Hindawi
Journal of Food Quality
Volume 2022, Article ID 5898937, 11 pages
https://doi.org/10.1155/2022/5898937

Research Article

Optimization of Emulsification and Microencapsulation of Balangu (*Lallemantia royleana*) Seed Oil by Surface Response Methodology

Parisa Izadi-NajafAbadi[1] and Asiye Ahmadi-Dastgerdi[2]

[1]*Department of Biotechnology and Food Science, Norwegian University of Science and Technology, Trondheim, Norway*
[2]*Department of Food Science and Technology, Ardestan Branch, Islamic Azad University, Ardestan, Iran*

Correspondence should be addressed to Asiye Ahmadi-Dastgerdi; as.ahmadi17@gmail.com

Received 13 April 2022; Revised 20 May 2022; Accepted 7 June 2022; Published 18 June 2022

Academic Editor: Walid Elfalleh

Copyright © 2022 Parisa Izadi-NajafAbadi and Asiye Ahmadi-Dastgerdi. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

Balangu (*Lallemantia royleana*) seed oil is a valuable source of omega-6 fatty acids that reduces the risk of cardiovascular diseases. Due to the high sensitivity of this oil to environmental factors, microencapsulation has been recommended to preserve valuable compounds of oils and prevent adverse environmental effects. In this study, the oil of balangu seeds was extracted using a combination of ultrasound and shaking incubation and was microencapsulated using an emulsification method. The process was optimized using the response surface methodology (RSM). For this purpose, the effect of three independent variables such as chitosan concentration (0–1.5%), sodium alginate concentration (0–4.5%), and pH (3–7) on emulsification and microencapsulation condition was analyzed. The results showed that the optimal conditions for emulsification and microencapsulation included 0.30% chitosan, 0.14% sodium alginate, and pH 3. Scanning electron microscopy (SEM) showed that the structure of the optimal sample was smooth, spherical, and without cracks, which confirms the success of emulsification and microencapsulation processes.

1. Introduction

Oil seeds are valuable sources of unsaturated fatty acids, protein, fiber, amino acids, antioxidants, and other nutrients [1]. Balangu (*Lallemantia Royleana*) from the mint family is one of the oilseeds that has about 30% of oil and is a rich source of omega-3 fatty acids. Balangu seed oil contains linolenic acid, which makes it a good source of omega-3 fatty acids. It is a very good source of tocopherol, phenolic compounds with high antioxidant activity [2, 3].

The conventional oil extraction methods such as Soxhlet extraction have limitations such as high solvent consumption, extraction time, loss of volatile compounds, low efficiency, and degradation of unsaturated fatty acids [4]. The shaking incubation extraction causes a complete contact of the solvent with the solid phase and thus increases the extraction efficiency [5]. The emergence of new methods of oil extraction eliminates the disadvantages of conventional methods of oil extraction. One of the new technologies for oil extraction is the use of ultrasound, which is more economical than other new extraction methods and reduces the amount of solvent [4]. In the extraction of vegetable oils (grape seeds, mango kernel, pomegranate kernel, and pistachio kernel) using ultrasound compared to the Soxhlet method, the oil extraction efficiency has increased and the oils were less oxidized [4, 6–8].

Oil protection from environmental factors is also important. Vegetable oils oxidize easily by heating, chemical reactions, and light. Microencapsulation is a good way to preserve valuable compounds of vegetable oils and prevent adverse environmental effects that have recently received much attention. In this technology, the target compounds are coated by the wall compounds to form microcapsule particles [9].

The emulsification is one of the microencapsulation methods which release the functional compounds. In emulsification, the continuous phase consists of aqueous solution and the dispersed phase consists of wall constituents [10]. Jafari et al. [11] showed that the smaller particles of the emulsion have the better microencapsulation process. Quispe-Condori et al. [12] reported that the microencapsulated polyunsaturated fatty acids of flaxseed oil were not degraded during the spray drying process but were reduced during the freeze drying process. Mohammadi et al. [13] showed that nanocapsulation of the olive leaf by emulsification method increased the stability of oil to oxidation. Asadpour et al. [14] showed the nanoemulsification of folic acid using maltodextrin and whey protein increases the emulsion particle size.

In this study, emulsification and microencapsulation of balangu seed oil have been investigated to preserve its valuable nutrients. The process conditions were optimized using the response surface methodology (RSM).

2. Materials and Methods

2.1. Materials. Whey protein concentrate and maltodextrin were obtained from Hirad Powder Co (Tehran, Iran). Chitosan, sodium alginate, and all chemical reagents were purchased from Merck Company (Germany).

2.2. Sample Preparation. Balangu seeds were purchased from the local market (Isfahan, Iran) in May 2021. After grinding, the powder was stored in plastic bags in the refrigerator [15].

2.3. Oil Extraction. In the Soxhlet method, 50 g of sample were placed in an extractor and the extraction was carried out with 300 ml of n-hexane for 8 hours at 40°C and the residual solvent was removed an oven (Model EO 155, Shimifann, Iran) at 30–40°C for 2 hours. In the shaking incubation-ultrasonic method, 50 g of Balangu seeds powder was placed in a laboratory incubator (Model Fan Azma Gostar, Tehran, Iran) and then in an ultrasonic bath (Model 300, Pulse, Italy) at 25 kHz. The oil was centrifuged (Model 2–16P, Sigma, Germany) at 7800 rpm for 20 minutes. Finally, the residual solvent was evaporated in a rotary evaporator (Model Heidolph, Germany) for 15 min at 115 rpm and 50°C [15].

2.4. Emulsification Process. The high energy emulsification method and ultraturrax homogenization-ultrasound combination were used to prepare oil in water emulsion (O/W). The wall material including chitosan (0–1.5%), sodium alginate (0–4.5%), whey protein concentrate (15%), and maltodextrin (10%) were mixed with balangu seed oil and distilled water. The mixture was homogeneous with ultraturrax for 3 minutes. The pH of the emulsion was adjusted (pH: 3–7) by 0.1 N sodium hydroxide and hydrochloric acid. The emulsion was homogenized for 6 minutes and placed in an ultrasound bath (25 kHz) at room temperature for 10 minutes. The final emulsion was stored in a freezer at −18°C [11].

2.5. Emulsification Tests

2.5.1. Particle Size and ζ-Potential Measurement. The particle size of the emulsions was measured using the dynamic light scattering instrument (Malvern Instruments Ltd., United Kingdom). ζ-potential was measured using a commercial microelectrophoresis instrument (Zetasizer ZA series, Malvern Instruments Ltd., UK). Samples were diluted using 10 mM phosphate buffer prior to analysis [16].

2.5.2. Emulsion Stability Index (Creaming Index). First, 10 ml of the emulsion was poured into a test tube and centrifuged at 7800 rpm for 20 minutes. Then, the total height of the emulsion and the height of the cream layer were measured using a ruler.

2.5.3. Turbidity. The emulsion (0.5 ml) was mixed with distilled water and the sample absorption was determined using a UV-visible spectrophotometer at 600 nm (Model T70, PG, United States) [17].

2.5.4. Antioxidant Activity. To measure the antioxidant activity, 3.5 ml of DPPH methanolic solution was added to 0.1 ml of emulsion, and after 10 minutes, the absorption sample was read at 517 nm using a spectrophotometer. The DPPH solution was considered as a control sample. The antioxidant activity was calculated using the following equation [18]:

$$\text{The radical inhibitors}\,(\%) = \frac{\text{adsorption control} - \text{adsorption sample}}{\text{adsorption control}} \times 100. \quad (1)$$

2.6. Microencapsulation Process. All prepared emulsions were dried in a freeze-dried for 24 hours (1 hPa and -20°C) using a freeze dryer (Model DW8030, USA). After drying, the powders were stored at −18°C for further analysis (Quispe-Condori et al., 2011).

2.6.1. Microencapsulation Efficiency. The microencapsulation efficiency was calculated using the following equation:

The efficiency of extracted oil = 1—(surface oil/total oil) × 100.

To measure surface oil, 0.5 g of the powder was mixed with 10 ml of hexane and its absorbance at 228 nm was determined. To measure total oil, the powder was dissolved in water (1 : 10) and 20 ml of hexane. The mixture was put through ultrasound for 2 minutes at 100 Hz at an ambient temperature using an ultrasound probe. The sample was centrifuged for 15 minutes at 7500 rpm and then its absorption was determined at 228 nm [19].

2.6.2. Moisture Content. The moisture content of the microcapsulated powder was calculated using the gravimetric method in the oven at 60°C for 2 h [20].

2.6.3. Oil Release Test in Simulated Gastric (SGF) and Intestinal Environment (SIF). To prepare the SGF solution, 2 g of sodium chloride was dissolved in 900 ml of distilled water and the pH of the solution was increased to 1.2 using 36% hydrochloric acid. Then, 3.2 g of pepsin was added to the solution and was kept at 4°C. To prepare the SIF solution, potassium hydrogen phosphate and water were mixed in a ratio of 6 : 400 (w/v) and 77 ml of sodium hydroxide. Then, using HCL and NaOH, the pH of the solution was adjusted to 6.8. Finally, 10 g of pancreatin was added to the solution and was kept at 4°C for later use.

In the digestion step, 2 g of the finely microencapsulated powder was mixed with 20 ml of SGF solution and placed in a water bath (38°C) for 2 hours. Then, 20 ml of SIF was added to the solution and placed in a vibrating water bath for 3 hours. Then, 25 ml of hexane was added to the solution. Finally, the oil and hexane were rotated at 60°C to evaporate the hexane. The oil release rate was calculated using the following equation [21].

$$\text{Oil release}\,(\%) = \frac{\text{primary weight} - \text{Second weight}}{\text{total oil}} \times 100. \tag{2}$$

2.6.4. Scanning Electron Microscopy (SEM). The optimal treatment was coated with a thin layer of gold alloy and then analyzed using scanning electron microscope (Zeiss, Germany) with a magnification of 5000 × and a voltage of 10 kV.

2.7. Experiment Design. The effects of three independent variables of chitosan concentration (0–1.5%), sodium alginate concentration (0–4.5%), and pH (3–7) on dependent variables were analyzed (Table 1).

The central composite design (CCD) using Minitab software version 16.1.1.0 was used to design 20 treatments (Table 2). The quadratic equation used for regression analysis of the experimental data for each dependent response as a function of independent variables is shown as follows:

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i=j=1}^{k} \beta_{ij} x_i x_j, \tag{3}$$

where β_0, β_i, β_{ii}, and β_{ij} are constant coefficient, linear regression coefficient, quadratic regression coefficient, and interaction regression coefficient between the two variables, respectively.

The mathematical models indicated the function of the independent variables on dependent responses as shown in the following equations:

3. Results and Discussion

3.1. Particle Size. The mean particle diameter and ζ-potential of optimal emulsion were 30.9–74.39 nm and—30.4 mV, respectively.

3.2. Creaming Index. The emulsion stability ranged from 6 to 32%. Three-dimensional response plots for the stability index are presented in Figure 1. The highest emulsion stability (52.4%) was observed at pH 3, 0% chitosan, and 4.5% sodium alginate and the lowest emulsion stability (2.89%) was observed at pH 7, 0.84% chitosan, and 3.4% sodium alginate.

According to Figure 1(a), at pH values 3 and 7, with increasing the concentration of chitosan from 0 to 0.5%, the stability index of the emulsion decreased and with further increase of the concentration from 1.2 to 1.5, the emulsion stability increased. Chitosan forms the electrostatic interactions between particles, the particles repel each other, and the emulsion is thermodynamically stable. At the high concentrations of chitosan, instability occurs due to coalescence and the flocculation of particles and creaminess increases [22]. Lements [23] reported that the emulsion stability at high concentrations of chitosan increased due to charge neutralization and bridging flocculation effects. In general, polysaccharides cause the droplets to come closer together and increase the instability and creaminess of the emulsion.

Figure 1(b) shows that in the presence of 4.5% sodium alginate, creaming increased with decreasing pH. As the pH decreases, the concentration of positive ions increases and these ions in the presence of sodium alginate cause electrostatic repulsion and increasing creaming [24].

As can be seen in Figure 1(c), at 1.5% chitosan and 4.5% sodium alginate, the creaming of emulsion was highest. But the least instability occurred when chitosan was 1% and sodium alginate was 1.5%. With increasing chitosan concentration from 0 to 1, the emulsion stability decreased and then increased with a further increase of chitosan concentration. Similar results were reported by Lements [25].

Table 1: Uncoded and coded levels of three independent variable of chitosan concentration, sodium alginate concentration, and pH used in the RSM design.

Coded independent variables levels	Uncoded values of different independent variables		
	X1: Chitosan concentration (%)	X2: Sodium alginate concentration (%)	X3: pH
$+\alpha$	1.50	4.50	7.00
1	1.19	3.58	6.18
0	0.75	2.25	5.00
−1	0.30	0.91	3.81
$-\alpha$	0.00	0.00	3.00

Table 2: Central composite experimental design for the three independent variables used in this study.

Run	X1: Chitosan concentration (%)	X2: Sodium alginate concentration (%)	X3: pH
1	0.75	2.25	5.00
2	1.19	3.58	6.18
3	0.75	2.25	5.00
4	1.19	0.91	6.18
5	0.75	2.25	5.00
6	0.75	2.25	5.00
7	0.75	0.00	5.00
8	0.75	4.50	5.00
9	1.19	3.58	3.81
10	0.30	3.58	6.18
11	1.50	2.25	5.00
12	0.75	2.25	5.00
13	0.30	3.58	3.81
14	0.30	0.91	6.18
15	1.19	0.91	3.81
16	0.75	2.25	7.00
17	0.75	2.25	5.00
18	0.75	2.25	3.00
19	0.00	2.25	5.00
20	0.30	0.91	3.81

3.3. Turbidity. The turbidity of colloidal systems depends on the size and number of particles (103). The emulsion turbidity was ranged from 0.24–0.78 nm. Three-dimensional response surface plots for emulsion turbidity are shown in Figure 2. The highest turbidity (1.3 nm) was reported at pH 7, 0% chitosan, and 4.5% sodium alginate and the lowest turbidity (0.21 nm) was reported at pH 4.3, 0.37% chitosan, and 0% sodium alginate.

Figure 2(a) shows that at pH values 3 and 7, the turbidity of the emulsion decreased by increasing the chitosan concentration and with chitosan increasing, the turbidity increased. It is due to the increase diameter of colloidal particles and their accumulation [26]. Neirynck et al. [27] confirmed that increasing the pectin-whey protein complex reduced turbidity due to emulsion stabilization, but excessive increase of these biopolymers due to their insolubility caused an increase of turbidity, which is consistent with the results of our study.

Figure 2(b) shows that at pH values 3 and 7, the turbidity of the emulsion increases with increasing concentration of sodium alginate, and the highest turbidity occurred at 4.5% sodium alginate and pH 7. Harnsilawat et al. [24] found that with the increasing alginate concentration, turbidity increased significantly due to the accumulation of biopolymers.

Figure 2(c) shows that at the lowest concentration of polysaccharides (0% sodium alginate and 0.4% chitosan) turbidity was the lowest. The highest turbidity was obtained in the presence of 4.5% sodium alginate and 0% chitosan. The high turbidity in a high levels of sodium alginate compared to chitosan can be related to the accumulation of particles or the formation of large particles in the presence of alginate.

3.4. Antioxidant Activity. The antioxidant activity of the emulsion ranged from 24 to 78%. Three-dimensional response surface plots for emulsion antioxidant activity are presented in Figure 3. The highest antioxidant properties were reported at pH value 3, 0.75% chitosan, and 3.54% alginate and the lowest antioxidant properties were reported at pH 4.6, 0.84% chitosan, and 0% sodium alginate.

With increasing pH, the antioxidant properties reduced and then increased (Figure 3(a)) because chitosan is an insoluble polysaccharide at alkaline pH with a negative charge but is soluble at acidic pH with a positive charge. At

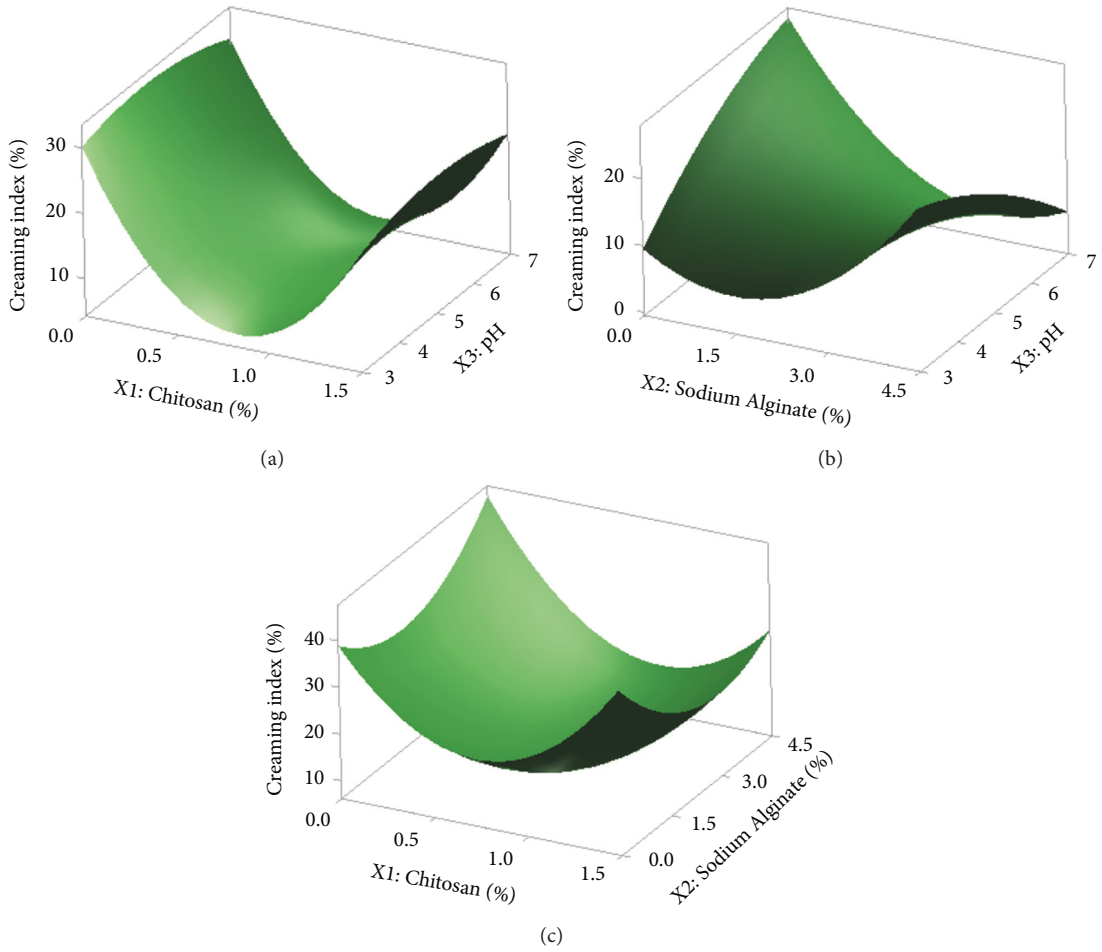


Figure 1: Three-dimensional plot of creaming of emulsion. (a) Creaming index vs. chitosan concentration. (b) Creaming vs. sodium alginate concentration. (c) Creaming vs. chitosan concentration and sodium alginate concentration.

higher pH, chitosan is insoluble and has high emulsifying properties [28, 29].

As can be seen in Figure 3(b), the highest antioxidant properties were obtained at pH 3 and 4.5% sodium alginate. Unlike chitosan, the use of this polysaccharide at lower pH increases its antioxidant properties [24]. However, the viscosity of sodium alginate is very high at low pH, its solubility is reduced, so in these conditions, it shows the highest emulsifying properties with high antioxidant activity [30].

Figure 3(c) shows the interaction of sodium alginate and chitosan. At 1.5% chitosan, with increasing the concentration of sodium alginate from 0 to 3.8%, the antioxidant properties increased, which can be attributed to high viscosity and stable emulsion [31]. With increasing the sodium alginate concentration from 3.8 to 4.5%, the antioxidant properties are constant. Sellimi et al. [30] stated that increasing the concentration of alginate were associated with increasing the antioxidant properties of the emulsion.

3.5. Microencapsulation Efficiency. The microencapsulation efficiency of balangu seed oil ranged from 77.36–90.28%. Three-dimensional response plots for microencapsulation efficiency are presented in Figure 4. According to these results, the highest microencapsulation efficiency (99.1%) was at pH 3, 1.5% chitosan, and 4.5% sodium alginate and the lowest microencapsulation efficiency (76.9%) at pH 5.4,1.2% chitosan, and 0% sodium alginate.

According to Figure 4(a), the interaction between chitosan concentration and pH shows that the highest microencapsulation efficiency is related to 1.5% chitosan with pH 3. The lowest microencapsulation efficiency was reported at pH 5. Chitosan has a high solubility in acidic conditions and bind to other polysaccharides and proteins and produces stable capsules that increase the microencapsulation efficiency, but at the alkaline pH, the positive charge of chitosan decreases, the coacervation will occur, which will reduce the microencapsulation efficiency [32].

As Figure 4(b) shows at pH equivalent to 3, the efficiency also increased with the increasing concentration of sodium alginate. Ahn et al. [33] found that increasing the concentration of wall materials increased the viscosity and the microencapsulation efficiency of sunflower oil.

Figure 4(c) shows that at 1.5% chitosan with increasing the concentration of sodium alginate, the microencapsulation efficiency increased and the highest microencapsulation

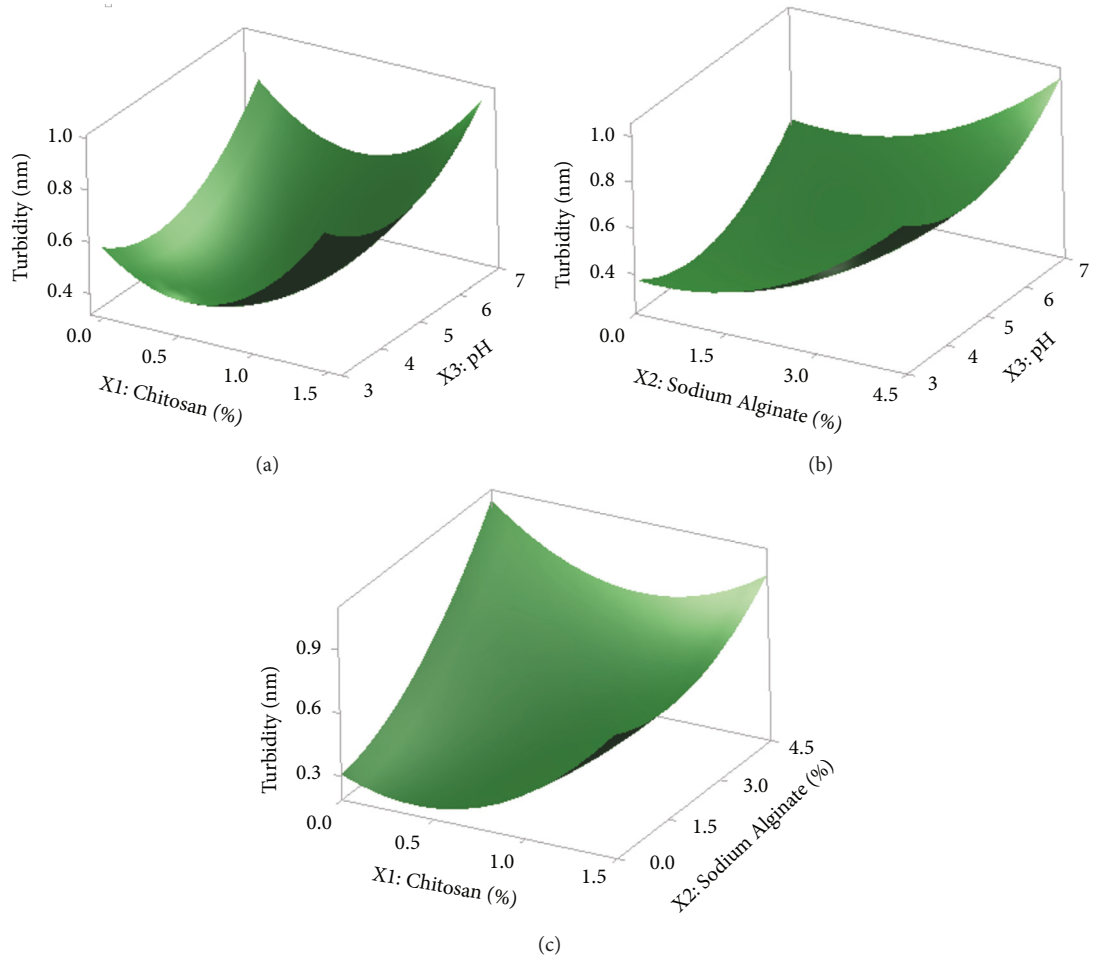


Figure 2: Three-dimensional plot of turbidity of emulsion. (a) Turbidity vs. chitosan concentration. (b) Turbidity vs. sodium alginate concentration. (c) Turbidity vs. chitosan concentration and sodium alginate concentration.

efficiency occurred at 1.5% chitosan and 4.5% sodium alginate. The lowest yield occurred at 1.2% chitosan and 0% sodium alginate, which showed the high importance of high concentrations of wall materials in increasing the efficiency of microencapsulation.

Frascareli et al. [34] observed that with increasing the concentration of the Arabic gum, the microencapsulation efficiency of coffee oil increased due to the high rate of emulsion formation and the smaller particle size of the emulsion.

3.6. Moisture Content. The moisture content of the microencapsulated balangu seed oil ranged from 2.12–4.12%. Three-dimensional response surface plots for the moisture content of the capsule are shown in Figure 5. The highest moisture content (5.88%) was observed at pH 3, 1.5% chitosan, and 4.5% sodium alginate and the lowest moisture content (1.78%) was observed at pH 4.7, 0.36% chitosan and 0.96% sodium alginate.

According to Figure 5(a), at pH 3, with increasing chitosan concentration from 0 to 1.5%, the moisture content of the capsule increased. Figure 5(b) similarly shows that the moisture content of the capsule increased with increasing sodium alginate. Increasing the concentration of wall materials increases the viscosity of the emulsion and makes it harder to water diffuse, thus increasing the humidity [34].

Figure 5(c) shows that chitosan and sodium alginate do have not a significant difference in moisture content, which is consistent with the results of Carneiro et al. [35].

3.7. Oil Release Test in Simulated Gastric (SGF) and Intestinal Environment (SIF). The release of balangu seed oil capsules ranged from 17.10–80.60%. Three-dimensional response plots for oil release rate are presented in Figure 6. The lowest release rate (15%) was observed at pH 3.33, 1.23% chitosan, and 4.36% sodium alginate. The highest release rate (91%) was observed at a pH value at 6.2, 0% chitosan, and 1.94% sodium alginate.

Figure 6(a) shows that the lowest release was related to pH 3 and 1.5% chitosan. At acidic pH, the electrostatic bond between chitosan and sodium alginate is strongest and prevents the release of contents of the capsule, but at higher pH, the release is greater.

Figure 6(b) shows that at the pH range of 3 and 7, increasing the concentration of sodium alginate caused a decrease in the release of balangu seed oil due to the

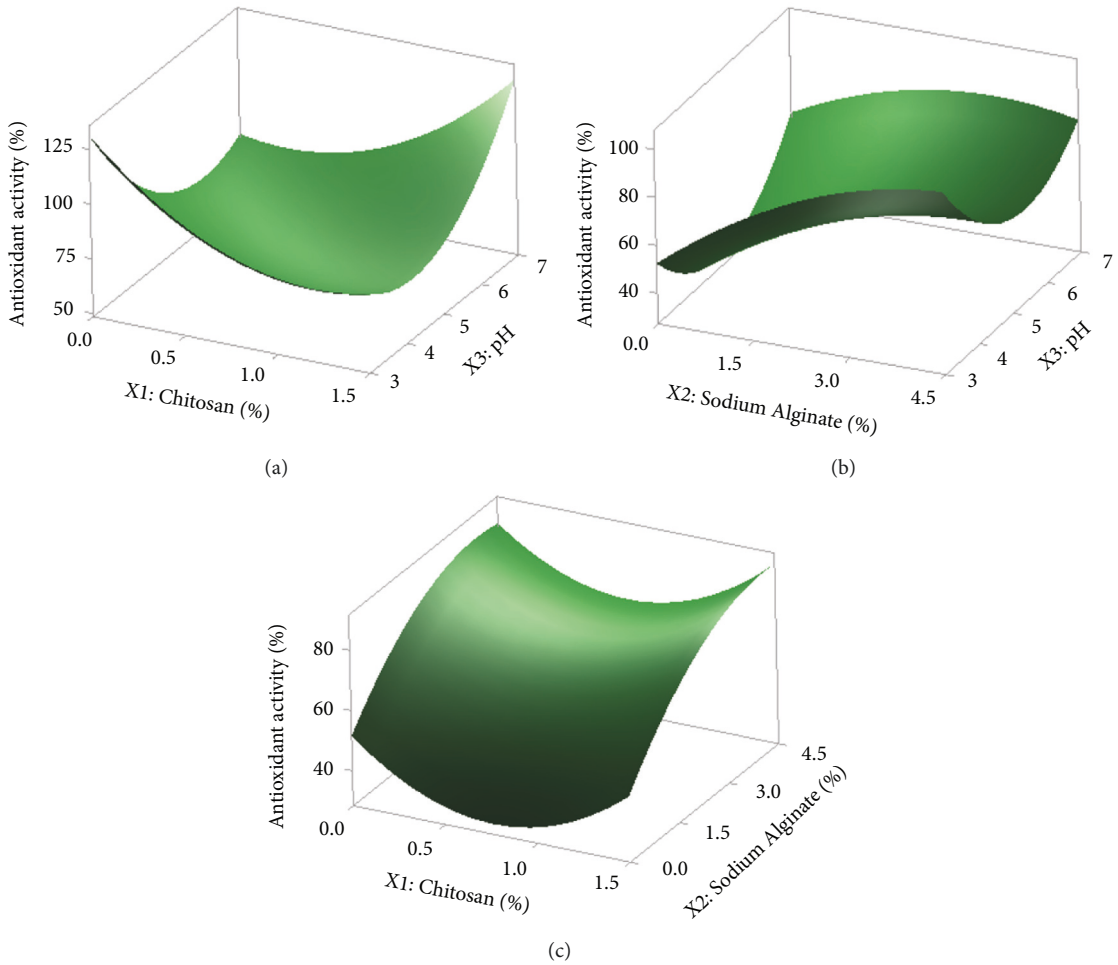


Figure 3: Three-dimensional plot of antioxidant activity of emulsion. (a) Antioxidant activity vs. chitosan concentration. (b) Antioxidant activity vs. sodium alginate concentration. (c) Antioxidant activity vs. chitosan concentration and sodium alginate concentration.

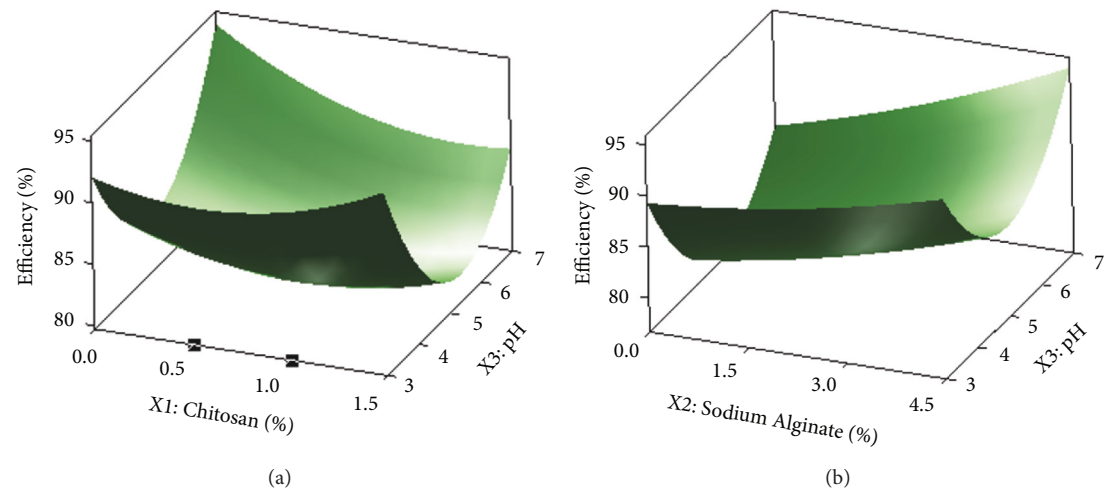
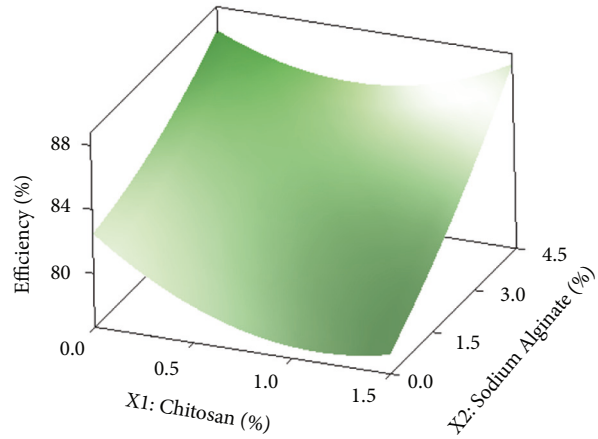
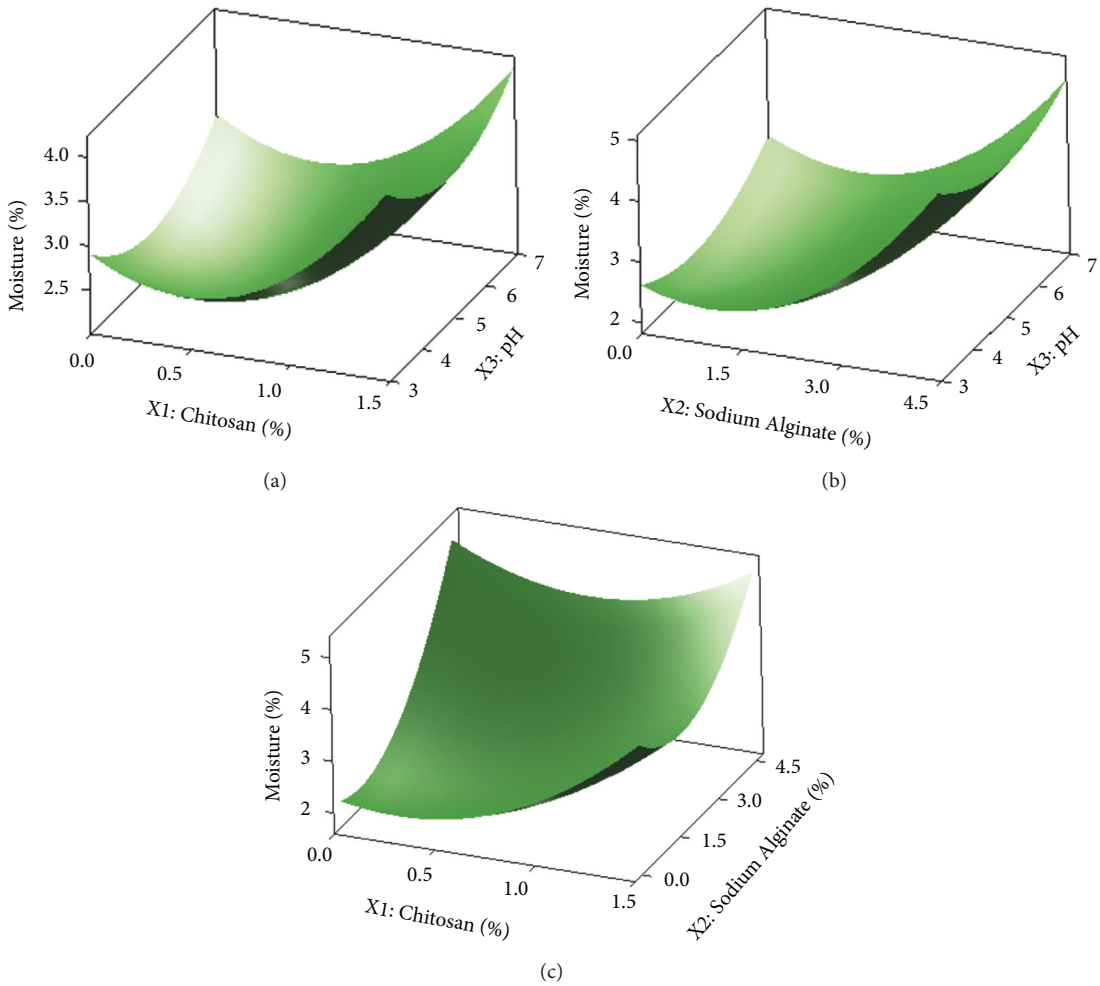


Figure 4: Continued.



(c)

Figure 4: Three-dimensional plot of microencapsulation efficiency. (a) Microencapsulation efficiency vs. chitosan concentration. (b) Microencapsulation efficiency vs. sodium alginate concentration. (c) Microencapsulation efficiency vs. chitosan concentration and sodium alginate concentration.



(a)

(b)

(c)

Figure 5: Three-dimensional plot of microencapsulate moisture content. (a) Microencapsulate moisture content vs. chitosan concentration. (b) Microencapsulate moisture content vs. sodium alginate concentration. (c) Microencapsulate moisture content vs. chitosan concentration and sodium alginate concentration.

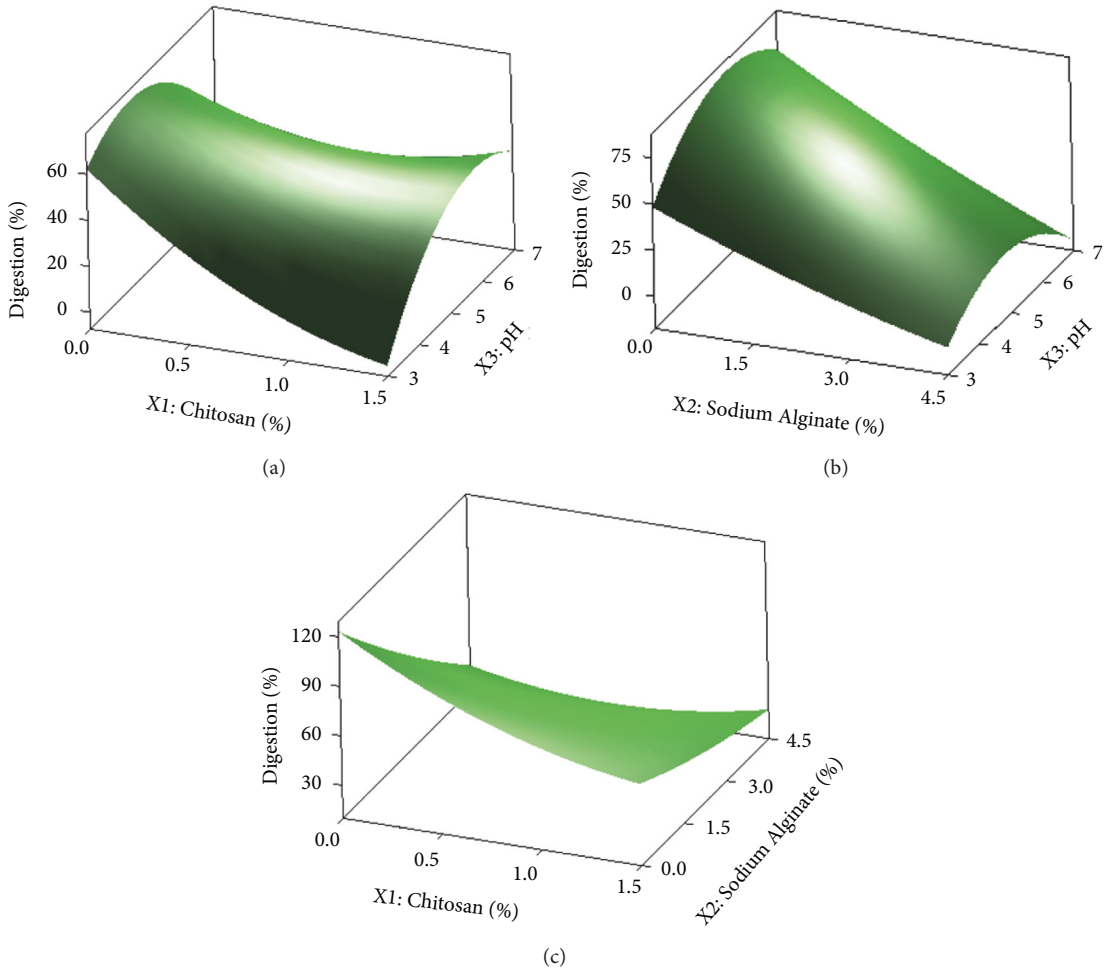


Figure 6: Three-dimensional plot of microencapsulation digestion. (a) Microencapsulation digestion vs. chitosan concentration. (b) Microencapsulation digestion vs. sodium alginate concentration. (c) Microencapsulation digestion vs. chitosan concentration and sodium alginate concentration.

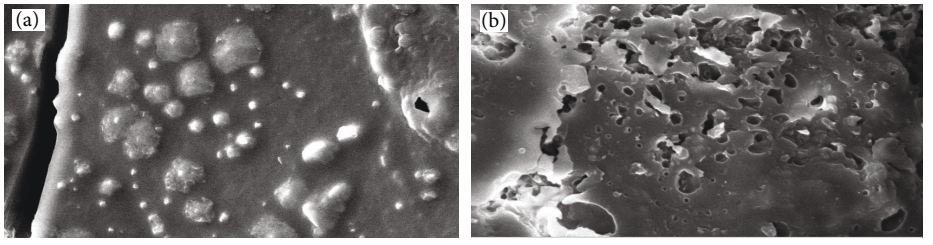


Figure 7: Scanning electron microscopy images of balangu seed powders with a magnification of 5000 ×; (a) the optimal sample (0.30% chitosan, 0.14% sodium alginate, and pH 3); (b) the worst sample including 1.5% chitosan, 0% sodium alginate, and pH 4.2.

increased rate of formation and high strength of the wall around the core, which reduces the loss of core compounds [36].

Figure 6(c) shows that the lowest oil release occurred at 4.5% sodium alginate and 0.1% chitosan. The low release of balangu seed oil at the low concentration of chitosan can be attributed to the high viscosity of chitosan [37]. Increasing the concentration of chitosan and sodium alginate increases the viscosity of the emulsion and decreases the release rate [11].

3.8. Optimization. The best sample in terms of microencapsulation efficiency included 0.30% chitosan, 0.14% sodium alginate, and pH value of 3.

3.9. Scanning Electron Microscopy (SEM). Microscopic observations showed that the structure of the optimal sample was smooth, spherical, and regular, and these data confirmed the successful microencapsulation (Figure 7). The outer surface of the particles also showed that the optimal

sample has different sizes and without cracking, which causes reduced oil release and increased material retention. It should be noted that despite some accumulation of particles, these can open over time and the particle size decreases [38].

The treatment with the lowest microencapsulation efficiency did not have a definite shape and had large pores and cracks. Because the microencapsulation was not done properly, the particles had a high dispersion.

4. Conclusion

Microencapsulation of balangu (*Lallemantia royleana*) seed oil extracted by the ultrasound-incubation method in polysaccharide and protein matrices indicated that this process has high efficiency, low release, good moisture, and high antioxidant properties and stability and good transparency of the emulsion showed that this process can be an efficient method to protect oil extracted from unsuitable conditions and it can be easily used in water-based foods. The structure of the optimal sample (0.30% chitosan, 0.14% sodium alginate, and pH 3) confirmed the success of emulsification and microencapsulation processes.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

References

[1] M. Z. Kyari, "Extraction and characterization of seed oils," *International Agrophysics*, vol. 22, no. 2, p. 139, 2008.

[2] K. Morteza-Semnani, "Essential oil composition of lallemantia iberica fisch. et C.A. Mey," *Journal of Essential Oil Research*, vol. 18, no. 2, pp. 164-165, 2006.

[3] M. Zlatanov, G. Antova, M. Angelova-Romova, S. Momchilova, S. Taneva, and B. Nikolova-Damyanova, "Lipid structure of *lallemantia* seed oil: a potential source of omega-3 and omega-6 fatty acids for nutritional supplements," *Journal of the American Oil Chemists Society*, vol. 89, no. 8, pp. 1393–1401, 2012.

[4] C. Da Porto, E. Porretto, and D. Decorti, "Comparison of ultrasound-assisted extraction with conventional extraction methods of oil and polyphenols from grape (*Vitis vinifera* L.) seeds," *Ultrasonics Sonochemistry*, vol. 20, no. 4, pp. 1076–1080, 2013.

[5] A. P. Schwab, J. Su, S. Wetzel, S. Pekarek, and M. K. Banks, "Extraction of petroleum hydrocarbons from soil by mechanical shaking," *Environmental Science & Technology*, vol. 33, no. 11, pp. 1940–1945, 1999.

[6] S. M. B. Hashemi, J. Michiels, S. H. Asadi Yousefabad, and M. Hosseini, "Kolkhoung (*Pistacia khinjuk*) kernel oil quality is affected by different parameters in pulsed ultrasound-assisted solvent extraction," *Industrial Crops and Products*, vol. 70, pp. 28–33, 2015.

[7] S. Samaram, H. Mirhosseini, C. P. Tan, and H. M. Ghazali, "Ultrasound-assisted extraction and solvent extraction of papaya seed oil: crystallization and thermal behavior, saturation degree, color and oxidative stability," *Industrial Crops and Products*, vol. 52, pp. 702–708, 2014.

[8] Y. Tian, Z. Xu, B. Zheng, and Y. Martin Lo, "Optimization of ultrasonic-assisted extraction of pomegranate (*Punica granatum* L.) seed oil," *Ultrasonics Sonochemistry*, vol. 20, no. 1, pp. 202–208, 2013.

[9] Z. Xiao, W. Liu, G. Zhu, R. Zhou, and Y. Niu, "A review of the preparation and application of flavour and essential oils microcapsules based on complex coacervation technology," *Journal of the Science of Food and Agriculture*, vol. 94, no. 8, pp. 1482–1494, 2014.

[10] H. D. Silva, M. Â. Cerqueira, and A. A. Vicente, "Nanoemulsions for food applications: development and characterization," *Food and Bioprocess Technology*, vol. 5, no. 3, pp. 854–867, 2012.

[11] S. M. Jafari, E. Assadpoor, B. Bhandari, and Y. He, "Nanoparticle encapsulation of fish oil by spray drying," *Food Research International*, vol. 41, no. 2, pp. 172–183, 2008.

[12] S. Quispe-Condori, M. D. A. Saldaña, and F. Temelli, "Microencapsulation of flax oil with zein using spray and freeze drying," *LWT—Food Science and Technology*, vol. 44, no. 9, pp. 1880–1887, 2011.

[13] A. Mohammadi, S. M. Jafari, A. F. Esfanjani, and S. Akhavan, "Application of nano-encapsulated olive leaf extract in controlling the oxidative stability of soybean oil," *Food Chemistry*, vol. 190, pp. 513–519, 2016.

[14] E. Assadpour, Y. Maghsoudlou, S. M. Jafari, M. Ghorbani, and M. Aalami, "Optimization of folic acid nano-emulsification and encapsulation by maltodextrin-whey protein double emulsions," *International Journal of Biological Macromolecules*, vol. 86, pp. 197–207, 2016.

[15] P. Izadi Najaf Abadi and S. Taghian Dinani, "Optimization of ultrasonic-shaking incubation extraction of oil from balangu (*lallemantia royleana*) seeds using response surface methodology," *RAdvFoodSci*, vol. 3, no. 2, pp. 299–322, 2020.

[16] Z. Zhang, R. Zhang, L. Zou, and D. J. McClements, "Protein encapsulation in alginate hydrogel beads: effect of pH on microgel stability, protein retention and protein release," *Food Hydrocolloids*, vol. 58, pp. 308–315, 2016.

[17] M.-G. Song, S.-H. Jho, J.-Y. Kim, and J.-D. Kim, "Rapid evaluation of water-in-oil (w/o) emulsion stability by turbidity ratio measurements," *Journal of Colloid and Interface Science*, vol. 230, no. 1, pp. 213–215, 2000.

[18] T. P. Sari, B. Mann, R. Kumar et al., "Preparation and characterization of nanoemulsion encapsulating curcumin," *Food Hydrocolloids*, vol. 43, 2015.

[19] T. Sathasivam, S. Muniyandy, L. H. Chuah, and P. Janarthanan, "Encapsulation of red palm oil in carboxymethyl sago cellulose beads by emulsification and vibration technology: physicochemical characterization and in vitro digestion," *Journal of Food Engineering*, vol. 231, pp. 10–21, 2018.

[20] C. I. Beristain, E. Azuara, and E. J. Vernon-Carter, "Effect of water activity on the stability to oxidation of spray—dried encapsulated orange peel oil using mesquite gum (*Prosopis juliflora*) as wall material," *Food Science*, vol. 67, no. 1, pp. 1–7, 2006.

[21] S. Wang, Y. Shi, and L. Han, "Development and evaluation of microencapsulated peony seed oil prepared by spray drying: oxidative stability and its release behavior during in-vitro digestion," *Journal of Food Engineering*, vol. 231, pp. 1–9, 2018.

[22] E. Dickinson and D. Eric, "Hydrocolloids as emulsifiers and emulsion stabilizers," *Food Hydrocolloids*, vol. 23, no. 6, pp. 1473–1482, 2009.

[23] S. Ogawa, E. A. Decker, and D. J. M. Clements, *Production and Characterization of O/W Emulsions Containing Cationic Droplets Stabilized by Lecithin – Chitosan Membranes*, ACS, Washington, DC, USA, 2003.

[24] T. Harnsilawat, R. Pongsawatmanit, and D. McClements, "Characterization of β-lactoglobulin-sodium alginate interactions in aqueous solutions: a calorimetry, light scattering, electrophoretic mobility and solubility study," *Food Hydrocolloids*, vol. 20, no. 5, pp. 577–585, 2006.

[25] Y. Suk Gu, E. A. Decker, and D. J. McClements, *Influence of pH and ι -Carrageenan Concentration on Physicochemical Properties and Stability of -Lactoglobulin-Stabilized Oil-In-Water Emulsions*, ACS Publications, Washington, DC, USA, 2004.

[26] T. Aoki, E. A. Decker, and D. J. McClements, "Influence of environmental stresses on stability of O/W emulsions containing droplets stabilized by multilayered membranes produced by a layer-by-layer electrostatic deposition technique," *Food Hydrocolloids*, vol. 19, no. 2, pp. 209–220, 2005.

[27] N. Neirynck, P. Van der Meeren, M. Lukaszewicz-lausecker, J. Cocquyt, D. Verbeken, and K. Dewettinck, "Influence of pH and biopolymer ratio on whey protein-pectin interactions in aqueous solutions and in O/W emulsions," *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 298, no. 1-2, pp. 99–107, 2007.

[28] Q. Gan, T. Wang, C. Cochrane, and P. McCarron, "Modulation of surface charge, particle size and morphological properties of chitosan-TPP nanoparticles intended for gene delivery," *Colloids and Surfaces, B: Biointerfaces*, vol. 44, no. 2-3, pp. 65–73, 2005.

[29] Z. Wei, C. Wang, S. Zou, H. Liu, and Z. Tong, "Chitosan nanoparticles as particular emulsifier for preparation of novel pH-responsive Pickering emulsions and PLGA microcapsules," *Polymer*, vol. 53, no. 6, pp. 1229–1235, 2012.

[30] S. Sellimi, I. Younes, H. B. Ayed et al., "Structural, physicochemical and antioxidant properties of sodium alginate isolated from a Tunisian brown seaweed," *International Journal of Biological Macromolecules*, vol. 72, pp. 1358–1367, 2015.

[31] M. Nakauma, T. Funami, S. Noda et al., "Comparison of sugar beet pectin, soybean soluble polysaccharide, and gum Arabic as food emulsifiers. 1. Effect of concentration, pH, and salts on the emulsifying properties," *Food Hydrocolloids*, vol. 22, no. 7, pp. 1254–1267, 2008.

[32] S. A. Agnihotri, N. N. Mallikarjuna, and T. M. Aminabhavi, "Recent advances on chitosan-based micro- and nanoparticles in drug delivery," *Journal of Controlled Release*, vol. 100, no. 1, pp. 5–28, 2004.

[33] J.-H. Ahn, Y.-P. Kim, Y.-M. Lee, E.-M. Seo, K.-W. Lee, and H.-S. Kim, "Optimization of microencapsulation of seed oil by response surface methodology," *Food Chemistry*, vol. 107, no. 1, pp. 98–105, 2008.

[34] E. C. Frascareli, V. M. Silva, R. V. Tonon, and M. D. Hubinger, "Effect of process conditions on the microencapsulation of coffee oil by spray drying," *Food and Bioproducts Processing*, vol. 90, no. 3, pp. 413–424, 2012.

[35] H. C. F. Carneiro, R. V. Tonon, C. R. F. Grosso, and M. D. Hubinger, "Encapsulation efficiency and oxidative stability of flaxseed oil microencapsulated by spray drying using different combinations of wall materials," *Journal of Food Engineering*, vol. 115, no. 4, pp. 443–451, 2013.

[36] W.-C. Hsieh, C.-P. Chang, and Y.-L. Gao, "Controlled release properties of chitosan encapsulated volatile citronella oil microcapsules by thermal treatments," *Colloids and Surfaces B: Biointerfaces*, vol. 53, no. 2, pp. 209–214, 2006.

[37] C. Peniche, I. Howland, O. Carrillo, C. Zaldívar, and W. Arguelles Monal, "Formation and stability of shark liver oil loaded chitosan/calcium alginate capsules," *Food Hydrocolloids*, vol. 18, pp. 285–287, 2004.

[38] A. Mohammadi, M. Hashemi, and S. M. Hosseini, "This study was undertaken to investigate the nanoencapsulation of *Zataria* SC," *Innovative Food Science & Emerging Technologies*, vol. 23, pp. 73–80, 2014.