Effect of Different Combinations of Emulsifier and Wall Materials on the Physical Properties of Spray-dried Microencapsulated Swida wilsoniana Oil

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A B S T R A C T

Spray drying was used to produce microencapsulated Wilson Dogwood (Swida wilsoniana) oil. The influences of the emulsifier and wall materials on the encapsulation were studied in order to produce high quality encapsulated S. wilsoniana oil. The emulsions were prepared by using lecithin (L) and Tween 80 (T) with different hydrophilic-lipophilic balance. Results indicate that the stable emulsion was obtained using T and L as the compound emulsifier at a ratio of 4/6 (w/w). By virtue of the compound emulsifier (T/L 4/6), the microencapsulation efficiency (MEE) reached 91%, and the oil loading up to 35% were achieved. We also examined the influence of the wall materials on the microencapsulation of S. wilsoniana oil. All the three wall materials exhibited high MEE (>85%), and the highest MEE (95.20%) was obtained with sodium caseinate/lactose. All the S. wilsoniana oil encapsulated with the three wall materials exhibited nearly spherical microcapsules without pores or cracks—thus protecting the oil from oxygen.

1. Introduction

Wilson Dogwood (Swida wilsoniana), a member of the Cornaceae family, is an important woody plant with high yield and oiliness. The S. wilsoniana plant can maintain maximum productivity for more than 50 years and has a life span of more than 200 years. The oil content of the dry fruit of S. wilsoniana is between 33% and 36% (Li et al., 2015). In China, S. wilsoniana oil usually is used to promote human health. With an unsaturated fatty acid content of about 72% (oleic acid, 29.83%; linoleic acid, 39.15%; linolenic acid, 2.59%), long-term consumption of S. wilsoniana oil is believed to reduce cholesterol and prevent hyperlipidemia (Fu et al., 2012). However, one of the major complications in the commercialization of S. wilsoniana oil is rapid oxidation during processing and storage, leading to the formation of some toxic substances and unpleasant tastes and odors.

Microcapsule technology offers protection from environmental conditions by changing the nature of the material or its performance. This technology can shield peculiar smells, improve stability, and prolong shelf life (Pu et al., 2011; Shamaei et al., 2017; Timilsena et al., 2017). To improve the stability of the oil and expand its application, stable microencapsulation and high microencapsulation efficiency (MEE) are needed. A high-quality microencapsulated oil product can be prepared by spray drying. Atomization of the liquid is the most important step in spray drying. Operating conditions depend upon the physical and chemical properties of the emulsifiers to be spray-dried (Wu and Mujumdar, 2006). However, rare information is available on the microencapsulation of S. wilsoniana oil by spray drying, nor are there any comparisons in the literatures about the microencapsulation properties of wall materials for S. wilsoniana oil. In recent years, special attention has been focused on improving encapsulation efficiency during spray drying to prevent oxidation and extend shelf life (Tonon et al., 2011; Botrel et al., 2017).

Emulsification plays a critical role in optimizing the encapsulation efficiency of oils (Li et al., 2013; Noello et al., 2016). Few studies have evaluated the effects of emulsifiers on emulsion and microencapsulation properties. Soy lecithin (L), is derived from...
Soybeans and is a commonly used emulsifier in the food industry (Letyagina et al., 2014). Soy lecithin has a zwitterionic phosphocholine headgroup and two hydrocarbon tails. Tween 80 (T) is a nonionic, single-tailed surfactant which is also used as an emulsifier in foods (Athas et al., 2014). The hydrophilie-lipophile balance (HLB) of lecithin is eight and it is indeed oil-soluble. In comparison, the HLB of Tween 80 is 15 and it is water-soluble. An effective emulsifier may be obtained by combining this two amphiphiles.

The aim of this work is to produce microencapsulated *S. wilsoniana* oil using spray drying and investigate the encapsulated powder properties of the various emulsifiers after spray drying. Furthermore, the ability of lecithin and Tween 80 in combination with various HLB to emulsify *S. wilsoniana* oil was examined along with the influence of emulsifiers on encapsulation efficiency (Fig. 1). The effect of wall materials on the encapsulation efficiency of the powders was also evaluated to better understand the physiochemical properties of the microencapsulants to produce high quality encapsulated powders with maximum recovery for energy savings.

![Fig. 1 Pictorial scheme of formation of Wilson Dogwood (Swida wilsoniana) oil microencapsulation](image)

2. Materials and Methods

2.1. Materials and chemicals

The *S. wilsoniana* oil was obtained from Hunan Academy of Agricultural Sciences (Hunan, China). Gum Arabic, lactose, sodium caseinate, and soy lecithin were purchased from Aladdin chemistry Co. Ltd. (China). Soy protein isolate was purchased from Shanghai Yuanye Bio-Technology Co., Ltd. (China). All other chemicals used were of analytical grade.

2.2. Emulsion preparation

The following emulsions were investigated. Model 1 contained *S. wilsoniana* oil, gum Arabic, lactose, and various ratios of Tween 80 (T) and soy lecithin (L) as emulsifiers (T, T/L 6/4, T/L 4/6, L). Model 2 contained *S. wilsoniana* oil, lactose, gum Arabic/sodium caseinate/soy protein isolate, and Tween 80 and soy lecithin (4/6, w/w) as emulsifiers.

In Model 1, lactose and gum Arabic (3:1) were stirred and dissolved in distilled water stirring for 2 h at 25 °C. Identifying emulsification could affect the encapsulation efficiency properties of the spray-dried encapsulated *S. wilsoniana* oil powder, and *S. wilsoniana* oil with fixed ratios of core/wall material (0.33) and 1% surfactant with respect to the oil were progressively added to the wall material solution prior to emulsion preparation and stirred for 5 min at 10 000 r/min (T25, IKA Labortechnik, Germany). The emulsion was then formed by using a homogenizer (30 MPa, three times; GYB 30-6S, China).

In Model 2, for the wall material, lactose and gum Arabic/sodium caseinate/soy protein isolate (3:1) were stirred and dissolved in distilled water with stirring for 2 h at 25 °C. The *S. wilsoniana* oil with fixed ratios of core/wall material (0.33) and 1% surfactant (T/L 4/6) with respect to the oil were progressively added to the wall material solution during pre-emulsion preparation and stirred for 5 min at 10 000 r/min. The emulsion was then formed by using a homogenizer.
2.3. Spray-drying of sample

Spray drying was done with a mini spray dryer (H-Spray Mini, Holves, China) equipped with a 0.7-mm standard diameter nozzle. The operational conditions were as follows: air inlet temperature was (180 ± 2) °C, the air outlet temperature was (90 ± 2) °C, and the flow rate was ~8 mL/min. The dry powders were collected, hermetically sealed in packages, and stored at −18 °C until analysis.

2.4. Characterization of emulsion

2.4.1. Emulsion droplet size

Laser light scattering (Mastersizer 2000 Malvern Instruments Ltd, Malvern, UK) was used to determine the size distribution and average diameter of the emulsion droplets. The volume mean diameter (D_{v3}) was reported as the emulsion size. The samples were dispersed in water by the wet method. Measurements were conducted at 25 °C.

2.4.2. Creaming stability

The stability of S. wilsoniana oil emulsion was tested in terms of its creaming index (CI) according to the methodology described by Amr and Binsi (Edris et al., 2016; Binsi et al., 2017). Immediately after emulsion preparation, 25 mL aliquots of each emulsion were transferred to a cylindrical graduated glass tube, sealed, and stored at ambient temperature for 24 h. The volume of the upper phase was measured with a vernier caliper after 24 h. Stability was calculated using the following equation:

\[ \text{Emulsion stability index} = \left( \frac{V}{V_0} \right) \times 100\% \]  \hspace{1cm} (1)

where \( V_0 \) is the emulsion initial volume and \( V \) is the upper phase volume.

2.4.3. Viscosity measurement

The viscosity of emulsions was measured at 20 °C by using a dynamic rheometer (HAAK MARS II, Germany) equipped with a cone and plate system (gap 0.103 mm cone angle 2°). Viscosity-shear rate plots were obtained by applying an increasing shear rate from 4 s\(^{-1}\) to 100 s\(^{-1}\). All measurements were performed in triplicate 24 h after the emulsions were prepared.

2.5. Particle characterization

2.5.1. Moisture content

The moisture content of the microcapsules (4 g) was determined gravimetrically by oven drying at 103 °C to a constant weight (Ng et al., 2014).

2.5.2. Particle size distribution

Particle size distribution was measured according to the methodology described by Li et al. (2013) using a laser light diffraction-instrument, Mastersizer 2000 (Malvern Instruments, Malvern, UK).

2.5.3. Scanning electron microscopy (SEM)

The surface appearance and morphology of the microencapsulate were examined using SEM (S3400-I, Hitachi Ltd., Japan). Powder samples were fixed onto double-sided sticky tape mounted on SEM stubs, and sputter-coated in gold.

2.5.4. Microencapsulation efficiency (ME) and oil loading

Total oil content and surface oil were determined in order to calculate MEE and oil loading according to equations (2) and (3), respectively (Strobel et al., 2016).

\[ \text{MEE} = \left( \frac{\text{total oil} - \text{surface oil}}{100\%} \right) / \text{total oil} \]  \hspace{1cm} (2)

\[ \text{Oil loading} = \left( \frac{\text{total oil/microcapsules taken}}{100\%} \right) \times 100\% \]  \hspace{1cm} (3)

Total oil was determined according to Pond’s method (Pont, 1955). De-emulsifiers were prepared using 10 g of sodium citrate and 10 g of sodium salicylate dissolved separately in deionized water, and then mixed with 18 mL of n-butanol; the volume was topped up to 90 mL with deionized water. Next, 10 g of microencapsulated powder was mixed with 20 mL of water in an Erlenmeyer flask with a stopper at 50 °C. After 15 mL of de-emulsifier was added, the mixture was shaken vigorously and put in a 70 °C water bath for 6 min. This dispersion was then centrifuged at 3000 r/min for 10 min at ambient temperature, followed by collection of the total oil.

Surface oil was measured according to the previously described methods (Velasco et al., 2006). Briefly, 10 g of the dried microcapsule powder was dispersed in 200 mL of light petroleum ether (60–80 °C) followed by vortexing at 25 °C in the dark for 15 min. The dispersion was then filtered using a Whatman No. 4 filter paper, collected in a round-bottom flask, and evaporated using a rotary evaporator at 30 °C to recover the oil (Velasco et al., 2006).
2.6. Statistical analysis

All experiments were performed at least in triplicate. Analysis of variance was carried out to assess differences between factor means. Differences were considered statistically significant at \( P < 0.05 \).

3. Results and Discussion

3.1. Emulsion characterization

3.1.1. Emulsion droplet size

Droplet size is an important indicator of emulsion stability. Results of this study show that the encapsulation efficiency of microencapsulates can be improved by decreasing the emulsion droplet size as the larger droplets may be breakdown during atomization leading to the higher surface oil (Tonon et al., 2011). The small oil droplets can be enclosed efficiently within the wall matrix of microcapsules (Jafari et al., 2008). As previously explained, emulsions were produced by using various emulsifiers. Four HLB emulsions with HLB in the range of 8–15 were prepared for the microencapsulation of S. wilsoniana oil. Table 1 presents the droplet mean diameters for each of these emulsions. Droplet mean diameter varied from \((4.43 \pm 0.017)\) μm to \((29.99 \pm 0.026)\) μm. The emulsions had relatively small droplet sizes except where the emulsifier was soy lecithin; in that case, droplet size was \((29.99 \pm 0.026)\) μm. The emulsions prepared by the mixture of Tween 80 and soy lecithin have relatively small droplet size, which may be attributed to the synergy between them: L and T can get stable at the oil–water interface, and the large headgroup of T prevents oil droplets coalescence by steric repulsions.

3.1.2. Creaming stability

Emulsion stability is important in microencapsulation. We found that emulsion stability was markedly affected by the type of emulsifier used, which further influenced the characteristics of the spray dried powder. Emulsion stability was measured using a CI (Table 1). A gradual phase separation occurred after 2 h in both Tween 80 and Tween 80/soy lecithin (6/4). After 24 h, a homogenous original phase fraction of \((45.27 \pm 1.30)\)% and \((10.89 \pm 2.43)\)% occurred, which were not suitable for spray drying. The other emulsions remained stable (CI = 0) for a 24 h period, which could be due to the formation of network structures in the emulsions (Ye and Singh, 2006). The results showed that neither T nor L was effective emulsifiers for spray-dried S. wilsoniana oil. A combination of T and L, especially at a weight ratio of 4/6, however, did provide an effective oil-in-water (O/W) emulsifier, were capable of forming stable emulsions that resist creaming through the synergy of L and T at the oil-water interface.

3.1.3. Emulsion viscosity

In processing, emulsion viscosity is important because it affects encapsulation efficiency and oil retention. The viscosities of the S. wilsoniana oil emulsions prepared with lactose and gum Arabic (as wall materials) in combination with various ratios of T and L (as emulsifiers) are shown in Fig. 2. All emulsions behaved like non-Newtonian fluids when subjected to shear stress below 25 S⁻¹ with a shear-thinning behavior. The viscosity was fairly insensitive at very high shear rates, indicating Newtonian behavior (Bakry et al., 2016). The T/L (6/4) emulsion had the lowest viscosity, which may be due to the differences in chemical composition, and potentially resulting in changes in emulsions properties (e.g., droplet size, interfacial properties) and increased viscosity. A moderate viscosity is desirable for spray drying. Increasing the viscosity to an optimal value can reduce the oscillation and circulation of internal droplets, resulting in improved oil retention (Jafari et al., 2008). Maximum viscosity was reached with the incorporation of T/L (4/6) surfactant.

3.2. Microencapsulation by spray drying

3.2.1. Moisture content

Moisture content is important in the powder-quality and shelf life of

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**Table 1** Droplet size distribution and creaming stability of Wilson Dogwood (Swida wilsoniana) oil emulsions with different combinations of emulsifier (Model 1)

<table>
<thead>
<tr>
<th>Emulsifier</th>
<th>Emulsion size (D₄₃, μm)</th>
<th>Creaming stability</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tween 80</td>
<td>5.67 ± 0.020</td>
<td>(45.27 ± 1.30)%</td>
</tr>
<tr>
<td>Tween 80/soy lecithin (6/4)</td>
<td>4.43 ± 0.017</td>
<td>(10.89 ± 2.43)%</td>
</tr>
<tr>
<td>Tween 80/soy lecithin (4/6)</td>
<td>5.01 ± 0.015</td>
<td>0</td>
</tr>
<tr>
<td>Soy lecithin</td>
<td>29.99 ± 0.026</td>
<td>0</td>
</tr>
</tbody>
</table>

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**Fig. 2** Viscosity of emulsions of S. wilsoniana oil stabilized by different emulsifiers, versus shear rate.
spray-dried products. Excessive moisture in these products causes powder agglomeration, microbial growth, and caking, which result in the release/oxidation of the oil. Table 2 shows that the moisture content of spray-dried microcapsules varied with the wall material from (1.08 ± 0.215)% to (2.89 ± 0.294)%. The moisture content of the sodium caseinate/lactose blend was the highest, while that of the gum Arabic/lactose blend was the lowest. This variation in moisture content between powders is related to the chemical structures of the different wall materials. Compared with other spray-dried microcapsules, the moisture content in our products was relatively low, which may be attributed to the high inlet and outlet air temperatures during spray drying. The relatively low moisture content was expected to minimize oxidation and microbial contamination of the core material.

3.2.2. Encapsulation efficiency and oil loading
Encapsulation efficiency refers to the potential of the wall material to encapsulate the core material inside the microcapsule; Oil loading is one of the most important parameters of microencapsulated powders. Since oil on the microcapsule surface is easily oxidized, the surface and total oil content of the microcapsules were used to evaluate encapsulation efficiency. As shown in Table 3, the use of compound surfactants (T/L, 4/6) markedly improved encapsulation efficiency (91.06 ± 0.96)%, indicating that only a little of oil remained unencapsulated in the Tween 80/soy lecithin (4/6) emulsified microcapsules. Furthermore, the payload of S. wilsoniana oil microcapsules with the Tween 80/soy lecithin (4/6) was (35.53 ± 2.21)%. This suggests that nearly all the oil is encapsulated within the shell matrix. The combinations (Tween 80/soy lecithin) are more stable because T/L blends have a synergistic effect, that is, in combination, they offer an effective emulsifier, which may positively influence encapsulation efficiency of the microencapsulated powders. The overall result was that the composition of emulsifier significantly influenced encapsulation efficiency and other measured data such as emulsion size. In addition, microencapsulated powders obtained from emulsions stabilized by (T/L, 4/6) had efficient encapsulation, with most of the S. wilsoniana oil being retained in the particles.

As shown in Table 4, the MEE of encapsulated S. wilsoniana oil ranged from (85.25 ± 1.50)% to (95.20 ± 1.85)%%. Sodium caseinate blends and gum Arabic blends had higher MEE (95.20 ± 1.85)% and (91.06 ± 0.96)%, which meant that both blends were effective in maintaining the S. wilsoniana oil inside the microparticles during spray drying, thus limiting oxidation during processing and storage. The sodium caseinate blends had higher MEE than gum Arabic and soy protein isolate blends, may due to the reaction between the amino groups of sodium caseinate and the carbonyl groups of lactose. These groups can form conjugates through the Maillard reaction. The Maillard reaction stabilizes oil microcapsules by changing the physical properties of the wall and contributing to wall formation (Ferreira et al., 2016). Such changes in the wall retain and protect the core material, thus increasing the MEE. Another reason for the higher MEE in the sodium caseinate blends is the susceptibility of the soy protein isolate to heat denaturation during spray drying (Rodea-González et al., 2012). The payload of gum Arabic blends was the most (35.53 ± 2.21)%, which meant the microparticles produced using gum Arabi had more total oil and less surface oil than those made with sodium caseinate or soy protein isolate. The surface oil was the most important factor that affected the microparticles stability since the oil droplets on the surface of the powders were easily to be oxidized.

3.2.3. Size distribution of microparticles
The sizes of the spray-dried microcapsules are shown in Table 2. Wall material with soy protein isolate had significantly (P < 0.05) smaller particle size than that of the other two wall materials. Particle sizes of spray-dried microcapsules with three wall materials were in the range of 3.21–8.69 μm. The sodium caseinate/lactose blend with particle size of (7.43 ± 0.020) μm had the highest MEE, while S. wilsoniana oil encapsulated with soy protein isolate/lactose had smaller particles sizes, which may be attributed to the soy protein isolate and lactose exhibited much higher ability to be redispersed in water when reconstituted emulsions (Tang and Li, 2013). Smaller microcapsules possess larger surface area which may cause more oil for migrating to the surface of the microcapsule during the spray drying or even during the storage period (Botrel et al., 2017). On the other hand, the

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Moisture content and particle size distribution of microcapsules using different wall materials (Model 2)</th>
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</thead>
<tbody>
<tr>
<td>Wall material</td>
<td>Moisture content (%)</td>
</tr>
<tr>
<td>Gum Arabic/lactose</td>
<td>1.08 ± 0.215</td>
</tr>
<tr>
<td>Sodium caseinate/lactose</td>
<td>2.89 ± 0.294</td>
</tr>
<tr>
<td>Soy protein isolate/lactose</td>
<td>1.34 ± 0.194</td>
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</tbody>
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<tr>
<th>Table 3</th>
<th>Microencapsulation efficiency (MEE (%)) and payload of S. wilsoniana oil with different surfactants (Model 1)</th>
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<tbody>
<tr>
<td>Emulsifier</td>
<td>MEE (%)</td>
</tr>
<tr>
<td>Tween 80</td>
<td>91.63 ± 0.93</td>
</tr>
<tr>
<td>Tween 80/soy lecithin (6/4)</td>
<td>87.90 ± 1.02</td>
</tr>
<tr>
<td>Tween 80/soy lecithin (4/6)</td>
<td>91.06 ± 0.96</td>
</tr>
<tr>
<td>Soy lecithin</td>
<td>51.04 ± 0.76</td>
</tr>
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</table>

<table>
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<tr>
<th>Table 4</th>
<th>Microencapsulation efficiency (MEE (%)) and payload of S. wilsoniana oil with different wall materials (Model 2)</th>
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<tbody>
<tr>
<td>Wall material</td>
<td>MEE (%)</td>
</tr>
<tr>
<td>Gum Arabic/lactose</td>
<td>91.06 ± 1.06</td>
</tr>
<tr>
<td>Sodium caseinate/lactose</td>
<td>95.20 ± 1.18</td>
</tr>
<tr>
<td>Soy protein isolate/lactose</td>
<td>85.25 ± 1.50</td>
</tr>
</tbody>
</table>
decrease in MEE value of the SL blend maybe also due to heat denaturation of the soy protein isolate during spray drying (Lim et al., 2012).

3.2.4. Scanning electron microscopy
Through scanning electron microscopy (SEM) observations (Fig. 3a, Fig. 3b), it is found that microcapsule particles encapsulated with gum Arabic/lactose had smooth surfaces. However, in the soy lecithin-emulsified particles, there were other larger, irregularly shaped agglomerates. The large agglomerates may result from the high viscosity of the feed emulsion or from the potential for partial flocculation of the feeding emulsion, which can lead to agglomerate formation during spray drying due to poor emulsion stability (Silva et al., 2016). The SEM images of S. wilsoniana oil microcapsules with various wall materials are shown in Figs. 3b–3d. The particles had smooth surfaces with no apparent cracks to permit the entry of gases and solvents, thus increasing the MEE (Rodea-González et al., 2012). Microcapsules obtained from soy protein isolate and lactose were much smaller than those produced using the other two wall materials. Large and small particles were formed, consistent with the dynamic light scattering data.

![SEM images of spray-dried emulsions of S. wilsoniana oil encapsulated with different wall materials](image)

a: gum Arabic/lactose, surfactant (L); b: gum Arabic/lactose, surfactant (T/L 4/6); c: sodium caseinate/lactose, surfactant (T/L 4/6); d: soy protein isolate/lactose, surfactant (T/L 4/6)

**Fig. 3** Scanning electron microscopy images of spray-dried emulsions of S. wilsoniana oil encapsulated with different wall materials

4. Conclusions
This study evaluated the influence of emulsion properties and wall materials on the encapsulation powder properties of S. wilsoniana oil microcapsules produced by spray drying. The S. wilsoniana oil emulsion characterization (oil particle size, emulsion stability, and viscosity) was revealed that samples emulsified with T/L (4/6) had greater emulsion stability and smaller oil particles. This finding was attributed to the formation of stable structure by the synergy between L and T. Three S. wilsoniana oil microcapsules were made by spray drying using various wall materials and with T/L (4/6) as the emulsifier. For each of the three wall materials, encapsulation efficiency exceeded 85%. Sodium caseinate/lactose had the highest encapsulation efficiency of 95.20%. The SEM images showed that, for each of the wall materials, the microcapsules were nearly spherical, without pores or cracks, offering protection from oxygen.

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Conflicts of interest

There are no conflicts to declare.

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