

PREPARATION OF ESSENTIAL OIL NANOEMULSION WITH CELLULOSE DERIVATIVES

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ABSTRACT

The pomelo (*Citrus maxima*) peel's essential oil (EO) with many potential applications was proposed to be encapsulated via simple emulsification. The small particle size and homogenous distribution were mainly aimed at improving stability and aqueous dispersion. The low-energy method of magnetic stirring was applied. Although the emulsion could be fabricated successfully with only polysorbate, long-term stability could not be achieved. Hydroxypropyl methylcellulose (HPMC), hydroxypropyl cellulose (HPC), hydroxyethyl cellulose (HEC), and sodium carboxymethyl cellulose (CMC) were used as stabilizers. The particle size was significantly reduced with the addition of cellulose derivatives. The samples using CMC and Tween 60/80 or HPC and Tween 80 could maintain the average particle size and polydispersity index (PDI) even after 30 days. Based on dynamic light scattering measurements, the majority of particles were in the region of less than 40 nm and a Z-average of 126 nm could be obtained. Although the PDI values were still high (> 2), the average size of approximately 100 nm was still detected after one month of storage. The results are comparable to the previous studies, and the proposed process might open a simple approach to achieving a nanoemulsion state without using high-energy dispersion methods.

Keywords: Emulsion, polysorbate, cellulose derivatives, essential oil, nano-technology.

1. INTRODUCTION

Essential oils (EOs) are a mixture of various organic compounds that could be isolated from plants by cold-pressing, hydro-distillation, or supercritical carbon dioxide extraction and have a broad antibacterial activity [1, 2]. Because of their oily characteristics, EOs could not be well dispersed in water [3] while most bio-related applications require an aqueous environment. In addition, EOs with many volatile compounds are highly sensitive when they are exposed to oxidation, humidity, and high temperature. To improve the stability and applicability of natural substances, microencapsulation technology could be applied [4-6]. The technique has been considered as an efficient approach to improve both the preservation and delivery capabilities of EOs [7]. The technique was based on introducing a shell covering the core of bio-active compounds and preventing the effect of harsh conditions [8]. The encapsulation not only provided the controlled releasing of the bio-substances, it also could be used to enhance their bio-functionality. The encapsulation of many types of EOs and the size of the particles have been proven to significantly affect anti-bacterial capability [9, 10]. To encapsulate and increase the dispersion of EOs in water, many chemical, physical, and mechanical processes have been applied. The spray-drying technique might be applied for

micro-encapsulating the bioactive compounds [11, 12]. However, because of the thermally sensitive characteristic of EOs, the low-temperature encapsulation method of freeze-drying might be preferred [13, 14]. Nanoprecipitation was also proposed as another approach to encapsulate EOs [15]. However, emulsification might be the most popular, economical, and conventional method to achieve the protecting effect as well as the improvement of dispersion properties [16, 17]. To successfully perform the encapsulation technique, two factors including the bioactive core (EOs) and the type of shell or wall must be considered. The wall materials with unique physical chemistry properties played the most important and main role in the stability and the delivery behavior. Various types of proteins [18, 19], polysaccharides [9, 20], and lipids [21] were used to encapsulate EOs or bioactive constituents. Many popular and commercially available surfactants were also applied to emulsify EOs. The aggregation of lemon grass's EO could be improved by adding 0.4 wt% sodium dodecyl sulfate (SDS) owing to the electrical repulsion of ionic surfactant [22]. Donsi et. al reported the nano-emulsion of D-limonene of terpenes mixture by using natural surfactants: soy lecithin, modified starch, or Tween20/glycerol monooleate [23]. The emulsion fabricated with lecithin had the size of 74-240 nm while the average diameter of the mixture prepared from Tween 20/glycerol mono oleate was 130-155 nm. All the emulsions could be stable for four weeks without observable creaming and changes in particle size. Food-grade emulsifiers also could be applied to fabricate stable emulsion [24]. Nano-emulsion (particle size of 60 nm) of corn oil also was prepared with Tween 20 and SDS. Although sodium caseinate and β -lactoglobulin could cause an increase in particle size (up to 150 nm), the system was more physically stable. In a similar approach, many commercially available surfactants of span 20/80, Tween 80 [25], and Triton X-100 [26] could be used to stabilize oil in water (o/w) emulsion. In addition, polymers with biocompatibility could be applied to stabilize the emulsion. Chitosan and cellulose-based materials could become potential candidates. The combination of polysorbate surfactant and chitosan had been proposed to fabricate citrus EO emulsion by Li et al. [27]. The bacterial cellulose nanocrystal and fish gelatin are also used in the preparation of cinnamon EO [28]. The water-soluble cellulose derivatives of hydroxyethyl cellulose (HEC), sodium carboxymethyl cellulose (CMC), hydroxypropyl cellulose (HPC), hydroxypropyl methylcellulose (HPMC) were widely used in the food industry because of their biocompatibility [29, 30].

However, to the best of our knowledge, the effect of the cellulose derivatives as a stabilizer for EOs o/w emulsion has not been investigated. In most processes for nano-emulsion, the high-pressure homogenizer [31, 32] or rotor-stator high-speed stirring [33] must be applied to achieve a good dispersion. There was no research that reports the ability to fabricate nanoemulsion with the low-energy methods of magnetic stirring.

The aim of this study is to prepare a highly dispersed and stable emulsion with a simple method. Therefore, in this study, pomelo (citrus maxima) peel's EO in which D-limonene is the major component [34] shall be used as the main target to disperse in aqueous media under the form of oil in water emulsion. The polysorbate (Tween 20, 60, and 80) shall be used as the main surfactant. The effect of cellulose derivatives on particle size and the stability of the emulsion shall be investigated. The simple magnetic stirrer shall be applied to confirm the ability to prepare highly dispersed emulsion with low energy.

2. EXPERIMENTAL SECTION

2.1. Materials

The materials used in this study are from similar sources compared to the previous publication [35]. Polysorbate surfactants (Tween 20, 60, and 80) were obtained from Guangdong Guanghua Sci-Tech Cop. Ltd (China). CMC was the product of Nippon Paper

Industries. HPC (viscosity 2%, 20 °C: 1000 - 5000 mPa.s was from Wako Pure Chemical Industries (Japan). HEC (viscosity: 1500-2500 mPa.s) was purchased from Aladdin Industrial Corporation (Shanghai, China). HPMC (E6 type) from Shandong Head (Shandong, China) was used. Essential oil (Pomelo D-92, AOTA) was generously supplied by AOTA International Joint Stock Company (Viet Nam).

2.2. Methods

2.2.1. Preparation of o/w emulsion

The preparation procedure was conducted based on previous research [27, 36]. The polysorbate surfactants of Tween 20, 60, or 80 (0.5 mL) were added into a beaker containing 100 mL double distilled water. The solution was stirred at room temperature using a magnetic stirrer (600 rpm) for 30 min. Then, 100 μ L of EO was added and the mixture was continued to stir at the same speed for 60 min. The emulsion samples fabricated using only surfactant shall be named T.20, T.60, and T.80 for Tween 20, 60, and 80, respectively. The emulsion systems stabilized by cellulose derivatives were prepared in a similar method using a polymer solution (0.5 wt%) instead of pure water. The cellulose solution was prepared by adding 0.5 g of CMC, HEC, HPC, or HPMC into 100 mL double distilled water. The solution was kept stirring for more than 2 h until all of the polymeric powder was completely dissolved. The emulsions prepared by combining polysorbate and the polymers shall be named “*polymer abbreviation. Tween number*”. For example, the samples using Tween 60 and CMC shall have the name of CMC.60.

2.2.2. Characterization

In this study, various methods of turbidity, size analysis, and infrared spectroscopy were applied based on the previous publications [27, 37, 38].

2.2.2.1. Turbidity

The emulsion solution was evaluated with a turbidity meter (Mi415, Milwaukee Instrument, USA). All turbidity values detected at room temperature shall be reported in Formazin Nephelometric Unit (FNU). To confirm the stability, the turbidity was also recorded in 1 min intervals for 22 min right after the preparation of the emulsion.

2.2.2.2. Dynamic light scattering (DLS)

The dynamic light scattering (DLS) technique was applied to determine the particle size. The equipment of Zetasizer Pro, Malvern, UK was chosen to characterize the size analysis. The measurement conditions included: forward scattering mode, detector's angle of 17°, equilibrated time of 60 s, and temperature of 25 °C. The Z-average (along with the polydispersity index) based on Cumulants analysis was reported. Size distribution by intensity and by number were also reported.

2.2.2.3. Fourier transform infrared (FTIR) spectroscopy

The polymers and the mixtures with Tween 80 were dissolved in water. For FTIR characterization, essential oil was not used. The solutions were cast on a petri dish and evaporated gradually at room temperature. The film samples were obtained, and the attenuated total reflection (ATR) technique was applied for FTIR measurement. The equipment of MIR/NIR Frontier Perkin Elmer was used in the 4000 - 400 cm^{-1} region.

3. RESULTS AND DISCUSSION

EO was naturally extracted from pomelo fruit peel via hydrodistillation. The components of the oil were characterized using GC-MS. The main component in the EOs used in this study is D-limonene (92-96%). The essential oil was first dispersed in water by using polysorbate surfactants. Turbidity values as a function of time might provide a convenient way to estimate the stability of the o/w emulsion. The turbidity was monitored within 20 min right after the emulsion preparation. For comparison, the sample prepared by simply stirring the oil in water without the presence of surfactants was also characterized by the turbidity meter. Because each type of sample has various turbidities, the values were normalized to compare the stability during the first 20 min. The results were summarized in Figure 2.

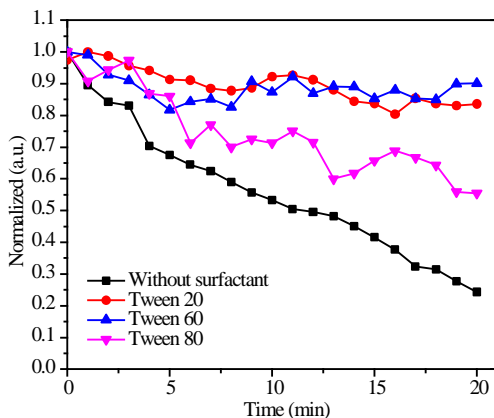


Figure 1. Normalized turbidity values of the samples with only polysorbate surfactants.

As can be observed, turbidity decreased rapidly in the absence of surfactant. However, the samples containing polysorbate displayed considerable improvement in stability despite a few fluctuations in turbidity. Unfortunately, the emulsion formed with Tween 20 and 60 showed aggregations after being stored at room temperature for 10 days. Although the samples with Tween 80 exhibited slightly more stability, phase separation was still observed after 16 days. It was concluded that the use of one type of polysorbate surfactant was insufficient in providing long-term stability to the EO emulsion. Therefore, water-soluble cellulose derivatives were added to the polysorbate emulsion, and the turbidity values were measured 20 min after preparation as shown in Figures 2 and 3. As a result, most samples with polymers displayed significant stability. However, the sample of HEC.60 showed a considerable reduction in the first 5 minutes, which could be a sign of aggregation. The sample with CMC exhibited the best stability, while turbidity fluctuations were still detected in the case of HPMC.

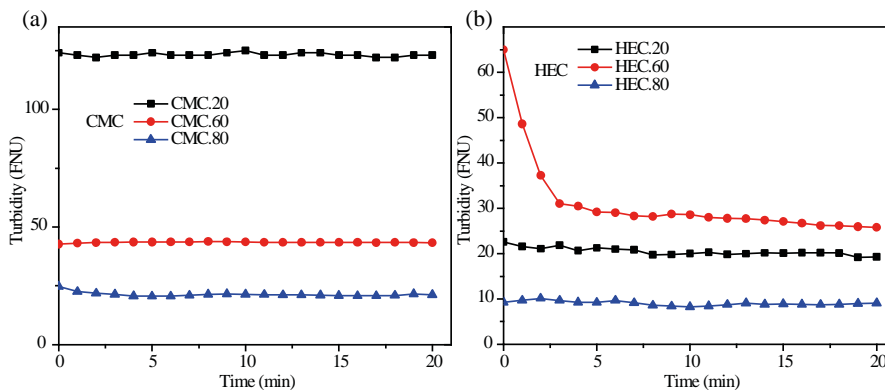


Figure 2. Turbidity values of the emulsion samples with CMC and HEC.

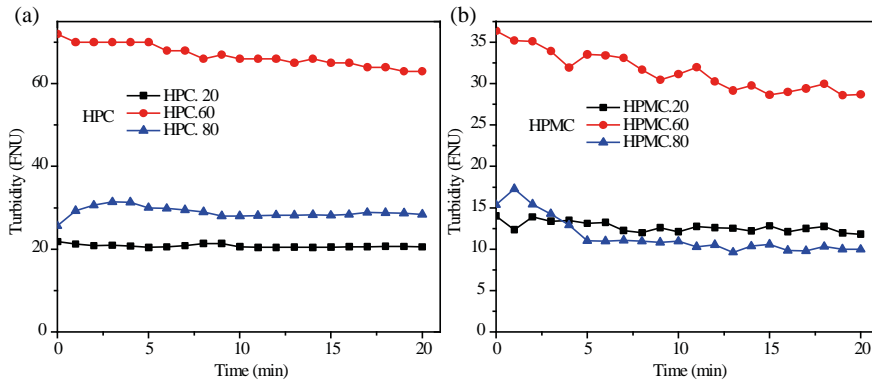


Figure 3. Turbidity values of the emulsion samples with HPC and HPMC.

The combination of Tween 20 or 80 and polymers showed high stability during the first 20 min. However, even with the assistance of polymers, the samples fabricated using Tween 20 showed a significant difference in turbidity after a long time of storage at room temperature. These results were reported in Figure 4. The CMC.20 and HEC.20 showed smaller turbidity while higher values were detected in HPC.20 and HPMC.20 after 15 and 30 days. Both cases with different directions still indicate the same tendency of instability. The aggregation and phase separation could be easily observed in all samples with Tween 20.

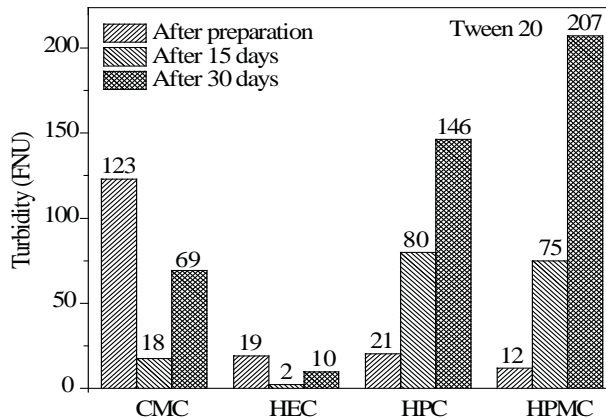


Figure 4. Turbidity values of emulsion fabricated with tween 20 and polymers.

The emulsion system with Tween 20 did not yield positive results. Therefore, particle size characterization was only conducted with Tween 60 and 80. However, the sample made with HEC and Tween 60 was highly unstable, and thus couldn't be characterized using the DLS technique to obtain reliable results. The DLS results (Z-average, PDI) and turbidity values for the samples that used Tween 60 in combination with cellulose derivatives were summarized in Table 1.

Table 1. DLS and turbidity results of emulsion prepared using Tween 60 and polymers

Samples	Z-Average (nm)	PDI	Turbidity (FNU)
T.60	1815 ± 424.7	13.77	33.99
CMC.60	641.7 ± 99.28	0.4017	43.29
HPC.60	532.3 ± 61.38	0.1929	63
HPMC.60	647.5 ± 207.2	1.33	28.7

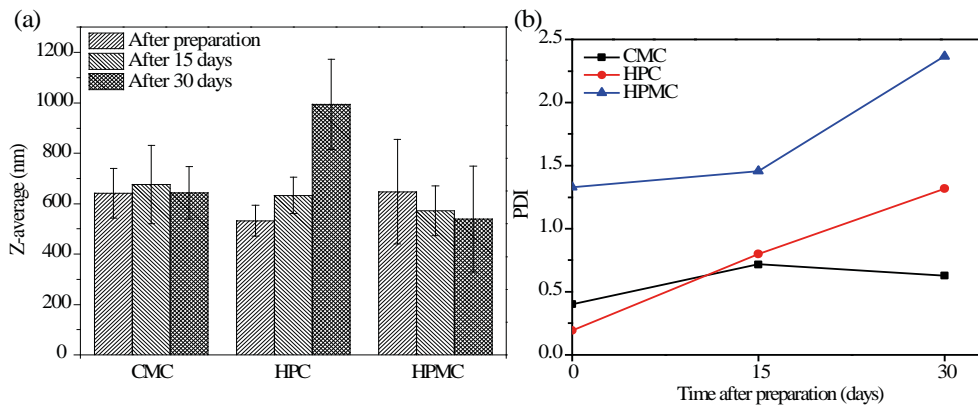


Figure 5. DLS results for samples fabricated with Tween 60 and polymers after long time storage.

As can be observed in Table 1, using polymers as a stabilizer greatly improved the emulsion's particle size and dispersity. Without polymers, the particle size was nearly 1.8 μm and the system was highly dispersed. Interestingly, by adding cellulose derivatives, the particle size was reduced by half in all cases. Additionally, PDI was significantly decreased, indicating a more homogeneous emulsion system. The smallest PDI was detected in HPC.60 samples. The addition of polymer didn't have a clear effect on turbidity. The DLS results suggest that even a low-shear method of magnetic stirring could produce a stable emulsion.

In the next investigation, the particle size was monitored after a long time of storage at room temperature and the results were summarized in Figure 5. Although the PDI values were small, the combination of HPC and Tween 60 couldn't maintain the same particle size after 30 days due to aggregation. The samples with CMC and HPMC showed similar particle sizes, but the PDI increased for the HPMC case, indicating instability. However, the PDI remained small for CMC.60 samples. Table 2 shows the results of the similar investigation with Tween 80.

Table 2. DLS and turbidity results of emulsion prepared using Tween 80 and polymers

Samples	Z-Average (nm)	PDI	Turbidity (FNU)
T.80	966.4 \pm 138.5	4.779	9.31
CMC.80	126.3 \pm 21.86	2.003	21.17
HEC.80	357.1 \pm 70.5	1.415	9.1
HPC.80	468.5 \pm 47.08	0.5678	28.4
HPMC.80	965.8 \pm 129.7	3.097	9.95

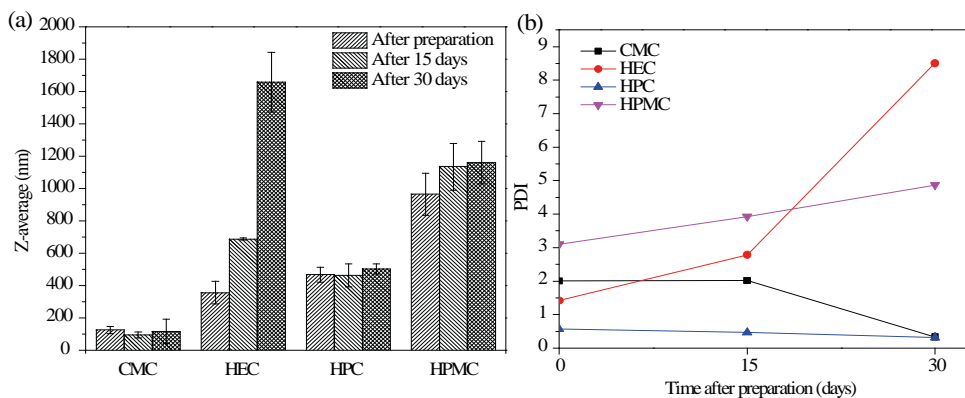


Figure 6. DLS results for samples fabricated with Tween 80 and polymers after long time storage.

Similarly, the particle size was rather large if only Tween 80 was used to disperse the EO in water as can be observed in Table 2. The large particle size might be the reason for the low stability of the emulsion with a single surfactant. Except for the case of HPMC.80, a significant reduction in particle size was also detected. The homogeneity of the emulsion was also improved when the obtained PDI values were smaller than before adding polymers. The addition of HPMC could not cause a noticeable change in the Tween 80 emulsion when the Z-average and PDI values were nearly unchanged. Interestingly, although the PDI values remained high (more than 2), the sample of CMC.80 could give a rather small average particle size (126 nm). The result is comparable to the previous nano-emulsion reports [23] and is better than that in the other publication with the assistance of chitosan [27]. This might open a new approach to fabricating a highly dispersed emulsion even with a simple and low-energy stirring method. Generally, the turbidity is smaller with the Tween 80 system. However, it could not guarantee small particle size and long-term stability. Even though the FNU turbidity of 9.31 was detected in the case of T.80, a particle size of 966 nm and a highly dispersed system ($PDI > 4$) were obtained. While the addition of CMC or HPC largely altered turbidity, the HEC.80 and HPMC.80 were nearly unchanged compared to the T.80 samples. This phenomenon might be because of less interaction between Tween 80 and HEC or HPMC. The change in particle size after a long period of storage might provide information about the stability of the emulsion. The results were conducted similarly with the Tween 60 system and were summarized in Figure 6. As can be seen, the sample with the addition of CMC could maintain a very small particle size even after 30 days. It indicates that the current combination of Tween 80 and CMC could strongly stabilize the emulsion of grapefruit peel essential oil. Although the size was not as small as CMC.80, HPC.80 also showed good stability in the Z-average and PDI after a month of storage. Even with the small turbidity values, the samples of HEC.80 and HPMC.80 showed the tendency of size increase, and the sign of phase separation could be observed after storing in the same conditions. The PDI in the case of CMC had a tendency towards slight reduction and the particle size was nearly unchanged during storage. Figure 7 presents the size distribution by scattering intensity and number of particles.

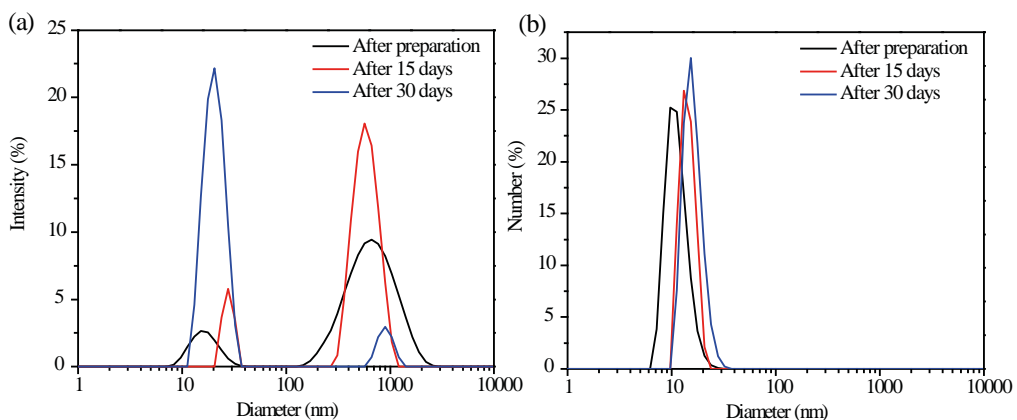


Figure 7. DLS particle size distribution for CMC.80 samples sample.

As can be observed in Figure 7a, there were two types of particle distribution systems. The large particles have a wide range from more than 100 nm to approximately 2.5 μm . Because of the presence of various diameter ranges, the rather large PDI value for this type of sample could be explained. However, in Figure 7b which is the size distribution as a function of a number of particles, the main number of particles was only less than 40 nm. The scattering intensity in the DLS technique is highly sensitive to the size of the particle. The bigger the size, the higher the signal. Even with a small number of large particles, the intensity signal might be significant. The result indicated that the emulsion in nanoscale dispersion could be

fabricated using a simple technique. Because of the small particle size, the sample with CMC and Tween 80 was highly stable even after long-term storage. The minor presence of the large particles might be due to some impurity. Besides, it is worth noting that the EOs from natural sources contained various components besides D-limonene. This might be also another reason for the high dispersity of the emulsion.

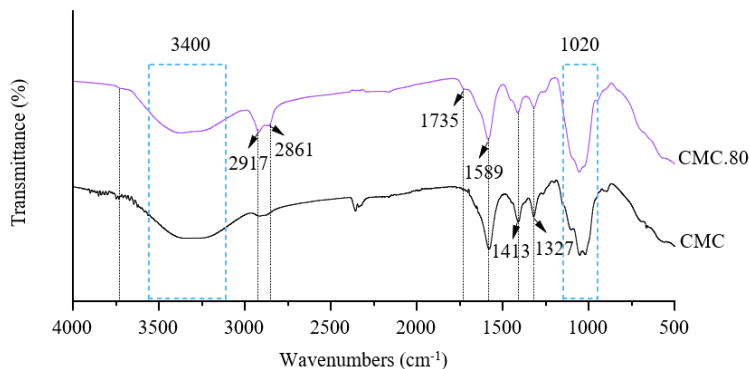


Figure 8. FTIR spectra of CMC and CMC blending with Tween 80.

Although the polymers in this study possessed a similar chemical structure, their effect on emulsion properties and stability were varied. All of the used cellulose derivatives could increase the viscosity of the solution. It is worth noting that the DLS characterization usually yields a higher particle size when the viscosity is higher. However, the obtained results were totally different. Therefore, it could be concluded that the cellulose derivatives, in this case, not only play a role as thickening agents but also they might have interacted with the used surfactant during the emulsion formation. Because CMC.80 showed the best and the most potential result, the FTIR measurement was conducted to confirm the chemical structure of CMC and the interaction with Tween 80. Figure 8 showed the FTIR spectra of CMC and CMC-Tween 80 films fabricated without the presence of essential oil. The broad peak at 3400 cm^{-1} represented the O-H stretching. The peak at 1589 cm^{-1} is evidence of carboxyl groups in the CMC molecules [39]. Similar to HPMC's FTIR spectra [40], the strong peak at 1020 cm^{-1} could be due to the cyclic rings with ether linkages. In general, most CMC.80's FTIR peaks were originally from CMC.80. There was only a few difference. The peak at 1735 cm^{-1} of C=O might belong to the ester group in Tween 80 molecular structure. The peaks at 2917 and 2861 cm^{-1} of aliphatic C-H could be observed clearly in only the CMC.80 mixture. The result showed that the CMC might have mostly covered the Tween 80 molecules. The FTIR peak shifting was not clearly detected indicating the intramolecular interaction was not too strong. It seems to provide a better stabilizing effect in this case. If Tween 80 and CMC firmly adhered to each other, the interaction of Tween 80 with EOs might be broken and lead to low stability. In addition, based on FTIR spectra, the carboxyl group of COO⁻ was not affected by combining with Tween 80. It also can provide a more substantial electrical charge surrounding the oil phase and prevent aggregation.

4. CONCLUSIONS

The o/w emulsion of pomelo peel's EO was successfully fabricated using polysorbate surfactants and magnetic stirring. The investigation of turbidity during the first 20 min after preparation showed a significant improvement compared to the samples without surfactants. However, when a single surfactant was used, the emulsion was unstable at room temperature after a long time of storage. Tween 60 and 80 samples were more stable than the case of Tween 20. However, the particle sizes of the Tween 60/80 emulsion were still large and highly poly-

dispersed. By adding cellulose derivatives (CMC, HPC, HPMC, and HEC), the particle size and PDI values of the emulsion based on Tween 60 or 80 could be significantly reduced. Many samples of CMC.60, CMC.80, and HPC.80 could maintain similar average particle size and PDI even after 30 days. CMC.80 sample could perform with the smallest Z-average (126 nm). After a month, the emulsion still could show a size of approximately 100 nm. Although with a relatively high PDI, DLS results for CMC.80 could show the size distribution of the major particles in the region of less than 40 nm. The study could open a new approach to fabricating stable emulsion with nano-size by using a simple dispersion method of magnetic stirring.

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TÓM TẮT

CHẾ TẠO NHŨ TƯƠNG NANO TINH DẦU VỚI DẪN XUẤT CỦA CELLULOSE

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Tinh dầu vỏ bưởi (*Citrus maxima*) (EO) với nhiều ứng dụng tiềm năng đã được sử dụng để bao gói thông qua phương pháp nhũ hóa đơn giản. Kích thước hạt nhỏ và sự phân bố đồng nhất là mục tiêu được hướng đến để cải thiện tính ổn định và khả năng phân tán trong nước. Phương pháp khuấy từ ít tiêu tốn năng lượng đã được áp dụng để chế tạo hệ nhũ tương. Mặc dù hệ nhũ tương có thể được chế tạo thành công chỉ với polysorbate nhưng không thể đạt được độ ổn định lâu dài. Trong nghiên cứu này, hydroxypropyl methylcellulose (HPMC), hydroxypropyl cellulose (HPC), hydroxyethyl cellulose (HEC) và sodium carboxymethyl cellulose (CMC) được sử dụng làm chất ổn định. Kích thước hạt giảm đáng kể khi bổ sung các dẫn xuất cellulose. Các mẫu sử dụng CMC và Tween 60/80 hoặc HPC và Tween 80 có thể duy trì kích thước hạt trung bình và chỉ số đa phân tán (PDI) sau 30 ngày. Dựa trên phép đo tán xạ ánh sáng động, phần lớn các hạt nhũ tương nằm trong vùng có kích thước nhỏ hơn 40 nm và có kích thước hạt trung bình (Z-average) là 126 nm. Mặc dù giá trị PDI vẫn còn cao (>2), kích thước hạt trung bình khoảng 100 nm vẫn được duy trì sau một tháng lưu trữ. Các kết quả có thể so sánh với các nghiên cứu trước đây và quy trình đề xuất trong nghiên cứu này có thể mở ra một cách tiếp cận đơn giản hơn để đạt được trạng thái nhũ tương nano mà không cần sử dụng các phương pháp phân tán năng lượng cao.

Từ khóa: Nhũ tương, polysorbate, dẫn xuất của cellulose, tinh dầu, công nghệ nano.