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Characterisation of Encapsulation Citronella Oil with Gelatin-Chitosan

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ABSTRACT. Citronella oil is one of the essential oils has been known recently due to its applicability in food and pharmaceutical industry. The main problems of essential oil are unstable and fragile volatile component. They could be degraded easily by oxidation, volatilization, heating and light if they are not protected from external factors such as encapsulation. Microencpasulation is a method whereby one material or mixture of the materials is coated by other material.This method is designed for protection, isolation and assist in storage. In the present study, the gelatine-chitosan microcapsules were prepared by complex coacervation.The marphology of the microcapsules, the mean particle size of microcapsules, thermal stability and release rate and release mechanisme of the microcapsules were investigated. On the optical microscope , the microcapsules were spherical shape and irregular size. The release of citronella oil from the capsules can be seen clearly with the rupture of spherical shape within two weeks. The mean particle size by the volume for this microcapsules is 363.176 µm which at acceptable range of particles size by the complex coacervation which is at range 1-500 µm. The span values more than two indicated that different width of size distribution shows not much difference and can be accepted. Analysis of result of thermal analysis reveal that incorporation of citronella oil by the microencapsulation process results in a complex coacervation with high thermal stability compared with the free oil, indicating that microcapsules protect the oils, making it more resistant to evaporates.

Keywords: *Microcapsules, Citronella oil, Complex coacervation, Gelatin, Chitosan;*

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1. INTRODUCTION

Many active compounds present in pharmaceutical, cosmetics and food products (essential oils and flavor) are instable compounds. They can suffer oxidation or volatilization or react with other formulation components. Microencapsulation is a feasible alternative to increase the stability of these compounds. There is a few works in literature on the encapsulation citronella oil .Some aspects of the microencapsulation process still need to be better understood as well as the influence of the process on the oil characteristic [1].The most important aspect in microencapsulation of given essential oil is to prevent the deterioration of the oil during the encapsulation step [2, 3]. The choice of microencapsulation methods is very much depend on the nature of core material [4]. Among the microencapsulation technique, the complex coacervation is suitable for encapsulate the high value active molecules and unstable substances such as polyphenols which are the major constituent in citronella oil [5]. Complex coacervation involved neutralization of two oppositely charge polymer in aqueous solution. The concentration of the wall material and pH of coacervation are the most important characters in the microencapsulation by coacervation. Furthermore, an additive which is cross linking agent such as formaldehyde or glutaraldehyde normally use as to harden the wall around the

core material [4]. Basically, the complex coacervation consist of three steps: formation an oil-in-water emulsion, formation of the coating and stabilization of the coating[6]. From the literature, the most favorable coating were the gelatin and Arabic gum [7,8,2]. Recently the microencapsulation with gelatin and chitosan also been considered by the researcher because the properties of gelatin and chitosan that contain thermal stability as when combine together to form the wall [3].

2. MATERIALS AND METHODS

2.1 Preparation of encapsulation citronella oil by complex coacervation

Materials. The materials used as wall materials in this research were Gelatine-B (type B, 260 bloom, from bovine) was supplied from Halagel Sdn. Bhd and Chitosan (R&M Chemical). Essential oil used as a core material was citronella essential oil (CO) which was extracted from lemongrass. Glutaraldehyde (50% aqueous solution) was used as a cross linker. Other reagents involved were distilled water, ethanol, acetic acid solution (1%v/v), hydrochloric acid (HCl) and sodium hydroxide (NaOH).

2.2 Encapsulation procedure. The encapsulated citronella oil (ECO) was made through the complex coacervation process. In this process the substances that were used are citronella oil, gelatin type B and chitosan.

2.3 Preparation of encapsulation solution. Before starting encapsulation process, preparation for two main solution A and solution B must been done first. Solution A were prepared by 30 min soaking 3.5 g of gelatin-B in 350 ml of deionized water prior one hour stirring at 50^0C until the solution were fully dissolved. Solution B was prepared by dissolving 0.1 g of chitosan in 100 ml of acetic acid aqueous solution for more 12 hours at the room temperature.

2.4 Microencapsulation CO by complex coacervation using chitosan-gelatin (B). There were seven steps involved in the complex coacervation including emulsification, coacervation, pH adjusting, dilution, cooling, cross linker and harvesting. After harvesting process, the processed mixtures was transferred into a separating funnel and undergo settling process for 24 hours. Three layers occurred whereby the top layer contains capsule in oil layer, middle layer contains water and diluted excess polymer and lastly bottom layer contains concentrated excess polymer. The bottom layer and middle layer were drained off and the top layer was rinsed with distilled water three times

2.5 Assessment of microcapsule morphology. The shape and morphology of ECO can be determined by using optical microscope (RZ-5, Meiji Techno, Japan).

2.6 Particle size and particle size distribution. Particle size and particle size distribution were determine by a laser particle size analyzer (Mastersizer 2000, Malvern Instrument Ltd., UK) at 1 min, 2500 rpm.

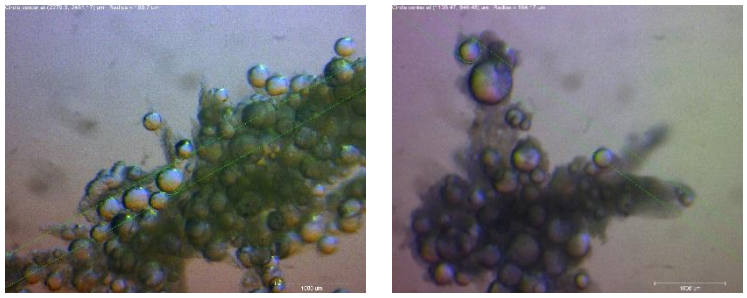
2.7 Thermal stability. The thermal stability of the microcapsules was studied by thermogravimetric analysis (TGA, SDTA851, and Mettler Toledo. The thermal behavior of gelatin, chitosan, citronella oil and microcapsules were analyzed by using heating rate 10 ˚C min^{-1}, temperature range between 25 -550 ˚C.

3. RESULTS AND DISCUSSION

3.1 Morphology of encapsulated citronella oil. The encapsulation of citronella oil by complex coacevation and using gelatin - chitosan as core shield was successfully. Fig. 1 shows the microcapsules of citronella oils under optical microscope.

Fig. 2 shows that the CO capsules are in spherical shape which having irregular size. Some of the capsules were attached together causing agglomerate due to the hardened free gelatin –B after cooling until the settling process. Agglomeration of the capsules during the wall formation was a common phenomenon in many microencapsulation process [10]. As the wall of the materials change from liquid to solid form, they often went through a sticky stage which makes the agglomeration difficult to avoid [10]. After the microcapsules were dispersed under the water, the spherical microcapsules can be seen clearly. Also the

color of citronella oil from the picture shows the yellow color indicated the origin color of citronella oil. It can be observed clearly in Fig. 2.



(A) (B)

Fig. 1 The microscopic image of the microcapsules under optical microscope : (A) Microcapsules forms agglomerates and (B) Microcapsules dispersed under water

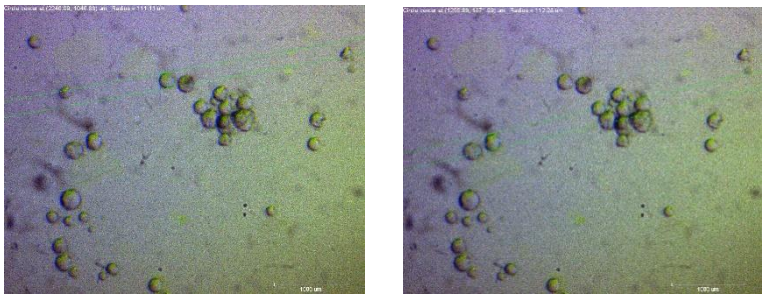


Fig. 2 The picture of the microcapsules dispersed in water

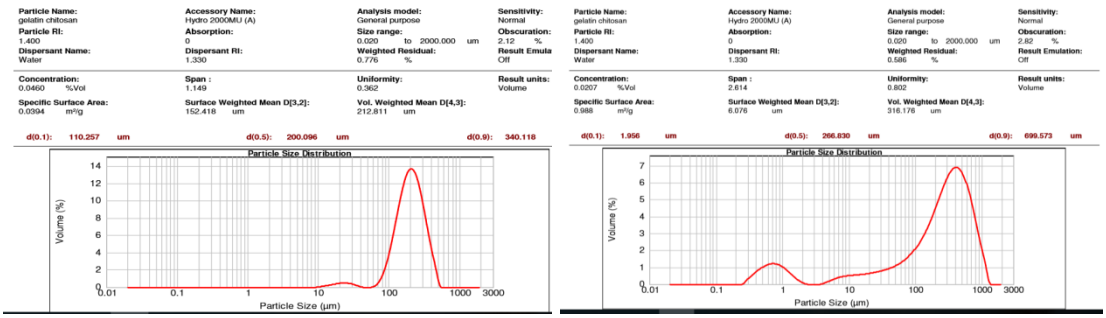
3.2 Particle size and Particle Size distribution. It was observed from Fig. 3 that this distribution is a unimodal distribution .Unimodal distribution is a distribution with one clear peak or most frequent peak. The values increase at first then, then rising to single peak then after that they decrease [11]. At the Malvern laser diffraction, the particle size was expressed as mean diameter over the volume distribution d (4,3) and size distribution (span) were calculated by equation 1.

$$\text{Span} = \left(\frac{(D(0.9)_- (D(0.1)}{D(0.5)}\right) \quad (1)$$

whereby D(0.9), D(0.5) and D(0.1) are the particle diameters at 10%, 50% and 90% of undersized particle calibration curve, respectively. In this experiment, the mean particle over volume is 363.176 µm. It twice from the reported by other researcher [12] but still considered moderate rate of microencapsulation that have been reported by other researcher whereby the range of microencapsulation by using complex coacervation is from 1 to 500 um [9].

Fig. 3 also shows that same particle size distribution measurement was quantified both relative to the total of particles and the volume of particles. It was observed 10% of total volume particles have diameter less than 2 um, 50% total of volume have diameter below 267 um meanwhile 90% of total volume particles has diameter less than 700 um. It represent 5.52% of the particles by volume. This indicated that microcapsules

contain the particle size acceptable range from 1 um to 500 um. The span values for this experiment is 2.6 and that means the different width of size distribution is not far and can be acceptable. The more values of span, the wider of distribution. Fig. 4 shows the mean particle size, particles size distribution and span.



(A) (B)

Fig. 3 Mean particle size particle distribution and span from two experiments A and B

Table 1 show microcapsules mean particles size obtained for two replicants of experiment. Although the obtained distribution has wide dispersions, the results pointed out for a good reproducibility.

Table 1 Mean volume particles for two experiments.

Experiments	Particle Size (um)
1	363.2
2	212.8
Average	288

3.3 Thermal stability of microcapsules. Thermograms become an important part for analysis of microcapsules formation. It is also provides information on each constituent of microcapsules which can used for their comparison. Fig. 4 shows the TGA curves and their first derivatives as function of time. Table 2 summarizes the most important thermographic results.

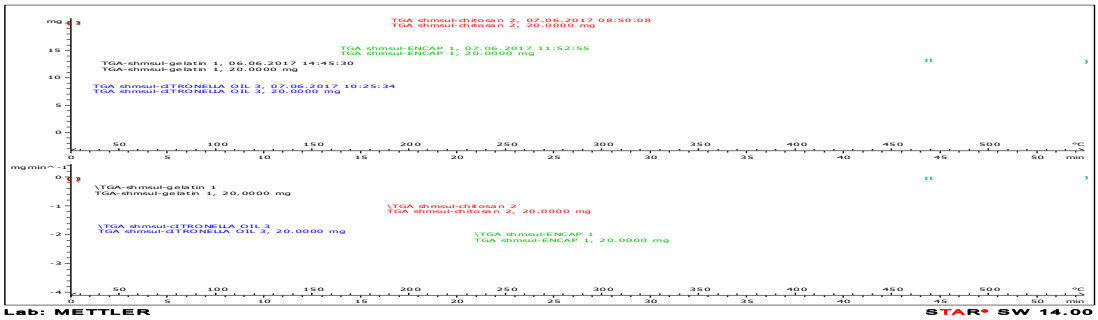


Fig. 4 The TGA curves and their first derivatives as function of time for gelatin, chitosan, citronella oil and encapsulation of citronella oil.

The thermogravimetric curves in Fig. 4 and Table 2 disclosed the distinct mass reduction or losses for each component and indicated the thermal decomposition in different stages. Citronella oil present mass reduction in single stage, while chitosan, gelatin and encapsulation of citronella oil present two stages [13].

Mass losses of citronella oil started at about 44 ºC and it end at 268 °C. Analysis of thermogram curve and its first derivative shows oil evaporated completely. Thus, this oil is highly volatile and its need to extend the

durability when will be used for other applications. It is slightly higher from reported by previous study [13]. Analysis of thermogram curve and its first derivative shows that the oil completely evaporated. This oils is highly volatile and requires protection to extend its durability when applied to surface. This act can be understood more by looking to the thermal behavior of jasmine oil [14]. It's indicating that the most of the mass of the jasmine oil was lost below 150 ºC and not tolerant with thermal durability.

For gelatin and chitosan, the first mass loss stage indicates evaporation of all the residual water present in these compounds [15,16]. The liberation of humidity seen in TGA and DTG curves in Fig. 4 occurs in initial temperature range of analysis, approximately above the boiling temperature. For this case, the mass loss of gelatin extend to 219 °C and 109 ºC for chitosan. Quantitatively mass loss for gelatin is 3% and chitosan is 7% from 20 mg samples.

Pyrolysis of gelatin and chitosan occurs in the same decomposition temperature range 240 °C to 450 ºC. These two compound present different behavior in decomposition zone. For mass loss (44% and 34%, respectively) and the residual mass is 55% and 65%, respectively.

In the thermogram for the microcapsules, two thermal events are also observed: the first stage of mass loss at the 35 ºC boiling and below 225 ºC [6]. In this stage 72% mass loss occurred. The second stage of mass loss occurred in the upper temperature range, beginning at 225 ºC and extended to 410 ºC. At this stage, likely most of the most of the protein chain such as gelatin and chitosan have been degraded. At this stage 24% of mass loss occurred. Active core material begin to degrade. Fig. 4 also shows that in the second stage of curve for this microcapsules occurs in a few seconds due to the high volatility of citronella oil. It can also been observed in the corresponding first derivative curve.

Table 2 Themogravimetric data for citronella oil, gelatin, chitosan and microcapsules

	Citronella	Gelatin	Chitosan	Microcapsule
Stage 1				
$\Delta T_{decomposition}$	44 -265^{0}C	30 -93^{0}C	21 – 109 ºC	32 – 233 ºC
T_{Max}	265^{0}C	93^{0}C	109^{0}C	233 ºC
Mass Loss	84%	3.3%	7%	72%
Stage 2				
$\Delta T_{decomposition}$		233 - 503 ºC	244 - 480 ºC	280 - 408 ºC
T_{Max}		347 ºC	340 ºC	408 ºC
Mass Loss		44%	34%	2%
Residual	16%	52.7%	59%	26%

4. SUMMARY

Citronella oil was successfully encapsulated by complex coacervation. The morphology shows that microcapsules were obtained with almost perfectly spherical shape. The particle size for these microcapsules is at an average of 288 µm and they are at acceptable range. Thermal analysis revealed that microencapsulation of oil improved its characteristic, making it less volatile and suitable for applied in higher temperature.

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