

PARTIAL HYDROPHOBIC MODIFICATION OF ALGINATE USING DODECENYL SUCCINIC ANHYDRIDE FOR FISH OIL ENCAPSULATION

Dyah Hesti Wardhani, Hana Nikma Ulya✉, Andri Cahyo Kumoro, Nita Aryanti

Department of Chemical Engineering, Faculty of Engineering, Universitas Diponegoro
Tembalang, Semarang 50239, Indonesia

ABSTRACT

Background. As a hydrophilic compound, alginate needs to be modified to perform both as an emulsifier and matrix for fish oil encapsulation. Dodecyl succinic anhydride (DSA) was selected for the modification due to its mild reaction conditions and the fact that no side product is generated. The purposes of this research were to prepare partial hydrophobic groups of modified alginate, which were subsequently used for a fish oil encapsulant.

Material and methods. The viscosity average molecular weight (Mv) of commercial alginate was adjusted using sonication. The degraded alginate was modified using DSA by mixing alginates in the desired ratio for 4 h at pH = 10. The fish oil emulsion was stabilized using the modified alginate and subsequently freeze-dried to obtain fish oil particles. The effect of the Mv of alginate and the alginate-DSA ratio was analyzed for emulsion stability. The performance of native and modified alginate in fish oil encapsulation were also compared.

Results. Alginate modification using DSA was influenced by the Mv of alginate as described by degree of substitution. This modification reduced the fish oil oxidation degree up to 8%. The modified alginate using 400 kg/mol and a mol ratio of 1:4 emulsion prevented separation for up to 18 h. The dynamic second-order model was well fitted to represent the separation emulsion ($R^2 > 0.89$). The fish oil encapsulation using the modified alginate showed higher loading capacity, and better anti-rehydration properties than the native one. The use of modified alginate also reduced the degree of oxidation <4% after 24 h.

Conclusion. Modification using DSA improved the alginate capability to stabilize the emulsion and encapsulating fish oil.

Keywords: Partial hydrophobic alginate, fish oil encapsulation, alginate oil affinity modification

INTRODUCTION

Omega-3, listed as a polyunsaturated fatty acid (PUFA), is mainly composed of alpha-linolenic acid (ALA), eicosapentaenoic acid (EPA), and docosahexaenoic acid (DHA). Omega-3 intake prevents hypertension, coronary diseases, diabetics, cancer, and other autoimmune diseases (Tur et al., 2012). Since most omega-3 sources were fish oils, their direct consumption was not preferable because of its flavor and taste. Omega-3 fortification also had the disadvantage of unstable properties,

which affected the food taste and caused carcinogenesis and pathogenesis of Alzheimer's (Kaushik et al., 2015). To protect and preserve the omega-3, encapsulation using a suitable matrix is needed.

In general, encapsulation consists of two main processes, i.e., precursor preparation and particle production (Kaushik et al., 2015). Oil-based omega-3 needed to be emulsified to form droplets in a water-based matrix solution, followed by solvent evaporation to

✉hananikmaa@gmail.com, phone 6224 7460058

produce the solids of encapsulated omega-3. Stable emulsion, which was influenced by the droplet size and viscosity, prevented the volatiles released and exhibited lower surface oil (Tonon et al., 2012).

Alginate has been widely used as a matrix for various active compounds, including lipophilic substances. Su et al. (2018) studied the alginate-soybean oil emulsion that remained stable in 2°C storage. However, the low temperature stabilized the emulsion by preventing droplet coalescence as the continuous phase became more viscous (Souza et al., 2015). Since alginate is a hydrophilic compound, a modification is necessary for application in encapsulating lipophilic compounds. Modified alginate for an emulsifier and encapsulant was reported by Falkeborg and Guo (2015), who reacted alginate with dodecenyl succinic anhydride (DSA). The use of cyclic anhydride carboxylic acid in hydrophobic group modification was preferable, as no extreme condition was needed for the reaction. In addition, this method did not produce organic acid as a side product (Thakur, 2014).

As mentioned before, oil-based omega-3 is susceptible to oxidation in atmospheric conditions. Therefore, freeze-drying is a more suitable method for omega-3 encapsulation, since it uses a low temperature and minimizes contact between the sample with the environment (Pang et al., 2017). Moreover, Anwar and Kunz (2011) found a higher oxidation rate occurred in encapsulated fish oil produced from a spray dryer than a freeze dryer. Integrated studies of alginate modification that is subsequently applied for encapsulation of fish oil using the freeze-drying method are still rare.

Since the alginate was extracted from natural sources, it had various molecular weights. The high molecular weight of alginate hindered its solubility, which inhibited its ionic reaction with the modifier compound (Miller-Chou and Koenig, 2003). This paper aims to determine the effect of alginate molecular weight and the ratio of alginate-DSA on the modified alginate properties. The use of modified alginate in emulsifying and encapsulating fish oil was further studied by comparing it with the native alginate.

MATERIALS AND METHODS

Materials

Sodium alginate and fish oil in food grade specification were bought from a local store (PT Multi Kimia

Raya, Semarang, Central Java, Indonesia), while DSA was obtained from Sigma-Aldrich (St. Louis, Missouri, US). Ethanol, iron (II) sulphate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), trichloroacetic acid (TCA), thiobarbituric acid (TBA), and other chemicals were analytical grade (Merck Chemical Co., Darmstadt, Hesse, Germany).

Alginate modifications

Sodium alginate was degraded using the ultrasonication process in a constant-stirred heterogeneous system to modify its molecular weight. Sodium alginate (70 g) was dispersed in 96% ethanol (150 ml) and was ultrasonicated using a KW 1801033 ultrasonic cleaner at 40 Hz (PT. Krisbow Indonesia, Jakarta, Indonesia). After the desired molecular weight was obtained, the sodium alginate was filtered and dried at ambient temperature.

Sodium alginate with different Mv (400, 100, and 40 kg/mol) was modified using the method described by Falkeborg and Guo (2015), with slight modifications. A homogeneous alginate solution was made by dissolving 1 g of sodium alginate in 100 ml distilled water with constant stirring. DSA was added in various ratios of alginate-DSA (1:1, 1:2, and 1:4, g/ml). The variable of modifications is presented in Table 1. The solution was maintained at pH 10 during the modification using NaOH (3%, w/v). After 4 h of reaction,

Table 1. Variables of modified alginate

Code	Viscosity average molecular weight (Mv) kg/mol	Alginate:DSA ratio (w/w)
400(1:1)	400	1:1
400(1:2)	400	1:2
400(1:4)	400	1:4
100(1:1)	100	1:1
100(1:2)	100	1:2
100(1:4)	100	1:4
40(1:1)	40	1:1
40(1:2)	40	1:2
40(1:4)	40	1:4

the mixture was precipitated in 150 ml 96% ethanol. The precipitate was separated and dried at room temperature.

Degree of substitution

The degree of substitution of alginate was determined using the method described by Zainal Abiddin et al. (2018). NaOH solution (0.5 M, 25 ml) was added to the modified alginate (0.5 g) suspension in distilled water (10 ml). The sample was shaken in an orbital shaker at room temperature for 24 h. The unreacted NaOH was titrated by a HCl solution (0.5 M) with a phenolphthalein indicator. The degree of substitution (DS) was calculated using Equations (1) and (2).

$$\% \text{substitution} = \frac{(V_b - V_s) \times 0.1 \times M \times 100}{W} \quad (1)$$

$$\text{DS} = \frac{162 \times \% \text{substitution}}{23\,800 - 237 \times \% \text{substitution}} \quad (2)$$

where:

- V_b – HCl volume for native alginate titration, ml,
- V_s – HCl volume for modified alginate titration, ml,
- M – HCl molarity,
- W – sample weight, g.

Fish oil emulsification and encapsulation

Emulsions of fish oil were prepared by adding fish oil (0.1 ml) to 10 ml of the modified alginate solution (1% w/v) and were homogenized using IKA RW homogenizer (IKA, Staufenim Breisgau, Germany) at 25,600 rpm for 2.5 min at room temperature. The emulsion of fish oil in native alginate was also prepared for comparison. To obtain fish oil particles, fish oil emulsion was initially frozen at -10°C for 48 h and freeze-dried at -50°C for 72 h (Innotech BK-FD10S, Inno Tech GmbH, Bochum, Germany).

Emulsion stabilization

The stabilization of the alginate-fish oil emulsion was analyzed visually based on the phase separation described by Huang et al. (2001). The emulsion was placed in test tubes and measured for the cream-serum separation over time and calculated for %separation using Equation (3).

$$\% \text{separation} = \frac{h_{\text{serum}}}{h_{\text{total}}} \times 100\% \quad (3)$$

The phase separation of the emulsions was modelled using a dynamic second-order model used by Cardoso-Ugarte et al. (2018), as described by Equation (4).

$$y(t) = k[1 - (1 + k_1(t - d))e^{-k_1 t}] \quad (4)$$

where:

- $y(t)$ – phase separation at t day, %,
- k – maximum phase separation,
- d – emulsion stable duration, day,
- k_1 – phase separation constant.

Zeta potential

The Zeta potential of the emulsions was analyzed using a Horiba SZ-100 (Horiba Ltd., Kyoto, Japan) with a scattering angle of 90° . The temperature of the holder was 24.8°C , the dispersion medium viscosity was 0.896 cps, and the electrode voltage was 3.3 kV.

Optical microscopy

The fresh emulsion was observed using a binocular microscope (Sinher XSZ-107BN, Ningbo Beilun Fangyuan Photoelectric Ltd., Zhejiang, China) at $40\times$ objective lens magnification. The droplets were captured using a digital camera connected to a PC.

Fish oil oxidation analysis

The oxidation degree of the fish oil was determined using the method used by Falkeborg and Guo (2015). The pre-emulsion solution (50 μl) was prepared by dispersed linoleic acid (2% v/v) and tween 20 (1% v/v) in an alginate solution (25 mg/ml, 1.5 ml). Iron (II) sulphate solution (25 M, 250 μl) was added before the mixture was incubated at room temperature for 15 min. The oxidation product was detected by adding TCA (2.5% v/v, 0.5 ml) and TBA (0.7% in 0.05 M KOH, 1 ml), followed with incubating the mixture in a waterbath at 100°C for 15 min. After being cooled at room temperature, the emulsion was centrifuged at 4,000 rpm for 3 min (EBA 270, Hettich, Tuttlingen, Germany). The absorbance of the supernatant was read at 532 nm using a spectrophotometer (T6 UV/VIS spectrophotometer, Pgeneral Instrument Plant, Beijing, China). The oxidation degree was calculated using Equation (5), where

A_s and A_0 are the absorbance of alginate-stabilized emulsion and fish oil emulsion, respectively.

$$\text{Oxidation degree, \%} = \frac{A_s}{A_0} \times 100\% \quad (5)$$

Scavenging activity of 2,2-diphenyl-1-picrylhydrazyl (DPPH)

DPPH scavenging activity was analyzed using the method described by Sun et al. (2015) by incubating alginate solution (1%, 1 ml) in a freshly prepared DPPH solution (in 95% ethanol, 4 ml). The solution was incubated in an orbital shaker at room temperature with minimum light. The absorbance was measured at initial and after 30 min incubation using a visible spectrophotometer set at 517 nm.

Fish oil particle properties

Loading capacity (LC) of fish oil particles was determined based on Strobel et al. (2016). The freeze-dried alginate-fish oil (1 g) was dissolved in sodium citrate solution (100 mM, 30 ml). N-hexane and propan-2-ol, 5 ml of each, were added to the solution. After centrifuging at 4000 rpm for 5 min, the organic phase was decanted and vaporized. The remaining oil was weighed as the mass of encapsulated oil used for loading capacity calculation (Equation (6)).

$$\text{Loading capacity, \%} = \frac{m_{\text{oil}}}{m_{\text{sample}}} \times 100\% \quad (6)$$

The oxidative stability of encapsulated fish oil in native and modified alginate solids was analyzed based on the method described by Falkeborg and Guo (2015) with a slight modification. The encapsulated sample (20 mg) was dissolved in sodium citrate solution (100 mM, 10 ml). Two milliliters of the solution were added by TCA (2.5%, 0.5 ml) and TBA (0.7% in 0.05 M of KOH solution, 1 ml). The solution was incubated in a waterbath for 15 min at 100°C. After being cooled to room temperature, the absorbance of the solution was measured using a visible spectrophotometer at 532 nm. The oxidation degree was calculated using Equation (5) with A_s and A_0 are the absorbance of the sample and the absorbance of the mixture of

linoleic acid (20 mg) and iron (II) sulphate solution (25 mM, 250 µl), respectively.

The moisture content of alginate-fish oil solid (0.5 g) was calculated after being oven-dried at 105°C until constant weight. The moisture content (MC) was calculated using Equation (7).

$$\text{MC, \%} = \frac{W_{\text{sample}} - W_{\text{driedsample}}}{W_{\text{sample}}} \times 100\% \quad (7)$$

RESULTS AND DISCUSSIONS

Alginate modification

Alginate with various Mv, the product of ultrasonication, was reacted with DSA in different ratios of alginate-DSA. The substitution degree of the modified alginates is shown in Figure 1a. The highest degree of substitution was obtained by a variable of 40 (1:4) with a value of 0.0896. A lower Mv of the alginate eased the substitution process and increased the degree of substitution after a 4-h reaction. A high Mv was a disadvantage for the modification process. As the effect of high Mv alginate dissolution, viscous alginate solution hindered the interaction between alginate and DSA, which reduced the reaction probability. Lowering the Mv of the alginate using the ultrasonication method could reduce the viscosity of the alginate solution and increase the degree of substitution of alginate-DSA. A higher concentration of DSA also increased the substitution degree, which was also found by Falkeborg et al. (2015). However, the reaction could not completely replace the hydroxyl groups of alginate with long-chain DSA groups because of the steric hindrance of the larger structure of modified alginate.

The ability of native and modified alginate to prevent oxidation was compared by detecting the propanal using a spectrophotometer. Propanal is a secondary product of oil oxidation produced after the addition of iron (II) sulphate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$). Figure 1b shows that the modified alginate reduced the oxidation degree by up to 8%. The antioxidant activity of the modified alginate was influenced by the presence of electronegative hydroxyl groups. The activity increased with the more hydroxyl groups of DSA integrated into the alginate, which improved the radical scavenging ability

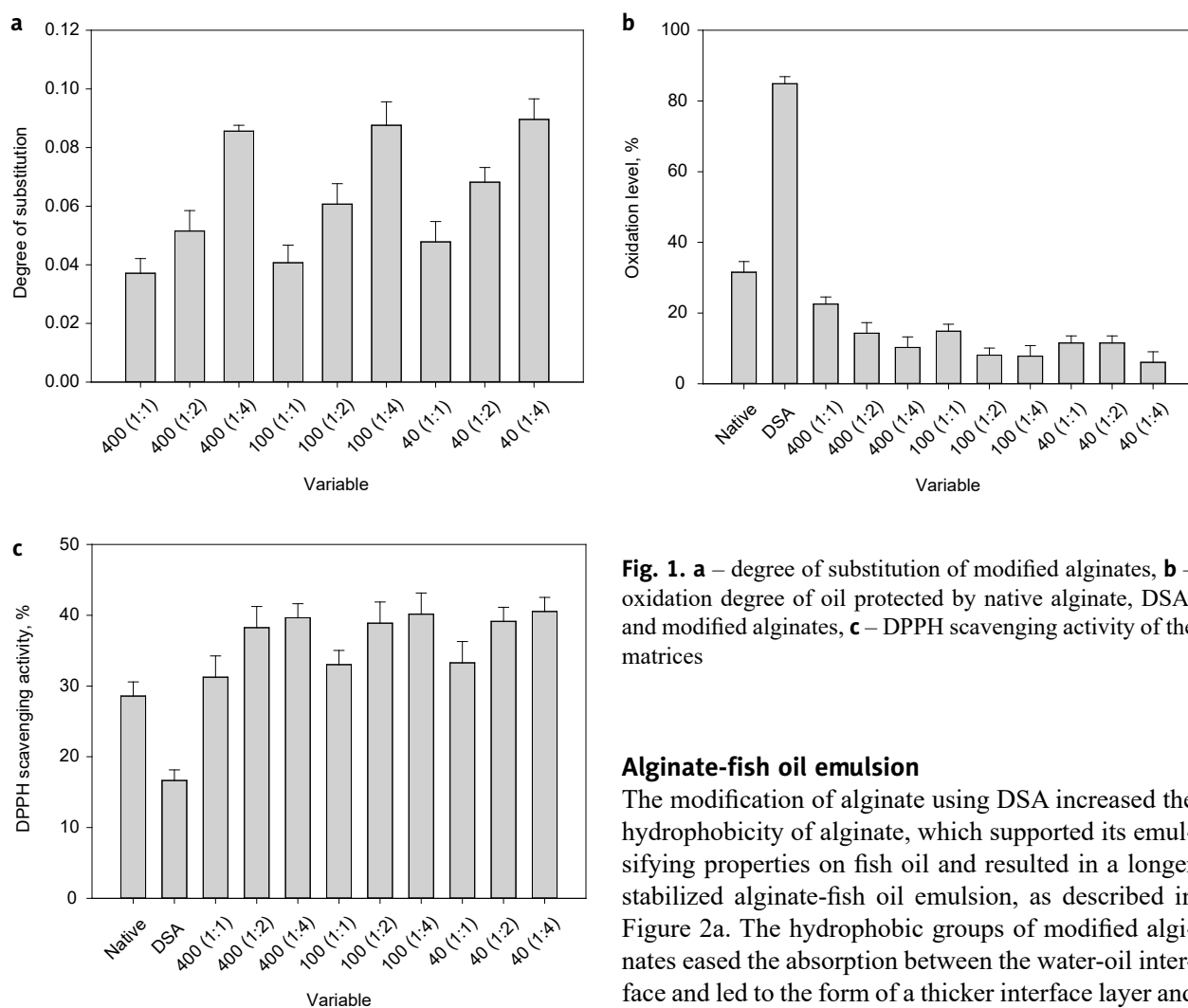


Fig. 1. **a** – degree of substitution of modified alginates, **b** – oxidation degree of oil protected by native alginate, DSA, and modified alginates, **c** – DPPH scavenging activity of the matrices

Alginate-fish oil emulsion

The modification of alginate using DSA increased the hydrophobicity of alginate, which supported its emulsifying properties on fish oil and resulted in a longer stabilized alginate-fish oil emulsion, as described in Figure 2a. The hydrophobic groups of modified alginates eased the absorption between the water-oil interface and led to the form of a thicker interface layer and increased the steric hindrance among the oil droplets in the emulsion (Lin et al., 2018).

Zeta potential is defined as a potential difference between the dispersion medium and the stationary layer of fluid surrounding the particle. A higher value of zeta potential indicates greater emulsion stability due to the repulsion of each charged particle which avoids aggregation (Barba et al., 2019). The zeta value of the emulsion droplet became more negative after alginate modification from -42.50 to -78.3 mV (Fig. 2b, 2d), which resulted in the smaller droplet formation (Fig. 2c, 2e). Moreover, the use of modified alginate also prevented droplet aggregation and produced more stable emulsions, as explained by Goodarzi and Zendejboudi (2019). The zeta potential value over ± 30 mV was claimed to provide repulsive forces between oil

(Falkeborg and Guo, 2015). However, the addition of native DSA directly to the oil emulsion will not maintain the emulsion form even after homogenizer treatment. DSA had strong hydrophobic properties, which was not soluble in water, particularly when the succinic ring was not completely opened (Wang et al., 2020).

The ability of alginate to protect against oxidation was also confirmed by the DPPH scavenging activity shown in Figure 1c. The modified alginate of the 40 (1:4) sample showed the highest inhibition activity (40.53%). The more hydroxyl groups in the modified alginate, the more DPPH radicals neutralized with the hydroxyl using an electron transfer mechanism and increased the inhibition activity (Škrovánková et al., 2012).

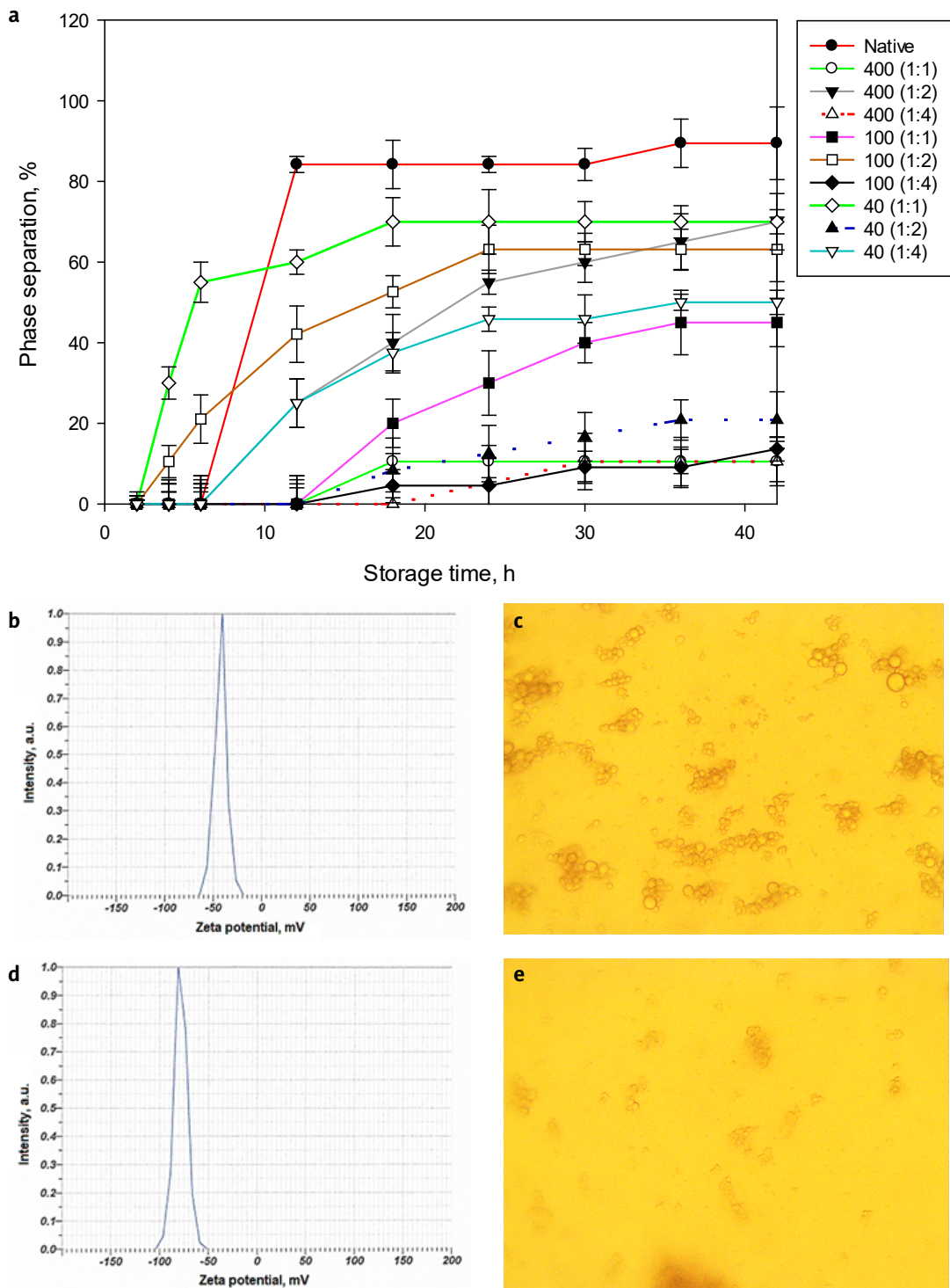


Fig. 2. a – phase separation of alginate-fish oil emulsion stabilized using native and modified alginate, b, c – zeta potential and optical micrograph of fish oil emulsions stabilized using native, d, e – zeta potential and optical micrograph of fish oil emulsions stabilized using modified alginate

droplets and physical stability enhancement in colloid suspension that prevented particle aggregation (Falkeborg and Guo, 2015).

Carboxylate groups of the native alginate structure contribute to negative surface charges in the native emulsion. In addition, DSA also contained carboxyl groups that led to more anionic charged particles, which contributed to the increase in the steric hindrance of the alginate after modification (Falkeborg and Guo, 2015). Carboxyl group addition from the hydrophobic chain of DSA exhibited a more negative charge, hence stabilizing the emulsion (Melanie et al., 2020). Moreover, the half-ester formation also contributed to the negative charge of the modified alginate (Kokubun et al., 2015). A similar result was reported by Pan et al. (2019) on modified dextrin using octenyl succinic anhydride that could stabilize the emulsion of dextrin-curcumin.

Figure 3 shows the creaming of emulsion at various alginate molecular weights and the ratio of

DSA-alginate. None of the emulsions were physically stable after the 42-h storage time. The creaming results, which are a visual description of the separation phase that occurred in the final incubation time, are presented in Figure 2a. This instability led to phase separation, where the oil droplets coalesced with each other and formed a larger one. Due to the oil's lower density than the surrounding liquid, the bigger droplets moved upwards to produce a creaming layer on top of the emulsions by the gravity influence (Falkeborg and Guo, 2015). Meanwhile, a transparent serum layer was formed at the bottom of the container because it contained no droplets that scatter light (Yang et al., 2012).

Although DSA-modified alginate increased the stability of the emulsion compared to the native, the most stable emulsion was obtained by the emulsion of 400(1:4) (Fig. 3). This could be due to the higher molecular weight of alginate leading to the higher viscosity of the alginate solution, which could help to stabilize

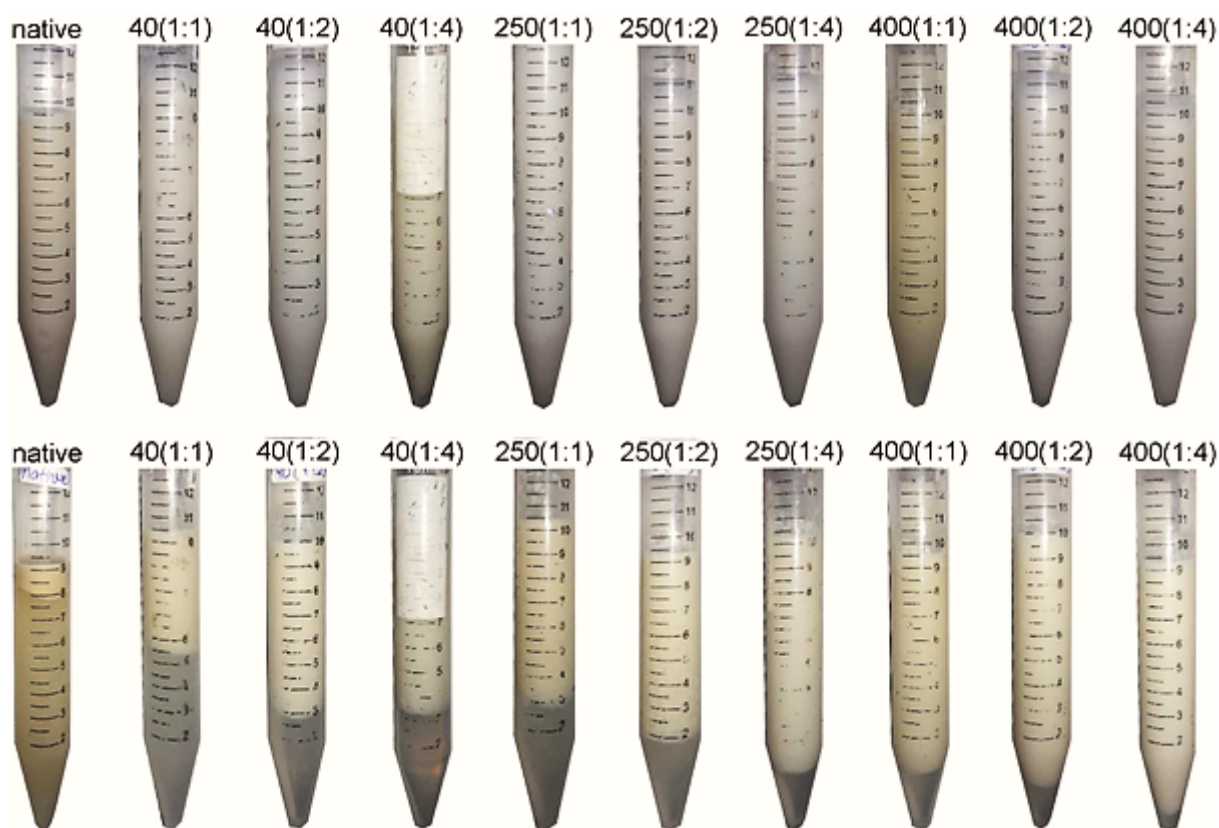


Fig. 3. Visual appearance of fish oil emulsions at initial storage (top) and after 42 h storage (bottom)

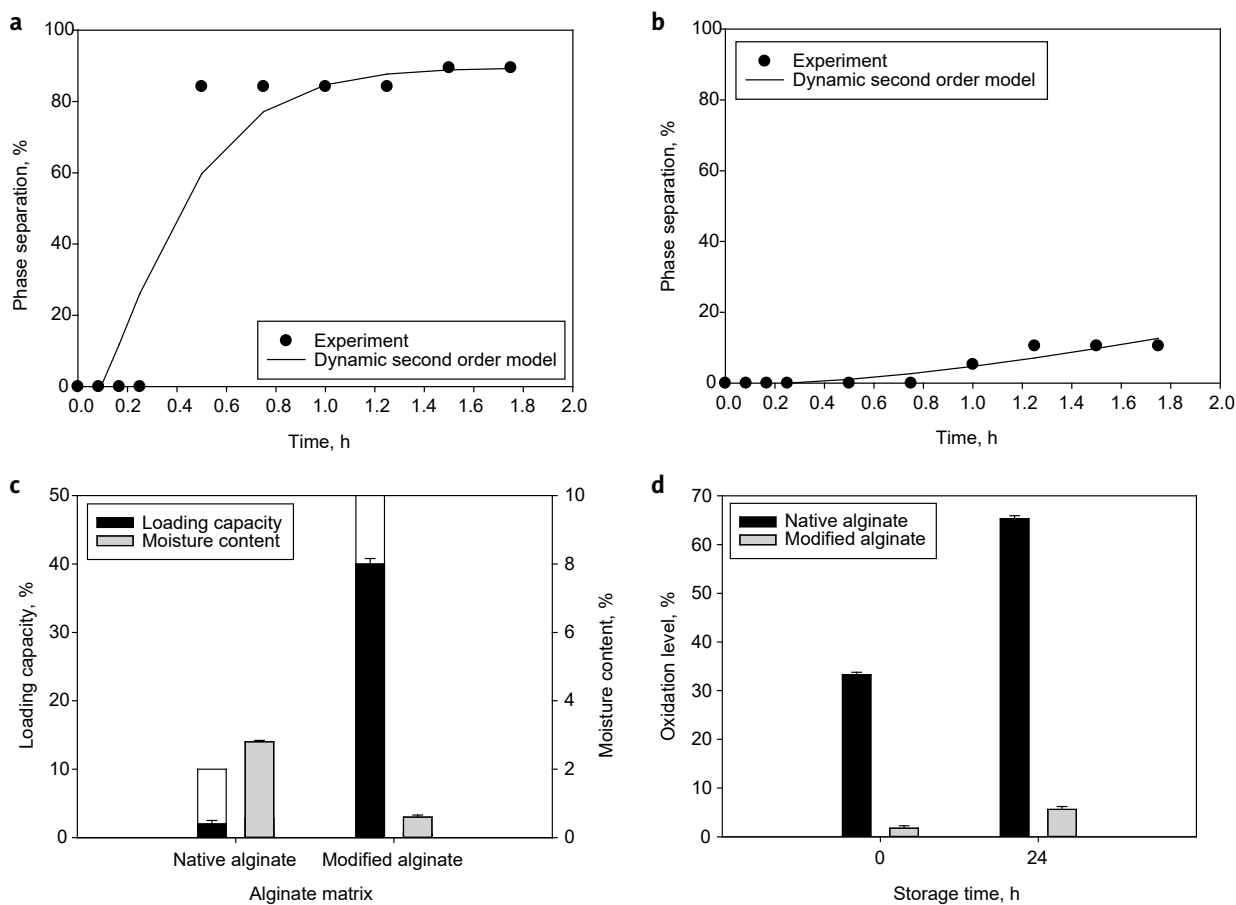


Fig. 4. Plots of dynamic second-order model of the phase separation of emulsion stabilized using: **a** – native alginate, **b** – modified alginate 400(1:4), **c** – loading capacity and moisture content, **d** – oxidation level of fish oil encapsulation

the emulsion by reinforcing the protective layer of oil droplets. This result was supported by Yang et al. (2012), who found that lower viscosity emulsion failed to slow down the upward movement of bigger droplets.

These results showed that the stability of the emulsion did not only consider the zeta potential value. Many other conditions that contribute to emulsion stability, such as viscosity, pH, ionic power, homogenization conditions, and the interaction between dispersed and continuous phases were reported to improve the emulsifying effect (Maphosa and Jideani, 2018).

The phase separation of fish oil emulsion stabilized using native alginate and 400(1:4) was modeled using a dynamic second-order model. The plots and the constants are shown in Figure 4 and Table 2, respectively. The regression values of the native alginate and

400(1:4) sample plot were 0.91 and 0.89. However, many factors influenced the rate of phase separation, such as the gravitation, diffusion mechanism of droplets to continuous phase, and floc formations (Dimitrova et al., 2000), which caused the emulsion's complex separation mechanism.

Table 2. Constants and regression value (R^2) of phase separation models

Variables	Constants			R^2
	k	k_1	d	
Native	89.47	4.682	0.024	0.91
400(1:4)	94.73	0.367	0.012	0.89

Encapsulated fish oil

Freeze-dried encapsulation of the most stable fish emulsion using 400 (1:4) samples was compared with that of the native alginate one. Figure 4 shows a significant difference in loading capacity between native and modified alginate matrix, with a value of 2 and 40%, respectively. The loading of the freeze-drying method was influenced by the oil droplet stability and freezing rate. The emulsion needed to be frozen before the drying process, which affected its stability. Accelerating the freezing rate was not suggested, as a drop in temperature would lead to oil droplet aggregation that moved to the continuous phase (Bejrappa et al., 2010).

The moisture content of encapsulated fish oil using native and modified alginate was analyzed on the seventh day after drying. The oil encapsulated using the native had a higher moisture content than the sample of modified alginate (Fig. 4c). Since there was no modification, native alginate was more hydrophilic and more easily absorbed the surrounding moisture (Shen et al., 2016). Meanwhile, the hydrophobic group linkage on the modified alginate chain prevented rehydration, as studied by Bismark et al. (2019), who modified starch using octenyl succinic anhydride. This condition was beneficial for protecting the fish oil from degradation.

The oxidation protection of native and modified alginate solids is shown in Figure 4d. After 24 h exposure to atmospheric conditions, the oxidation degree of fish oil encapsulated by native alginate increased by 32.03%, while the modified alginate sample registered a slight increase of only 3.85%. The presence of both hydrophilic and hydrophobic groups in the modified alginate built a strong barrier between oil and water, which reduced the oxidation probability (Bhandari and Singhal, 2002).

CONCLUSION

Alginate modification to increase its hydrophobic properties using DSA as a modifier was successful. Lower viscosity average molecular weight (M_v) and alginate:DSA ratio (w/w) led to a higher substitution degree being obtained. The antioxidant activity was increased after alginate modification, which reduced the oxidation degree of the fish oil to 8%. This modified alginate delayed the phase separation up to 18 h. However, the higher stability of the emulsion was produced by the emulsion prepared with a higher molecular weight

of alginate. Furthermore, freeze-dried encapsulated fish oil with the modified alginate had better performances on loading capacity, antioxidation, and anti-rehydration properties than those of the native alginate.

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