Received: August 8, 2023 | Revised: September 7, 2023 | Accepted: September 10, 2023

Microencapsulation of moringa oil in bio-polymer by simple solvent evaporation technique

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ABSTRACT

Moringa oil (MO) contains various bioactive components and pharmacology. It is attractive to use as a raw ingredient in various products. However, there are limitations on its direct utilization, especially MO's instability and hastening the active ingredient's degradation from external environmental factors, including temperature, humidity, oxidation, light, and heat. To solve these problems, in this work, microencapsulation of MO using different biopolymers as cellulose acetate butyrate (CAB), ethyl cellulose (EC), and poly(L-lactic acid) (PLLA) were carried out by a simple solvent evaporation technique. The prepared polymer microcapsule suspensions were highly colloidal stable for all types of biopolymers and ratios. The spherical biopolymer capsules were formed to a micrometer size after solvent evaporation under all conditions. However, when the microcapsules were dried, aggregation was found with the polymer microcapsules at a ratio of PLLA to MO of 50:50 for all three types of polymers, possibly due to the low amount of polymer to completely encapsulate all of MO. When polymer contents increased to 70%, the dried spherical polymer microcapsules were smoothly produced. Using 70% polymers, the PLLA microcapsule surface was smoother than the polymer microcapsules prepared by CAB and EC which exhibited the dent or hole on the outer surface. Micrometer size, spherical polymer capsules with a core-shell morphology were fabricated. Due to the higher hydrophilicity of the polymer than the MO, the polymer moves outward, forming a strong shell around the MO. Then, all three biopolymers can be used for the microencapsulation of MO at a suitable polymer to MO ratio. However, using PLLA at a ratio of PLLA to MO of 70:30 presented the highest encapsulation efficiency (74.08%), which may be due to its high molecular weight. Because of the non-toxicity and biodegradability of biopolymers, the fabricated microcapsules would be well applied in cosmetic products.

Keywords: Moringa oil, Microencapsulation, Solvent evaporation technique, Poly-L-lactic acid

INTRODUCTION

Moringa oil (MO) is extracted from parts of the Moringa tree (Drumstick tree), such as leaves and seeds. It is currently widely used as a raw material in the health and beauty industries [1]. MO is a natural extract that is 100% natural, providing a chemical-free and non-toxicity. Because of its high antioxidant activity and other beneficial biological and pharmacological properties, natural MO is increasingly used as an anti-inflammatory, anti-bacterial, anti-fungal, etc. [2-4]. Although MO can be used directly, several restrictions remain such as the accelerated degradation of bioactive components when exposed to environmental variables, including heat, light, oxygen depletion, and moisture or the reaction with other ingredients in the product. This decreases the product's efficiency and lifespan. To solve these problems, encapsulation technology is therefore applied for encapsulating active substances.

Encapsulation has gained much attention and is widely applied to various products. If a highly efficient encapsulation technology is used to store MO, the quality of MO will be improved. This active ingredient is more stable after being encapsulated because it is protected from degradation or reaction with the external environment and other substances in the emulsion of the cosmetic product [5, 6]. The encapsulation increases the response's surface area and controls the release of bioactive components at the desired rate as well. Physical encapsulation

DOI: 10.60101/jarst.2023.253714

techniques are typically simple, repeatable, and scalable for the industry. Among encapsulation techniques, solvent evaporation is one of the most simple and efficient methods to produce polymer capsules encapsulating various substances [7-9]. It is a repeatable, low-cost, and easily scalable method for the industry at room temperature. The microencapsulation of jasmine oil with polymethyl methacrylate was successfully prepared by solvent evaporation in an oil-in-water emulsion (O/W) system. The amount of encapsulated jasmine oil (23.04%) and the encapsulation efficiency was 72% [7]. Therefore, the possibility of using this technique in the production of microcapsules is interesting.

Biopolymers, produced from renewable resources, are used in pharmaceutical and cosmetic products because of their non-toxicity. Various groups of them were used especially polyesters, including poly(L-lactic acid) (PLLA), and cellulose derivatives. PLLA and cellulose derivatives e. g., ethyl cellulose (EC) and cellulose acetate butyrate (CAB) are well-known biopolymers used for the preparation of polymer capsules [10-13]. Because they are natural polymers that are readily available, inexpensive, non-toxic, biologically friendly, and can be biodegradable [14-17]. Spherical CAB microcapsules containing diltiazem resin were prepared by solvent evaporation. The microcapsule size increased with the concentration of CAB [18]. EC microcapsules were produced for the encapsulation of probiotics to extend their viability [13]. In addition, the EC microcapsules were successfully prepared and applied to the fabric samples. The textile surfaces desired antibacterial activities have been achieved to an acceptable level [19]. As well as a well-known biopolymer, PLLA capsules containing linalool were non-toxic to human cells and prevented the growth of *Staphylococcus aureus*, *Escherichia coli*, and *Candida albicans* [20]. The spherical and smooth surface PLLA microcapsules containing phase change material, Rubitherm 27 (RT27), with a high encapsulation efficiency (96%) were fabricated [21]. Polymer capsules encapsulating vitamin E were successfully created utilizing PLLA by the conventional solvent evaporation method. Meanwhile, the particle size distribution of PLLA capsules encapsulating vitamin E using phase inversion emulsification is narrower than that of capsules produced via the conventional solvent evaporation method [9].

Therefore, in this work, the preparation of polymer microcapsules encapsulating MO by a simple solvent evaporation method is studied. Various types of biopolymers e. g. PLLA, EC, and CAB are used. The influences of polymer types and polymer: MO ratios on the microcapsule morphology and encapsulation efficiency of MO are determined.

MATERIALS AND METHODS

Moringa oil (MO; Organic Thailand), cellulose acetate butyrate (CAB; laboratory reagent, Sigma-Aldrich; Mn 77,000), ethyl cellulose (EC; laboratory reagent, Sigma-Aldrich; Mn 51,000), and poly(L-lactic acid) (PLLA; commercial, B. C. Polymer Marketing; Mn 95,000) were used as biopolymers as received. Polyvinyl alcohol (PVA; analytical reagent, Sigma-Aldrich) was used as a surfactant. Ethyl acetate (EA; 99.9% purity, RCI Labscan), dichloromethane (DCM; 99.9% purity, RCI Labscan), and tetrahydrofuran (THF; analytical reagent, RCI Labscan) were used as solvents as received. Deionized water was used throughout the study.

Solubility test

Biopolymers of CAB, EC, and PLLA were used as microcapsule shells. Before the microcapsule preparation, polymers and MO must be completely dissolved in appropriate solvent. Then, solubility testing was studied at a 1:1 ratio of polymers to MO with various polymer:solvent ratios under the conditions shown in Tables 1 and 2. CAB and EC could be dissolved in EA and PLLA in DCM.

Table 1 Reagent amounts for the solubility testing of CAB and PLLA.

Chemicals		Polymer: MO: Solvent		
		1: 1: 5	1: 1: 6	1: 1: 7
MO	g	1.00	1.00	1.00
Polymer[a]	g	1.00	1.00	1.00
Solvent[b]	g	5.00	6.00	7.00

[a]; CAB and PLLA
[b]; CAB using EA and PLLA using DCM

Table 2 Reagent amounts for the solubility testing of EC.

Chemicals		EC: MO: EA		
		1: 1: 9	1: 1: 10	1: 1: 11
MO	g	1.00	1.00	1.00
EC	g	1.00	1.00	1.00
EA	g	9.00	10.00	11.00

Preparation of polymer microcapsule encapsulated MO

PLLA was homogeneously dissolved in DCM and CAB, or EC was homogeneously dissolved in EA before mixed with MO as an oil phase. It was then poured into PVA aqueous solution before homogenizing at 5,000 rpm for 5 min to generate oil droplet suspension. The suspension was gentle stirred for solvent evaporation to produce polymer microcapsules, as shown in Fig. 1. The effects of the type of polymer and the ratio of polymer:MO were investigated under the conditions listed in Table 3.

DOI: 10.60101/jarst.2023.253714

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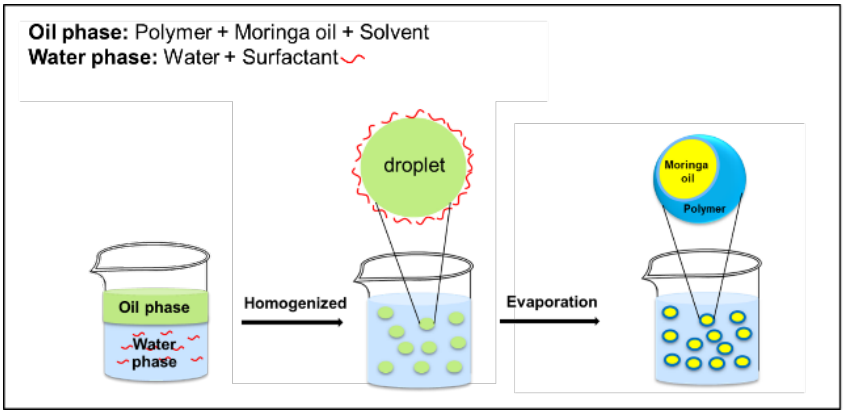

Figure 1 Schematic diagram for the preparation of polymer microcapsules by solvent evaporation.

Table 3 Reagent amounts for the preparation of polymer microcapsules by solvent evaporation at various polymer:MO ratios.

Chemicals			Polymer: MO	
			50: 50	70: 30
Oil	Polymer [a]	g	1.25	1.75
	MO	g	1.25	0.75
Water	PVA	g	0.23	0.23
	Water	g	22.5	22.5

[a]; PLLA, CAB and EC

Characterization

The suspensions of oil droplets and microcapsules encapsulated MO after solvent evaporation were observed with an optical microscope (OM, SK-100EB & SK-100 ET, Seek Inter) to investigate their shape and inner structure. A scanning electron microscope (SEM, JSM 6510, JEOL) was used to study the surface morphology of the microcapsules by distribution of the dried polymer microcapsules on a nickel SEM stub and coating it with Au. The amount of the encapsulated MO was measured by a UV-visible spectrophotometer (Lambda 35, Perkin Elmer). About weighed 0.1 g polymer microcapsules and MO standards [22, 23] were dissolved in tetrahydrofuran and adjusted the volume of the solution to 25 ml. Then, the absorbances of the standard solution at various concentrations and the sample solution were measured at λ_{max} 293 nm. The measured absorbance of the sample solution was used to calculate the amount of MO encapsulated in polymer microcapsules using the following equations.

MO in polymer microcapsules from experimental 0.1 g.

$$W_{MO;\ Capsules\ (Exp.)}\ (mg) = \frac{[MO] \times 25\ mL}{1{,}000} \quad (1)$$

MO in polymer microcapsules from theory 0.1 g.

$$W_{MO;\ Capsules\ (th.)}\ (mg) = \frac{W_{MO;\ th.}\ (g) \times 0.1\ g}{[PLLA\ (g) + MO\ (g)]} \times 1{,}000 \quad (2)$$

Loading efficiency (%)

$$\%L_E = \frac{W_{MO;Capsules\ (Exp.)}\ (mg)}{0.1\ (g) \times 10^3} \times 100 \quad (3)$$

$$\%L_{th} = \frac{W_{MO;Capsules\ (th.)}\ (mg)}{0.1\ (g) \times 10^3} \times 100 \quad (4)$$

Encapsulation efficiency (%)

$$\%EE = \frac{\%L_E}{\%L_{th}} \times 100 \quad (5)$$

Where

[MO] is the concentration of MO from calculation with the calibration curve (mg/L).

$W_{MO;Capsules\ (Exp.)}$ is the weight of MO (mg) in the polymer microcapsule obtained from the experiment.

$W_{MO;Capsules\ (th.)}$ is the weight of MO (mg) in the polymer microcapsule from the calculation.

L_E is the loading percent of MO in polymer microcapsule obtained from the experiment.

L_{th} is the loading percent of MO in polymer microcapsule obtained from the calculation.

RESULTS AND DISCUSSIONS

Solubility test

In the first step of polymer capsule preparation, the homogeneous oil droplets are required. Then, it is necessary to completely dissolve polymer and core substance using the appropriate solvent type and ratio. CAB, EC, and PLLA, biopolymers, were used as polymer shells at a ratio of polymer: MO of 1: 1. The prepared oil-phase solution requires the minimum amount of solvent that can completely dissolve the MO and polymer providing low viscosity suitable for internal phase separation. Polymers and MO were completely dissolved giving clear solutions for all conditions, as shown in Fig. 2. EA, a non-toxic solvent [24, 25], is a good solvent for CAB and EC. 1: 1: 6 and 1: 1: 10 were suitable ratios for CAB: MO: EA and EC: MO: EA, respectively, giving a homogeneous solution with low viscosity. For PLLA, DCM was used as a good solvent [26] at a ratio of 1: 1: 6.

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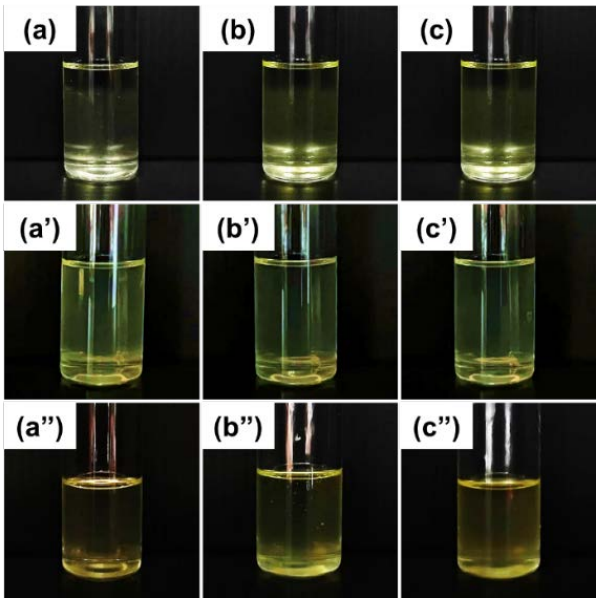

Figure 2 Solubility testing of CAB (a-c), EC (a'-c'), and PLLA (a''-c'') in a solvent at various ratios of Polymer: MO: solvent (w/w); CAB: MO: EA at 1: 1: 5 (a), 1: 1: 6 (b), and 1: 1: 7 (c); EC: MO: EA at 1: 1: 9 (a'), 1: 1: 10 (b'), and 1: 1: 11 (c') and PLLA: MO: DCM at 1: 1: 5 (a''), 1: 1: 6 (b''), and 1: 1: 7 (c'').

Effect of polymer type and polymer: MO ratio

The polymer microcapsules were prepared by the solvent evaporation method in an O/W system. The colloidally stable milky suspensions of the polymer/MO microcapsules were obtained for all types and ratios, as shown in Fig. 3.

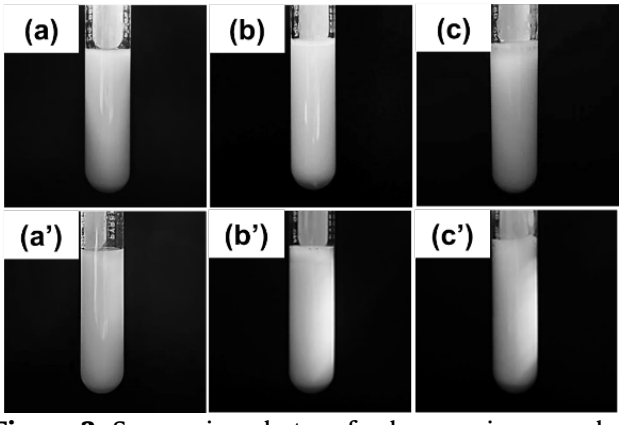

Figure 3 Suspension photos of polymer microcapsules using polymer: MO at 50: 50 (a-c) and 70: 30 (a'-c') of various polymers: CAB (a, a'), EC (b, b'), and PLLA (c, c').

The prepared polymer microcapsules were observed by optical microscope. Spherical microcapsules in a micrometer size were formed in all conditions, as shown in Figs. 4 and 5. The microcapsules were well distributed without coagulation. After drying, the polymer microcapsule powders were observed by SEM. Using EC, the microcapsules were spherical for both EC:MO ratios, as shown in Figs. 6b and 5b'. Rough surfaces or holes, which may be due to the evaporation of solvent were observed. However, at low polymer content (50:50), some aggregation of polymer microcapsules was found. The non-spherical CAB/MO microcapsules were formed with large aggregation due to the distribution of oil on the surface at a ratio of 50: 50, as shown in Fig. 6a. It is possibly because the low amount of CAB is not enough to completely encapsulate all of MO. When CAB content is increased to 70%, more stable spherical CAB/MO microcapsules are produced, as shown in Fig. 6a'. Similarly with CAB, stable PLLA/MO microcapsules at a ratio of 50:50 could not be prepared in dry state. Some aggregation of PLLA/MO microcapsules was clearly observed, as shown in Fig. 6c. In contrast, the spherical PLLA/MO microcapsules were formed at a ratio of 70:30 as shown in Fig. 6c'. Moreover, smoother surface than in the other conditions was clearly observed. After crushed, a core-shell morphology of the prepared microcapsules using all three types of polymers were clearly observed, as shown in Fig. 7. Phase separation of dissolved polymer chains and MO is smoothly proceeded during solvent evaporation leading to the formation of polymer shell enveloping the MO core. Therefore, all three types of polymers can be used to prepare stable MO encapsulated microcapsules at appropriate polymer to MO ratio.

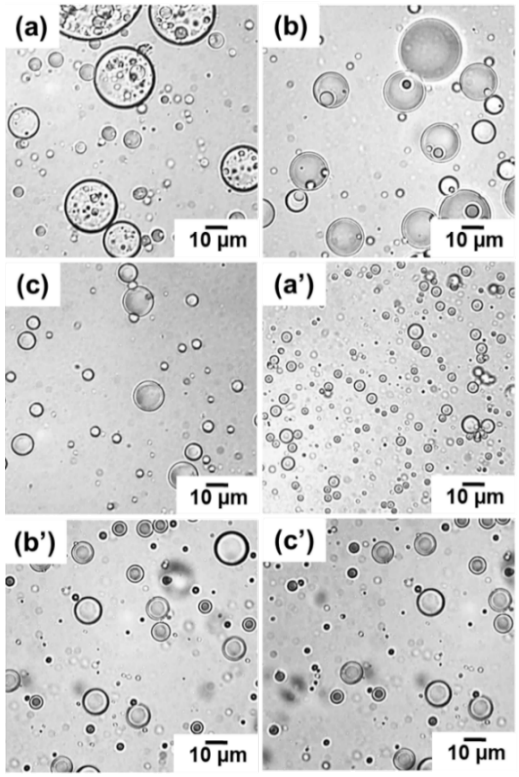

Figure 4 Optical micrographs of polymer microcapsules using Polymer: MO at 50: 50 before (a-c) and after solvent evaporation (a'-c') of various polymers: CAB (a, a'), EC (b, b'), and PLLA (c, c').

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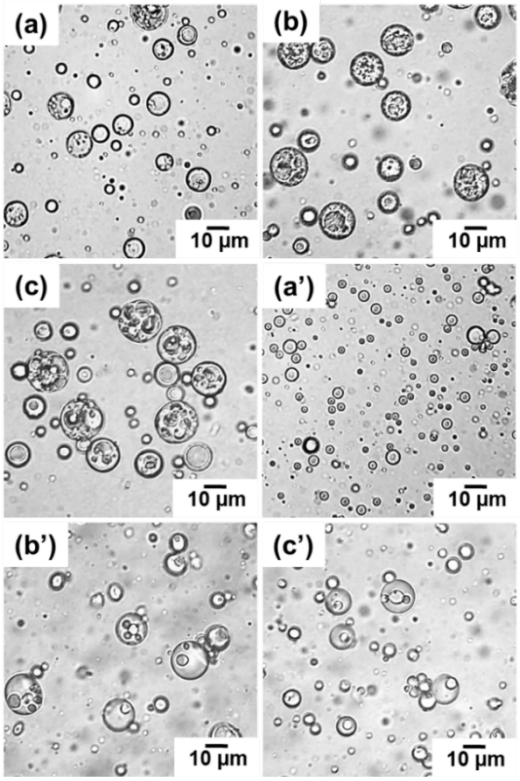

Figure 5 Optical micrographs of polymer microcapsules using Polymer: MO at 70: 30 before (a-c) and after solvent evaporation (a'-c') of various polymers: CAB (a, a'), EC (b, b'), and PLLA (c, c').

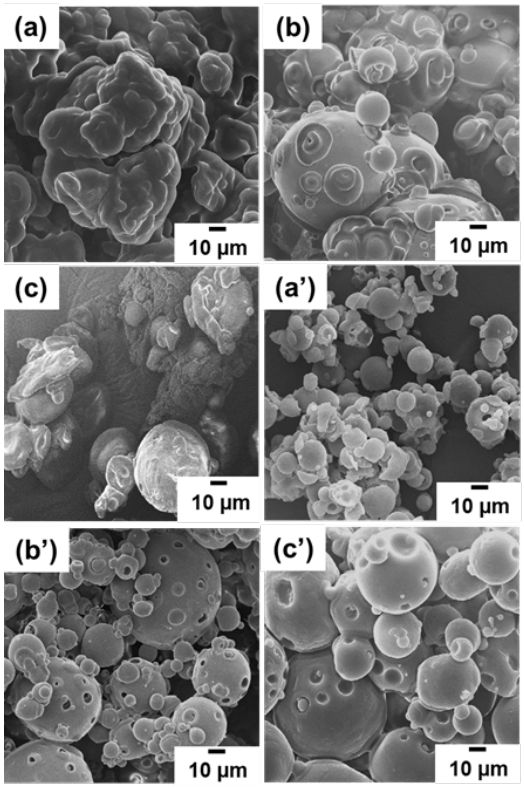

Figure 6 SEM micrographs of polymer microcapsules using Polymer: MO at 50: 50 (a-c) and 70: 30 (a'-c') of various polymers: CAB (a, a'), EC (b, b'), and PLLA (c, c').

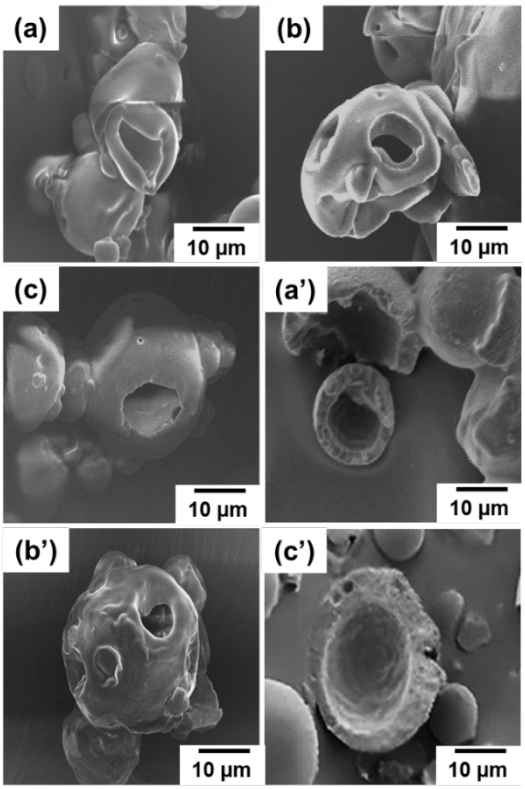

Figure 7 SEM micrograph of crushed microcapsules using Polymer: MO at 50: 50 (a-c) and 70: 30 (a'-b') of various polymers: CAB (a, a'), EC (b, b'), and PLLA (c, c').

Encapsulation efficiency

The loading and encapsulation efficiency (EE) of the prepared microcapsules were determined by UV-visible spectrophotometry, as shown in Table 4. The measured absorbance peak of the sample solution was used to calculate the amount of MO encapsulated in polymer microcapsules compared with the standard peaks of MO, as shown in Fig. 8. Corresponding with SEM observation, polymer microcapsules of all three kinds of polymers at a ratio of 50:50 presented low EE which may be due to the excessive amount of MO. Polymers are insufficient to totally encapsulate MO. EE increased with polymer content due to the larger amount of polymer needed to encapsulate MO for all types of polymers. Using PLLA at a ratio of 70:30 provided the highest EE at about 74%. This is probably due to the higher MW of PLLA (95,000 g/mol) used in this work than that of CAB (77,000 g/mol) and EC (51,000 g/mol). Using high MW polymer chains, MO was encapsulated more completely and formed a stronger shell [27]. As seen, only the dent occurs on the outer surface of PLLA microcapsules, as shown in Fig.7b' and 7c'. In contrast, the lowest %EE values of both EC/MO microcapsule ratios were obtained. Due to the rough surface with holes of the EC/MO microcapsules, some MO may loss to the outer region resulting in a reduction of the encapsulated MO content.

DOI: 10.60101/jarst.2023.253714

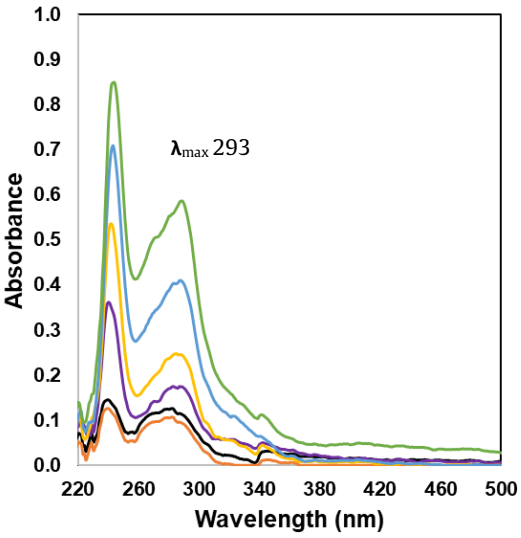

Figure 8 UV spectrum of MO standards (ppm); 2,000 (—), 4,000 (—), 6,000 (—), 8,000 (—) and 10,000 (—) and EC/MO microcapsules solution (—).

Table 4 Loading and encapsulation efficiency of polymer microcapsules prepared by solvent evaporation using various types of polymer.

Microcapsules	Polymer: MO	Loading (%wt)		Encapsulation (%wt)
		Experiment	Theory	
CAB/MO	50: 50	15.98 ± 0.17	50.44	31.68 ± 0.33
	70: 30	12.72 ± 0.03	30.02	42.37 ± 0.10
EC/MO	50: 50	12.34 ± 0.30	50.38	24.49 ± 0.58
	70: 30	12.28 ± 0.29	30.06	40.85 ± 0.96
PLLA/MO	50: 50	14.38 ± 0.16	51.13	28.12± 0.31
	70: 30	22.41 ± 0.29	30.25	74.08 ± 0.96

CONCLUSIONS

The stable spherical biopolymer microcapsules were successfully produced by simple solvent evaporation. All three biopolymer kinds, CAB, EC, and PLLA, can be used to produce spherical microcapsules encapsulating MO at appropriate polymer to MO ratio. Using PLLA, a stable core-shell microcapsule with a smooth surface were formed. The loading and EE increased with polymer content. Using a ratio of PLLA:MO of 70:30 presented the highest loading and EE. Because of its non-toxicity and biodegradability, it would be well applied to cosmetic products.

ACKNOWLEDGEMENT

This research project is supported by National Research Council of Thailand (NRCT): Contract No. N41A640334. The authors also thank BEAUTY MY COSMED CO., LTD. for their support.

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