0.26 \times 10^{-4} \pm 0.02 \times 10^{-4}$ g H_2O/g solid/minute) at 75% RH. The low hygroscopicity observed in cocrystallized powders were also found in other studies such as β-karoten cocrystals [59], yerba mate tea cocrystals [15], and marjoram cocrystals [16].

Vacuum drying resulted in powders with hygroscopicity rate of $0.29 \times 10^{-4} \pm 0.05 \times 10^{-4}$ g H_2O/g solid/minute. This low hygroscopicity is in contrast with the high hygroscopicity found at vacuum-dried licorice extract powders [60] due to high crystalline structure of powders obtained in this study compared to the amorphous structure typically found in vacuum-dried products. Plating resulted in powders with highest hygroscopicity rate of $0.48 \times 10^{-4} \pm 0.06 \times 10^{-4}$ g H_2O/g solid/minute. It can be inferred from results obtained that the rate of hygroscopicity trend is inversely correlated with the degree of crystallinity of the powders. Literature states that the higher crystallinity results in slower moisture intake because strong intermolecular forces between crystal's molecules create less accessibility in the structure [23, 61, 62].

Besides hygroscopicity rate, angle of repose was also analysed and presented on Table 3. Cocrystallization showed the highest angle of repose (47.87 ± 0.19°), followed by vacuum-dried powders (37.36 ± 0.69°), and the plated powders resulted in the lowest angle of repose (27.77 ± 0.42°). The lower angle of repose represents better flowability resulting in easier application and transportation [63]. Cocrystallization resulted in powders with poor flowability, vacuum drying resulted in powders with moderate flowability, and plating resulted in powders with free flowability. Angle of repose results are proportionate with the moisture content because higher moisture content increases friction coefficient between particles due to increased adhesion between particle and the surface [64]. Thus,

Table 4: Physicochemical properties of moringa leaves extract powders.

Samples	Hygroscopicity rate (g H_2O/g solid/minute)	Moisture content (% g/g solid)	Angle of repose (°)
Cocrystallized powders	$0.26 \times 10^{-4} \pm 0.02 \times 10^{-4}$	3.30 ± 1.03	47.87 ± 0.19
Vacuum-dried powders	$0.29 \times 10^{-4} \pm 0.05 \times 10^{-4}$	1.55 ± 0.14	37.36 ± 0.69
Plated powders	$0.48 \times 10^{-4} \pm 0.06 \times 10^{-4}$	1.20 ± 0.15	27.77 ± 0.42

moisture content becomes a critical control point in managing flowability of a material and an increased value will decrease its flowability [65].

4. Conclusion

The study found that cocrystallization, vacuum drying, and plating exhibited a stable antioxidant activity of moringa leaves extract during the storage despite of their modest processes. Cocrystallization is applicable to provide the strongest and most stable antioxidant activity of moringa leaves extract due to the high crystallinity and pores formed on the structure. However, the powders showed poor flowability due to their moisture content. Further studies are therefore necessary to determine better method in producing cocrystals with less moisture content.

Data Availability

No data were used to support the findings of this study.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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