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Microencapsulation of Ethyl Acetate Extract from Green Coffee Beans (*Coffea Canephora*) by Spray Drying Method

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ABSTRACT

Coffee contains caffeine and chlorogenic acid (CGA) as the main constituents benefiting human health. Extract of green coffee beans (GCB) has some limitations in terms of its unpleasant flavour, aroma, and phytoconstituents bioactivity. This study aimed to encapsulate the crude ethyl acetate (EtOAc) extracts of Gayo Robusta GCB (Coffea canephora) to overcome its limitation. Microencapsulation was carried out by spray drying method using whey protein concentrate (WPC) as a coating material to produce nutraceutical supplement microparticles. The size of microparticles, morphology, and physicochemical characteristics were investigated. We found that the yield of microparticles was 39.5%, and the mean volume diameter was 1.367 µm (span 1.162). The morphology of the microparticles was irregular microspheres with dense, wrinkled, shriveled, compact, and relatively homogeneous structural shape. The physicochemical properties measurement indicated that the microparticles had a radical scavenging activity value (RSA) of 374.53 µg/mL, total phenol content (TPC) of 6.92 mg GAE/g, caffeine content of 9.12%, and CGA level of 7.19%. The spray drying microencapsulation using WPC was able to engulf and package the unpleasant flavour and aroma of the crude extract of Gayo Robusta GCB, producing an abundant yield, small and narrow range-sized particle, as well as protecting and carrying considerable amounts of phytoconstituents bioactivity.

Keywords: spray drying microencapsulation, green coffee bean, chlorogenic acid, caffeine, whey protein concentrate

INTRODUCTION

The word "coffee" was originated from the Turkish word "KAUVEH" and the Arabic word "QAHWAH". The "KAUVEH or QAHWAH" means "infusion or beverage or wine or some type of wine" which indicates an appetite suppressant (Garg, 2016). Coffee contains a complex chemical mixture of phytoconstituents (cellulose, soluble carbohydrates, insoluble polysaccharides, nonvolatile and volatile compounds, aliphatic acids, nitrogenous compounds, polyphenols, proteins, lipids, amino acids, vitamins, and minerals). Caffeine and CGA are important from nutraceutical supplements perspectives (Garg, 2016). The de Melo Pereira *et al*. (2020) have been reviewed relating metabolic and physiological effects of coffee consumption on human health, such as type 2 diabetes risk reduction, neurological diseases prevention (Parkinson's, Alzheimer's, cognitive impairment, and dementia), protective factors against depression and suicidal behaviour, anti cancer activity, hepatic injury and cirrhosis prevention, human gut microbiota establishment, reduction of risk factors for cardiovascular

diseases, and positive effects on the gastrointestinal tract (de Melo Pereira *et al*., 2020).

The GCB contains phytoconstituents (volatile compounds) that are responsible for the unpleasant flavour and aroma, causing nausea and vomiting upon inhaling, such as bit, sour, pungent, rancid, and repulsive (Garg, 2016). Therefore, before being served for consumption and other applications, the green coffee beans are processed through fermentation, soaking, drying, defatting, decaffeinating, and blending or roasting to reduce or eliminate unpleasant odour and taste. However, the processing of GCB causes changes in chemical and physical parameters (Cid and de Peña, 2016) which of course changes the concentration, content, and activity of phytoconstituents, as well as inducing changes in the structure of the constituents and formation of new compounds. Alternative methods that can be done are extracting, fractionating, or isolating the phytoconstituents, but the extracts and fractions can still have an unpleasant odour and taste from the solvents, while the isolates have to be derived in a complicated way. The processing, preserving, and packaging of GCB can be difficult, expensive, and not feasible (Belviso and Barbosa-Pereira, 2019).

A promising approach to overcome these problems is microencapsulation using whey protein concentrate (WPC) by the spray-drying method. Encapsulation is a process of engulfing or a way to package the materials of solids or droplets of liquids or gases in a compatible thin solid shell (Al Shannaq and Farid, 2015; Fang and Bhandari, 2012). The spray drying microencapsulation has served to render undesirable functionality of flavour and aroma, physicochemical desire change in sensory perception, and provide stability of the active ingredient (Vasisht, 2014). One of the microencapsulation techniques is spray drying that is defined as the liquid state transformation becomes the dry particulate form by spraying the liquid into a hot drying chamber. It is a process suitable for the conversion of various liquid formulations (aqueous and organic solutions, emulsions, and suspensions). This technique is a simple, fast, and scalable drying technology (Arpagaus *et al*., 2017).

For spray drying microencapsulation, there are several types of shell materials such as polysaccharide-based, lipid-based, protein-based, and polymer-based (Sobel *et al*., 2014). The selection of protein-based shell materials must consider safety requirements. The shell material must be food-grade, biodegradable, and capable of forming a barrier between the active agent and the medium (Henrique Rodrigues do Amaral *et al*., 2019). Whey is the liquid remaining from the precipitation and curd removal during cheese manufacturing (Ramos *et al*., 2016). The WPC is one of the polymeric shell materials which possesses healthy benefits as a glutathione enhancer and sports nutrition. WPC has been applied widely in pharmaceuticals, foods, and beverage ingredients as an agent in gelation technology, thermal stabilization, and emulsification. Meanwhile, the WPC is an excellent shell material and suitable carrier for its ability to package and deliver phytoconstituents (Ramos *et al*., 2016). Gelation properties of globular proteins allow for the development of micro-and nanostructures (Khaire and Gogate, 2019).

In recent years, several studies have been reported on spray-drying microencapsulation for some coffee extracts using other shell materials. (Abrahão *et al*., 2019; Ballesteros *et al*., 2017; Desai *et al*., 2019, 2020; Gilbert Stanley, 2020; Silva Faria *et al*., 2020). Also, previous studies reported using WPC for another extract type (Calva-Estrada *et al*., 2018; Lee *et al*., 2015; Oliveira *et al*., 2018; Premi and Sharma, 2017; Rocha *et al*., 2019; Sarabandi *et al*., 2018). A previous study reported that the hydroxycinnamic and CGA of GCB have an inclusion effect with WPC (Budryn *et al*., 2015). Spray drying microencapsulation of crude EtOAc extract of Gayo Robusta GCB using WPC has not been reported. The present study aimed to prepare, formulate, and produce microparticles for nutraceuticals supplements. Furthermore, yield product, particle size distribution, particle morphology, radical scavenging activity, total phenolic content, caffeine, and CGA content were investigated.

MATERIALS AND METHODS

Chemicals and plant material

Analytical grade n-hexane, ethyl acetate (EtOAc), and ethanol (EtOH) were purchased from Merck. Instant whey protein concentrate (IWPC 80) was purchased from Milkspecialties. Chromatography-grade caffeine and chlorogenic acid (CGA) standards were purchased from Sigma-Aldrich. The coffee bean of Coffea canephora was collected from Gayo highland Aceh, Indonesia.

Coffee beans preparation

The coffee bean was prepared according to Garg's (2016) procedure with major modifications. Coffee beans (5 kg) were pulped and dried in dry

air using a winnowing tray at 32 °C for 7 days. The parchment of the coffee beans was removed, and the bean was dried again for 7 days. The silver skins on the coffee beans were removed, and the beans were dried for 3 days to get dried green coffee beans (GCB). The dried GCB was ground in a blender and sifted with sieve number 40. The coarse GCB was collected in the vessel and stored in a dark and dry place before the extraction process.

Extractions procedure

The extraction procedure was performed by using maceration, according to a procedure by Khoddami *et al*. (2013) with some modifications. 200 g of coarse GCB was macerated with 1 L of n-hexane at room temperature for 24 hours and then filtered by a vacuum-assisted filter. The filtrate was collected, and then n-hexane residue was dried and remacerated with 1L of EtOAc at room temperature for 24h and then filtered. The EtOAc filtrate was collected and filtered. Taken together, two filtrates were collected from this maceration process, and all filtrates were evaporated by using a rotary evaporator (Buchi). The concentrated filtrates were collected in dark bottles, weighed, and stored in a refrigerator. The maceration process was done in triplicate.

Microencapsulation process

The microencapsulation was processed by using a spray dryer (GEA Niro) according to a procedure by Abrahão *et al*. (2019) with some modifications. The 100 mg of EtOAc extract was dispersed in WPC solution (100mg/100mL) to obtain 200 ppm of feed solution. The feed solution was homogenized by using an Ultra-Turrax (IKA T18) at 40°C, 1,000rpm for 45min, and stored in a refrigerator to complete hydrolysis until after night. The spray drying was performed at an inlet temperature of 180°C, outlet air temperature of 70°C, and feed flow rate of 5mL/min. The microparticles were collected in an airtight plastic bag, weighed, and stored at a desiccator for future analysis. The process was performed in triplicate.

Extracts and Microparticles yield

The extracts yield was calculated according to Fikry *et al*. (2019). Extracts yield (%) was expressed as a ratio of the weight of the extracted solids to the initial sample weight. The physical characteristics of the extract were determined visually and tactilely.

The microparticles yield was calculated according to the Gonçalves *et al*. (2017). Microparticles yield (%) was expressed as a ratio of the mass of microparticles obtained from the spray dryer to the mass of initial solid content in the feed solution. The physical characteristics of microparticles were determined visually and tactilely.

Particle size distribution and morphology

The particle size distribution (PSD) was analyzed by PSA (Beckman Coulter LS 13 320) according to a procedure by Kusmayadi *et al*. (2019). The sample was dripped on the test equipment. The PSD of the sample was presented as the mean volume diameter and the span was calculated using the following equation: span = $[(d_{90}-d_{10})/d_{50}]$.

Particle morphology was examined by using SEM (Leo 40 XVP) according to Kuck and Noreña's (2016) procedure. The SEM was completed with microanalysis systems of ray-X (Quantax EDS and Espirit Software). The sample was attaced to double-sided adhesive tape, nailed to a stub, and coated with gold. The SEM was operated at 15 kV from 1.0 to 7.0k magnification.

Radical scavenging activity assay

Radical scavenging activity was determined by 2,2-diphenyl-1-picrylhydrazyl (DPPH) method according to a procedure by Abrahão *et al*. (2019) using a spectrophotometer (Shimadzu UV-1601). Shortly, 0.07g of the sample were diluted in the distilled water (25mL) at 25°C and agitated by a vortex. The sample solution (2.5mL) was pipetted into the tube containing 1mL of DPPH solution (0.3 mM) reagent followed by stirring. After 30min, the absorbance (Abs) was recorded using a spectrophotometer at 518nm. The absorbance values were converted into radical scavenging activity (%RSA) using the following equation: %RSA=100-{[(Abs sample-Abs spiked blank)]x100/Abs control}. Ethanol (1.0mL) containing a sample solution (2.5 mL) was used as a spiked blank. DPPH solution (1.0 mL; 0.3 mM) in ethanol (2.5mL) was used as a negative control.

Total phenolic assay

Total phenolic content (TPC) was determined according to a procedure by Abrahão *et al*. (2019) using a spectrophotometer (Shimadzu UV-1601). Shortly, the samples (0.07g) were diluted in the distilled water (25 mL) at 25°C and agitated by a vortex. The sample solution (200µL)

was pipetted into the tube containing the reagent of Folin-Ciocalteu (200 µL). Then, 200µL of the saturated solution of the calcium carbonate (10% w/v) was added together with distilled water (2mL) and incubated in the dark for 60min at room temperature. The absorbance was measured using a spectrophotometer at 765nm. The TPC was calculated using linear regression obtained from the standard curve of gallic acid (0–80µg/mL). The TPC value was expressed in milligrams of gallic acid equivalent per gram (mg GAE/g) of dry matter.

Caffeine content

The caffeine content was determined according to procedures by Tine *et al*. (2017) and Vinson *et al*. (2019) using LC-MS/MS with some modifications. The LC instrument was a Flexar LC Perkin-Elmer with Flexar LC PE200 column oven (LUNA 3U C18 column). The MS/MS conditions were carried on an AB Sciex 3200 QTRAP fitted with an APCI ion source. The APCI was operated in the positive mode. 5mg of sample and caffeine standard were dissolved in acetonitrile/H_2O (1:1v/v) to obtain the final solution concentration of 50ppm and filtered using a polytetrafluoroethylene membrane (0.2µm). Solutions of caffeine standard were diluted with acetonitrile/H_2O (1:1 v/v) to obtain a calibration curve with 5 concentration points ranging from 2 to 10µg/mL. The caffeine (2µg/mL) standard solutions were directly injected in the MS/MS apparatus at the 10µL/min flow rate. The mass spectra (MIM-EPI) of caffeine were recorded of m/z 195 at 138 Da. The software for data acquisition and data analysis was Analyst 1.5.2. Each experiment was done in triplicate.

Chlorogenic acids content

The CGA content was determined according to procedures by Tine *et al*. (2017) and Vinson *et al*. (2019) using LC-MS/MS with some modifications. The LC instrument was a Flexar LC Perkin-Elmer with Flexar LC PE200 column oven (LUNA 3U C18 column). The MS/MS measurements were carried on an AB Sciex 3200 QTRAP fitted with an APCI ion source. The APCI was operated in the negative mode. 5 mg of sample and standard were dissolved in acetonitrile/H_2O (1:1 v/v) to obtain the final solution concentration of 50 ppm and filtered using the polytetrafluoroethylene membrane (0.2µm). Solutions of the CGA standard were diluted with acetonitrile/H_2O (1:1 v/v) to obtain a calibration curve with 5 concentration points ranging from 10to 50µg/mL. The CGA (20µg/mL) standard solutions were directly injected in the MS/MS apparatus at the 10µL/min flow rate. The mass spectra (MIM-EPI) of CGA were recorded of m/z 359 at 191 Da. The software for data acquisition and data analysis was Analyst 1.5.2. Each experiment was done in triplicate.

Sensory evaluation

The procedure was performed according to Varastegani *et al*., (2017) and Watts, (1989) with some modifications. Sensory evaluation was conducted with 5 panelists (researcher and colleagues). Panelists wrote down their perceptions concerning appearance (colour), aroma, taste, texture (mouthfeel), and overall acceptability of the samples. The samples were served to the panelists with designated codes (GCE: green coffee extract; MGC: microgreen coffee) by single-blinded evaluation. The hedonic scale of 9-point was performed with 1 score equals to extremely dislike and 9 score equals to extremely like (1 = dislike extremely; 2 = dislike very much; 3 = dislike moderately; 4 = dislike slightly; 5 = neither like nor dislike; 6 = like slightly; 7 = like moderately; 8 = like very much; 9 = like extremely). The results were analyzed using simple ANOVA analysis.

RESULTS AND DISCUSSION

Extracts and microparticles yield

The yield of crude EtOAc extract was 0.54%, and its characteristics (Table I). The yield of crude EtOAc extract was higher than that in Villanueva *et al*. (2011), who used ethyl lactate. Typically, EtOAc is used for decaffeinated or fractioned coffee beans (Pietsch, 2017), and it is the most effective solvent (Yashin *et al*., 2013).

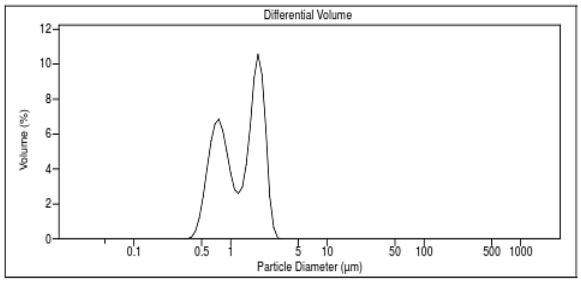
The yield of microparticles was 39.5%, and their characteristics (Table I). The yield of microparticles was lower than Desai *et al*. (2019), Sakawulan *et al*. (2018), and Nosari *et al*. (2015), but it was higher than Desai *et al*. (2020). The higher inlet temperature contributed to better production yield because the solvent was removed immediately by continuous flow, but it was inconsistent and independent (Arpagaus *et al*., 2017; Sakawulan *et al*., 2018). Variations in the yield might be caused by particle depositions around the spray cap and the chamber wall as well as cyclone efficiency (Jacobs, 2014), due to surface stickiness and cohesive particles (Gonçalves *et al*., 2017).

Table 1. Product yields and physical characteristics of crude EtOAc extract of GCB and the microparticles

Samples	Yields (%)	Physical Characteristics (visually and tactile)
Crude EtOAc extract	0.54 ±0.02	Semi-solid formed, yellowish-green color, smooth sandy texture, bitter flavor, coffee distinctive odor, and stung EtOAc aroma
Microparticles	39.5 ±0.89	Fine powder formed, smooth, greenish-yellow color, not bitter, savory flavor, roasted coffee, and milk distinctive odor, and fragrant aroma

Microparticles size distribution

The mean volume diameter and span of microparticles were 1.367µm and 1.162, respectively (Figure 1). Prior to spray drying, the extract showed a sandy and gritty texture (Table I) indicating relatively large particle sizes. A fine and homogeneous powder was obtained. In this study, the particles in the feed solution flocculated with an unadjusted pH value. It indicated that the particles were less uniform relatively, as well as a slightly wide size distribution. The mean volume diameter was lower than others (Abrahão *et al*., 2019; Desai *et al*., 2019). A study by Desai *et al*. (2020) has reported a particle size of 0.08 µm and span of 0.03 by using a nanospray dryer. The particle size is strongly influenced by the spray mesh size and solid concentration, and weakly influenced by the inlet temperature (Arpagaus *et al*., 2017).



Volume Statistics (Arithmetic)

Calculations from 0.017 µm to 2000 µm

Volume:	100%		
Mean:	1.367 µm	S.D.:	0.622 µm
Median:	1.358 µm	Variance:	0.386 µm²
Mean/Median ratio:	1.007	C.V.:	45.5%
Mode:	1.919 µm	Skewness:	0.219 Right skewed
		Kurtosis:	-1.267 Platykurtic

d_{10}: 0.617 µm d_{50}: 1.358 µm d_{90}: 2.196 µm

<10%	<25%	<50%	<75%	<90%
0.617 µm	0.768 µm	1.358 µm	1.907 µm	2.196 µm

Figure 1. The PSA analysis result of microparticle

Microparticles morphology

The images of microparticles showed irregular, spherical, dense, wrinkled, shriveled, homogenous, compact, not hollow, and porous particle morphology, indicated the particles were dispersed throughout the matrix (Figure 2). The particle morphology was similar to others (Abrahão *et al*., 2019; Desai *et al*., 2019, 2020). Arpagaus *et al*. (2017) reported the morphology of microparticles including being dense, hollow, porous, spherical, wrinkled, shriveled, or doughnut-like in shape. The particle morphology was dependent on the shell materials (Gonçalves *et al*., 2017), feed properties (material type, solid concentration, solvent, and surfactant), and drying temperatures (Arpagaus *et al*., 2017). The slow-drying produced more compact particles, while fast-drying led to hollow particles (Arpagaus *et al*., 2017).

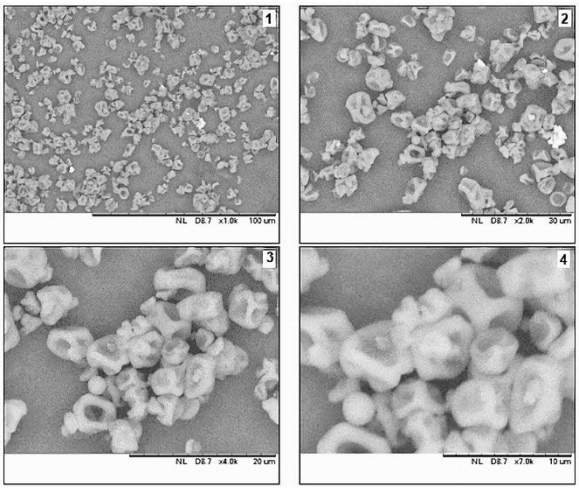


Figure 2. The SEM analysis results of microparticles at 1.0k (1); 2.0k (2); 4.0k (3); and 7.0k (4) magnifications

Radical scavenging activities

The DPPH IC_{50} values of crude EtOAc extract and microparticle were 101.97 and 374.53 µg/mL, respectively. DPPH IC_{50} value of the microparticles was higher than crude EtOAc extract (Table II), showing a decrease in RSA (2.67 times). The result was corroborated by others who reported the decrease of RSA related to the spray drying microencapsulation of coffee extracts using other shell materials. The retention of RSA was lower than Desai *et al*. (2020) and Sakawulan *et al*. (2018), but the RSA of microparticles was higher than those reported by them.

Table II. Parameter values of crude EtOAc extract of GCB and the microparticles

Parameters	Crude EtOAc extract	Microparticle
DPPH IC_{50} (μg/mL)	101.97±1.92	374.53±1.04
Total phenolic content (mg GAE/g)	2.913±0.28	6.926±0.767
CGA content (%)	8.67±0.08	7.19±0.06
Caffeine content (%)	11.57±0.07	9.20±0.04
Mean of particle diameter (μm)		1.367±0.622
Span		1.162

Table III. Sensory perception of crude EtOAc extract and the microparticles

Sensory evaluation	Crude EtOAc extract	Microparticle
Appearance (colour)	5.6±0.89	7.2±0.45
Aroma	4.6±0.55	7.8±0.45
Taste	1.6±0.55	7.6±0.55
Texture (mouthfeel)	1.8±0.45	7.6±0.55
Overall acceptability	3.4±1.52	8.4±0.55

Exposure to a high temperature decreased the RSA (Bastías-Montes *et al*., 2019; Desai *et al*., 2019; Frascareli *et al*., 2012) by triggering several reactions (phenolic degradations, polymerization, and Maillard reactions) which could create new compounds (Abrahão *et al*., 2019; Sakawulan *et al*., 2018).

Total phenolics content

The TPC of crude EtOAc extract and microparticles were 2.913 and 6.926 mg GAE/g, respectively (Table 2). TPC of microparticles was higher than that of crude EtOAc extract (1.37 times). The result was corroborated with Abrahão *et al*. (2019), who reported the increase of TPC using other shell materials, but the TPC increase was lower than those reported by them. On the contrary, other studies have reported a decrease in TPC (Ballesteros *et al*., 2017; Desai *et al*., 2019). The TPC of our microparticles was lower than Abrahão *et al*. (2019) and Desai *et al*. (2019), but it was higher than Ballesteros *et al*. (2017). The TPC increased due to the phenolics molecular structure alternation, physicochemical transformations of the shell material, phenolics conversion (Papoutsis *et al*., 2018), and newly formed equal phenolics (Abrahão *et al*., 2019).

Caffeine content

The caffeine contents of crude EtOAc extract and microparticles were 11.57 and 9.2 %, respectively (Table II). The caffeine content in the microparticles was lower than in crude EtOAc extract (0.25 times), and its encapsulation efficiency was 75%. The result was corroborated by others who reported the decrease of caffeine content related to the spray drying of different coffee extracts using other shell materials. The retention of caffeine content and encapsulation efficiency were higher than Desai *et al*. (2020) and Abrahão *et al*. (2019). Also, the caffeine content in our microparticles was higher than theirs. The caffeine was thermally resistant; thus the loss of caffeine might be due to the water-soluble properties of caffeine, leading to being partially carried over by the water vapour (Garg, 2016).

Chlorogenic acids content

The CGA contents in crude EtOAc extract and microparticles were 8.67 and 7.19%, respectively (Table II). CGA content in microparticles was lower than in crude EtOAc extract (0.2 times), and its encapsulation efficiency was 80%. The result was corroborated by others who reported the decrease of CGA content related to the spray drying of different coffee extracts using other shall materials. The retention of CGA content and encapsulation efficiency was lower than Desai *et al*. (2020); (2019), but it was higher than Abrahão *et al*. (2019). However, the CGA content of microparticles was higher than them. CGA is thermal unstable and thus the inlet temperature could trigger several reactions (isomerization, epimerization, lactonization) and degraded the compound into low molecular weight compounds (phenol and catechol), and to less extent lead to incorporation into melanoidin through covalent or

non-covalent bonds (Abrahão *et al*., 2019; Aguiar *et al*., 2016).

Sensory perception

Sensory evaluation was carried out to determine the level of preference for microparticles produced by spray-drying microencapsulation. The panelists tasted the samples and rated their overall delicacy using the 9-point hedonic scale. The sensory perception were evaluated by the panelists (Tabel 3). The simple ANOVA results depicted higher variations in the preferences for the crude extract, compared with the microparticles. The panelists' perception differed significantly ($P<0.01$) during sensory evaluation of the microparticles. Hence, the spray drying of crude EtOAc extracts using WPC brought about the variations in the panelists' expectation and actual physical and sensorial cues. On the other hand, the spray drying microencapsulation using WPC was able to engulf and package the unpleasant flavour and aroma of the crude EtOAc extract of Gayo Robusta GCB.

CONCLUSION

Spray drying microencapsulation produced microparticles with an abundant yield of 39.5%, a mean volume diameter of 1.367µm, and a span range of 1.162. The microparticles had a slightly wide size distribution and appeared as irregular microspheres with compact and smooth-structured morphology. The spray drying microencapsulation using whey protein concentrate succeeded to engulf and package the unpleasant flavour and aroma of crude EtOAc extracts of Gayo Robusta GCB. The spray drying encapsulated considerable amounts of phytoconstituents in the microparticles and retained their bioactivity. The radical scavenging activity of the microparticles was 374.53µg/mL, with the total phenolic, caffeine, and chlorogenic acid contents were 6.92g GAE/kg, 9.12%, and 7.19%, respectively. The microparticles could be potentially applied in the formulation of nutraceutical supplement for pharmaceutical or food and beverage ingredients.

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