Characterization of Nanoencapsulated *Centella asiatica* and *Zingiber officinale* Extract Using Combination of Malto Dextrin and Gum Arabic as Matrix

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Abstract. This research investigated nanoencapsulation of *Centella asiatica* and *Zingiber officinale* extract. The encapsulated extract was used as a complex matrix of multi-layered interfacial membranes between malto dextrin and gum Arabic. Characterization of nanoencapsulation using Transmission Electron Microscope (TEM), Fourier Transform Infrared Spectroscopy (FTIR) and BET surface area (SA) showed the morphology, functional group and cumulative adsorption in the surface area of pores. The TEM image of the nanoencapsulated powders of *Centella asiatica* and *Zingiber officinale* extract showed a nearly spherical shape with the particle size of 664 nm from its average radius.

1. Introduction

Nanoencapsulation allows protection of sensitive and bioactive pharmaceuticals component, as well as cosmetics and food ingredients from unfavorable environmental conditions, eradication of incompatibilities, solubilization or masking of unpleasant taste or odor. It can be produced as capsules in the range of submicrometer (from 10 to 1000 nm) in the colloidal systems [1-8]. The delivery system is defined as one in which a bioactive material is entrapped in a carrier to control the rate of bioactive release. Carriers can protect a bioactive component from unfavorable environmental conditions e.g. oxidation, pH and enzymes degradation [8-10]. Nanocarriers provide more surface area and have the potential to enhance solubility and improve bioavailability. Furthermore, it can ameliorate controlled release and targeting of the encapsulated food and cosmetics ingredients [11,12].

Centella asiatica can stimulate collagen biosynthesis and increase the incorporation of proline, thus modulating the metabolism of connective tissue. Asiaticoside, this pure triterpene from *Centella asiatica* is known to stimulate vascular and perivascular, with specific activity on collagen type I, whose reduction significant in aging and increased the tensile strength of newly formed skin [13-16]. This active agent is soluble in water and easy to defect at the high temperature. Ginger (*Zingiber officinale*) is known to have antioxidant properties which help to neutralize free radicals in human body. An antioxidant compound in a sufficient amount, substantially maintains the activity of collagenase and elastase in the skin [17]. This both active agent can be protected by encapsulation of malto dextrin and gum arabic with the proper ratio.

Gum arabic, a natural composite polysaccharide derived from exudates of A. Senegal and A. seyal trees, is one of the most common wall material used in encapsulation   due to its low viscosity, good emulsifying, high stabilizing and film-forming properties [18,19]. It has the ability of better retention of volatile substances and effective protection against oxidation[20,21]. Gum arabic has a highly branched structure which results in compact spheroidal conformation. Other matrix materials used was maltodextrin which has numerous properties that allow them to be used for diverse purposes in both the food and pharmaceutical industries. Their ability to retain water and form gels explains their choice as efficient food stabilizers in the microencapsulation of vitamins, minerals, colourants as well as fat and oils. However, the poor film-forming ability, hygroscopicity and turbidity of maltodextrins account for their inability to protect volatile compounds during spray drying [22-25]. Therefore, this work studied the nanoencapsulation of *Centella asiatica* extract and *Zingiber officinalle* extract by spray drying with malto dextrin and gum arabic as wall materials. The nanoencapsulation was evaluated for the particles size, BET surface and transmition electron microscope to observe the nanostructure of encapsulated powders. Meanwhile, FTIR finds out the chemical structure of the nanoencapsulation.

2. Material and method

2.1. Material
A sample of *Centella asiatica* (L.) was obtained from plantation area in Solo (Indonesia) with *Centella asiatica* extract using standardized Gotukula VDE by product number: 1614X623N. *Zingiber officinale* herbs extract procured from market herbs (Indonesia), furthermore *Zingiber officinale* extract using standardized Red Ginger VDE by product number: 740X226N with extract grade was nutraceutical. Gum arabic was purchased from Merck KGaA (Germany) with spray dried powder. Ethanol food grade purchased from PT. Brataco (Indonesia) with the purity of 70%. Maltodextrin was purchased from PT. SetiaGuna (Indonesia) and distilled water was obtained from a Milli-Q water purification system (Research center for chemistry, Indonesia). All of the materials mentioned above employed directly except the gum arabic and malto dextrin which was milled by a ball mill.

2.2. Method

2.2.1. Milling of raw materials. Malto dextrin and gum arabic were prepared by milling in a high-energy ball mill machine type Hem-E3D for 4 hours with the ball to powder ratio 10:1, speed of 1450 rpm and cycle time of 10 minutes in order to obtain more fine raw materials and the nano-sized scale.

2.2.2. Preparation of carrier agent solution. The carrier agent, i.e. gum arabic and malto dextrin which would be used as wall solutions were dissolved in distilled water and also were stirred with Thermo scientific cimarec for ±20 minutes until homogen. The solution of carrier agents was then mixed with the core materials i.e*Centella asiatica* and *Zingiber officinale* extract (30% solid contents). The carrier agent and active agent were in a weight ratio of 4: 1.

2.2.3. Preparation of active agent. The core materials (active agent) consisted of asiaticoside extract 2.5% w/w and gingerol extract 1% w/w of solid contents. Furthermore, active agent solution was prepared with ethanol and then stirred with Ultra-Turrax homogenizer Yellow line DI 25 digital for ± 15 minutes with a speed of 10,000 rpm until it became homogeneous.

2.2.4. Preparation of nanoencapsulation. Carrier agent solutions were combined with the active agent containing asiaticoside extract (2.5% w/w solid content) and gingerol extract (1% w/w solid content). Active agent slowly poured drop by drop into 50% of carrier agent solutions and stirred to reach homogeneity with Ultra-Turrax homogenizer Yellow line DI 25 digital for ±30 minutes with a speed

of 15,000 rpm. Then, the remaining carrier agent solution was slowly added to 30% solid contents and stirred with Ultra-Turrax homogenizer Yellow line DI 25 for ±20 minutes with a speed of 15,000 rpm until homogen. The last step was spray drying process where 1000-2000 ml of feed solutions were prepared.

2.2.5. Spray drying process. The feed solutions of nanoencapsulation were spray dried to get the product in powder form. The dried process operated at inlet air temperature of 100 – 180°C and outlet air temperature of 60 – 80°C. The temperature of feed solutions was 25°C. The principle of this technique was based on dissolving or dispersing the active agent in solution.

2.3. Characterization of nanoencapsulation

2.3.1. Particle size analyzer (PSA). 5 mg nanoencapsulation was dispersed in 3 ml distilled water. The particle size was determined by Beckman Coulter DelsaNano C (USA). The measurement of particles size distribution and a polydispersity index of samples was done at 25°C and sample was read three times.

2.3.2. Fourier transform infra-red (FT-IR) spectroscopy. FT-IR spectroscopy was a technique based on the vibration of the atoms of a molecule. An infrared spectrum was commonly obtained by passing infrared radiation through a sample and determination what fraction of the incident radiation was absorbed in a particular energy. Chemical structure of the obtained nanoencapsulation by FT-IR spectroscopy using IR Prestige-21 Shimadzu.

2.3.3. Transmission electron microscope (TEM). Transmission electron microscope (TEM) was applied to observe the morphology of nanoencapsulate product, carrier agent and solution of nanoencapsulation before spray drying process. A number of samples were placed on one surface of a double-faced adhesive tape stuck to the sample stage, then coated with copper under a vacuum condition to enhance their electric ability and observed on a JEOL (JEM-1400) electron microscope.

2.3.4. BET surface area (SA) analysis. Surface area affects for example dissolution rates of pharmaceuticals, the adsorption capacity of air, water purifiers, the activity of an industrial catalyst and the processing of most powders and porous materials. For determining the ability adsorption of nanoencapsule, it can be obtained with surface area measurment by TriStar II 3020 Version 2.00.

3. Resultsand discussion

3.1. Particle size analyzer
In the analysis, the particle size has a multi modal particle size distribution. Table 1 shows the results of particle size and polydispersity index. Raw materials of maltodextrin and gum arabic that were milled previously have a radius of 555.6 nm and 708.95 nm, respectively. Meanwhile, the polydispersity indexes are 0.411 and 0.511, respectively. The smallest particle was produced by nanoencapsulation of *Centella asiatica* and *Zingiber officinale* extract has the mean radius of 664.1 nm and has a low polydispersity index of 0.485. It indicates a fairly uniform size that represents homogenous particles. This uniform size also was influenced by the composition of the carrier agent as the wall in encapsulated material. The appropriate composition can obtain a better stability and accuracy release.

Tabel 1. Particle size and polydispersity index

Sample	Particle size in radius (nm)	Polydispersity index (PDI)
Maltodextrin	555.6	0.411
Gum arabic	708.95	0.511
Nanoencapsulation of of *Centella asiatica* and *Zingiber officinale* extract	664.1	0.485

3.2. Fourier transform infra-red (FTIR) spectroscopy

Figure 1 shows the FT-IR spectra of nanoencapsuled materials containing active agent (*Centella asiatica* and *Zingiber officinale* extract) and the carrier agent. The FT-IR spectra of *Zingiberofficinale* extract (Fig. 1a) shows the peak at a wave number of 1605 cm^{-1} which is identified as C=C vibration of in an aromatic ring. Meanwhile, the peaks at wave numbers of 2930 cm^{-1} and 3475 cm^{-1} are stretching vibration of C-H and –OH. The results are confirming that the gingerolwas contained in *Zingiber officinale* extract. The IR spectra of *Centella asiatica* extract (Fig. 1b) shows some principal peaks at wave numbers 1050 cm^{-1}, 1450 cm^{-1}, 1750 cm^{-1}, 2950 cm^{-1} and 3450 cm^{-1} refer to the stretching of C=C, C-O-C, C-C, C-H and –OH that confirming the purity of *Centella asiatica* extract was in the form of compounds asiaticoside. From the spectra, we can see that (Fig.1c-e) the intensity of peaks in 1637 cm^{-1} came from the aromatic ring (C=C) vibration sources from gingerols. Stretching vibration of –OH at peaks in 3437 cm^{-1} and stretching vibration of C=O at 1627 cm^{-1}, stretching vibration of –OH at wave number 3437 cm^{-1} from gum arabic and malto dextrin. Others peak in 2927 cm^{-1} was stretching vibration of C-H on the benzene ring from asiaticoside. It can be said that the encapsulated material contained the extract and also the matrix from the certain peaks that it showed.

3.3. Transmition electron microscope (TEM)

Morphological analysis of the nanoencapsule product, carrier agent and solution of nanoencapsulation before spray drying process were conducted using transmission electron microscope (TEM). Figure 2(a) shows some irregularly shaped particles and also some spherical shape.The morphology of the outer surface of the capsule (Fig. 2(b)) show the present of carrier agents i.e combination of gum arabic and malto dextrin and also the morphology of the encapsulated-material. The morphology of carrier agent without an active agent was clearly observed as a spherical shape, respectively.The nanoencapsule product was successfully formed with an active agent from *Centella asiatica* extract and *Zingiber officinale* extract as it is shown in Figure 2(c) that describes spherical shapes with a smooth surface. The TEM images do not show any pores or crack in the nanoencapsulated-form of *Centella asiatica* and *Zingiber officinale* extract with the carrier agent as wall material. The figure also showed the morphology of outer surface of the nanoencapsule which resulted from the interaction of inhomogeous reaction kinetics, fluid-induced shear force and shell-determined elastic forces [26]. However, the size of nanoencapsulated material was slightly bigger than the carrier agent itself. This is due to the ability of encapsulated active materials to expanse the size of nanoparticles [27].
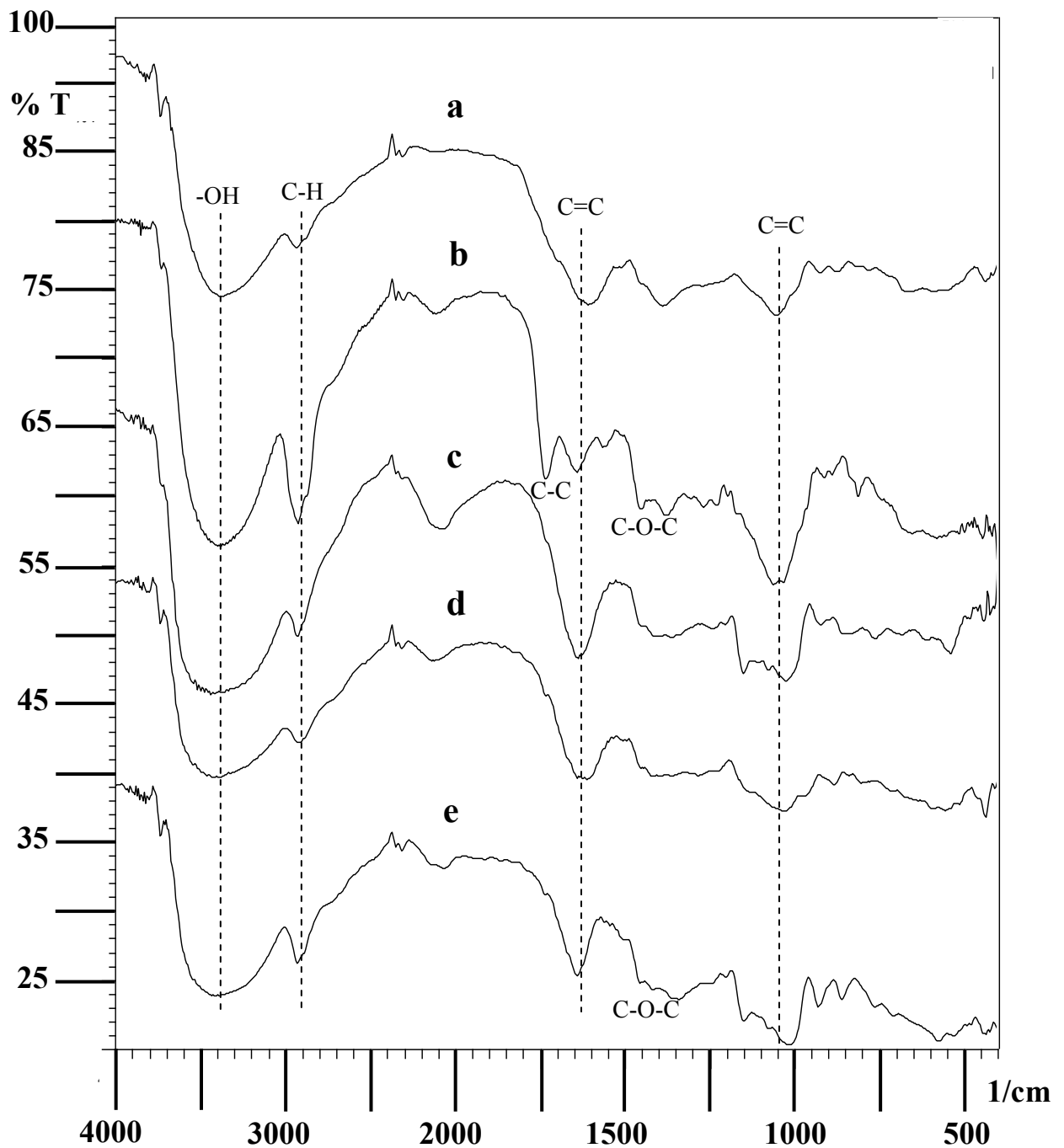
Figure 1. FT-IR spectrum: (a) *Zingiber officinale* extract; (b) *Centella asiatica* extract; (c) Gum arabic; (d) Dextrin and (e) nanoencapsulation of *Centella asiatica* extract and *Zingiber officinale* extract
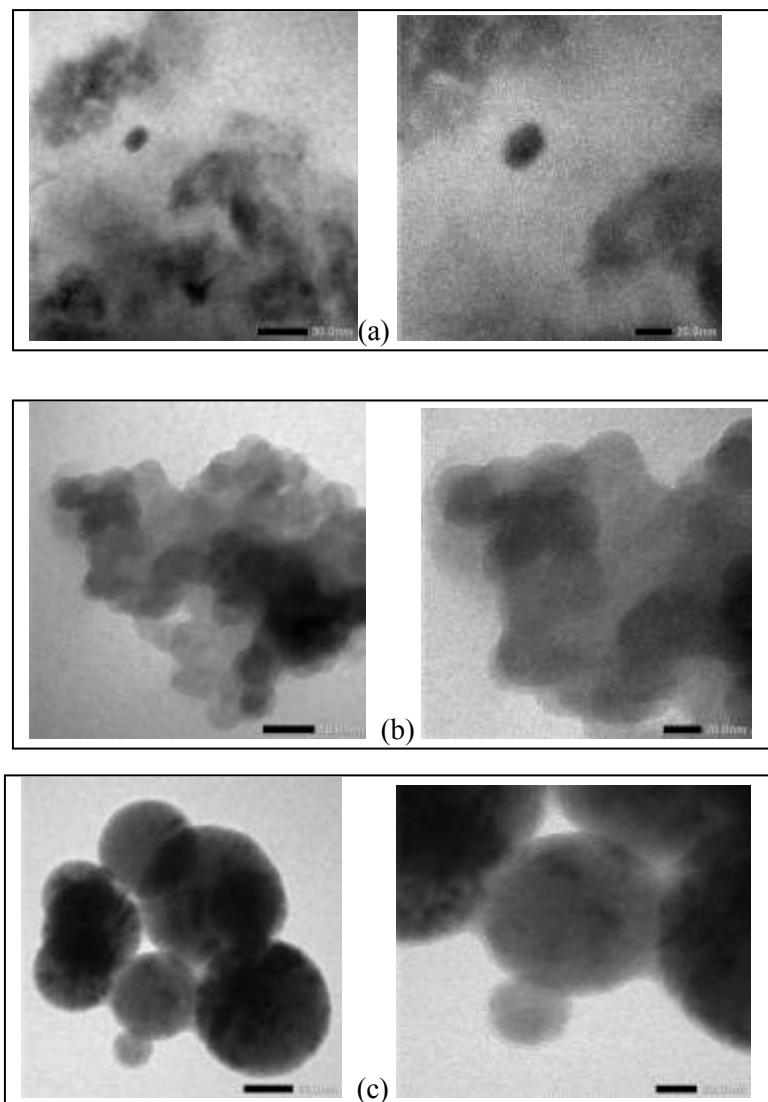
Figure 2. Transmission electron microscope (TEM) image: (a) solution of nanoencapsulation before spray drying process, (b) The wall (carrier agent), (c) nanoencapsule powder of extract *Centella asiatica* and *Zingiber officinale*

3.4. BET Surface area (SA) of nanoencapsule

A surface area analysis of nanoencapsulated-powder was conducted to describe the changes in multilayer adsoption of inert molecules onto a surface of nanoencapsule. The results obtained a single point surface area of nanoencapsule that was 0.3876 m^2/g, cumulative adsorption volume of pores was 1.6×10^{-4} cm^3/g. The analysis assumes that adsorption occurs by multilayer formation and that the number of adsorbed layers was infinite [28].

4. Conclusion

Nanaoencapsule of *Centella asiatica* and *Zingiber officinale* extract using malto dextrin and gum arabic as a matrix has an average radius of 664.1 nm. Morphology of this product has a spherical

shape and smooth surface with the surface area at 0.3876 m^2/g. This work would promote the application of this nanoencapsulation system product for cosmetic and pharmaceutical industries.

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