

Preparation and Evaluation of Capsule Shell Films Formulated from Brown Seaweed Alginate (*Sargassum crassifolium*)

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Abstract: Brown seaweed (*Sargassum crassifolium*) grows a lot along the coast of Indonesia, especially in the Pangandaran area. Brown seaweed may serve as a viable natural source of sodium alginate, a biopolymer extensively utilized in pharmaceutical dosage forms. Most capsule shells are made of gelatin, but as more people seek alternatives to animal products, alginate-based materials are being explored. This study aimed to extract sodium alginate from *Sargassum crassifolium* and fabricate capsule-shell films, subsequently evaluating their physicochemical properties. To obtain sodium alginate powder, the extraction process involved the following steps: acid pretreatment, alkaline immersion, sodium carbonate extraction, bleaching, precipitation, and drying. The extracted material had a moisture content of 12% and a yield of 26.21%. FTIR spectroscopy confirmed the structure of the extracted material by showing that its functional groups, such as hydroxyl, carbonyl, C–O–H, and C–O–C, were the same as those of standard sodium alginate. Capsule-shell films were prepared by varying the alginate concentration in the casting volume to 3%, 4%, and 5%. Organoleptic testing showed that all formulations produced films that were slightly brown, opaque, and felt like paper. Tests on thickness and weight showed that films with higher alginate concentrations were thicker and heavier. F3-1 had a thickness value that was closest to what is needed for commercial hard capsules, and F1-2 had a moisture content range that was acceptable for commercial capsule shells. The moisture content of the formulations ranged from 8% to 24%, depending on the temperature, humidity, and length of time they were dried. All formulations produced films that could be used, but F3-1 had the best thickness, and F1-2 had the correct moisture content, supporting their potential application as non-gelatin capsule-shell alternatives.

Keywords: Capsule Shell Films; *Sargassum crassifolium*; Sodium Alginate.

Introduction

Capsule shells, or capsule dosage forms, are very common in the pharmaceutical field because they can hide the bad taste of drugs. Capsule shells also keep the active pharmaceutical ingredients stable by protecting them from the environment [1]. There are two main types of pharmaceutical capsules: soft and hard. Capsule shells are usually made of gelatin, but they can also be made of starch or other materials that work well [2], [3].

Brown seaweed (*Sargassum crassifolium*) is one of Indonesia's many marine biological resources. The country is one of the world's biggest producers of seaweed. This species thrives along numerous Indonesian coastlines, especially in regions characterized by dead coral plates on the seabed [4], [5]. The *Sargassum* family has proteins, vitamin C, tannins, phenolics, and other important chemicals that are the main source of alginate [6], [7].

Alginate is a thickening or emulsifying agent in high demand across many industries, including food, non-food, medical, and pharmaceutical. But right now, the entire country's supply is imported. Brown seaweed alginate is a major part that strengthens the cell wall because it has a lot of it, up to 40% of the dry weight [5], [8]. There are three levels of quality standards for using brown seaweed as a source of alginate: industrial grade, food grade, and pharmaceutical grade. To get the right quality of alginate, you need both the right raw materials and a multistep

extraction process. The process of extracting alginate usually includes demineralization, neutralization, extraction, filtration, precipitation, and bleaching in that order [5], [9], [10].

Researchers are still exploring how to use brown seaweed (*Sargassum crassifolium*) to produce pharmaceutical products from marine natural resources. The goal of this study is to create and study capsule shell films made from sodium alginate taken from brown seaweed (*Sargassum crassifolium*).

Research Methods

Extraction of Na-Alginate from Brown Seaweed

The extraction process was based on the method described by Saji et al (2022) with modifications [11]. The dried brown seaweed was first soaked in 1% HCl at a 1:30 (w/v) ratio for 1 hour, then washed with distilled water until the pH reached neutrality. After that, the *Sargassum* was soaked in 0.5% NaOH, then extracted with 2% Na₂CO₃ at a 1:30 (w/v) ratio at 60–70 °C for 2 hours. Then, the mixture was filtered to remove the residue and the filtrate.

The obtained filtrate was bleached with 10% H₂O₂ under continuous stirring, then allowed to stand for 30 minutes. To make alginic acid, 15% HCl was added until the solution's pH was between 2 and 3. We used 10% NaOH to neutralise the pH, converting the alginic acid to sodium

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alginate. Then, precipitation was done by adding isopropyl alcohol in a 1:2 (v/v) ratio. The sample was filtered and then dried in an oven (Mettler) for about 24 hours.

The dried material was ground in a grinder to obtain *Sargassum* sodium alginate powder. Characterization included identification of sodium alginate functional groups using FTIR spectroscopy (PerkinElmer) and determination of its moisture content using a halogen moisture analyzer (MB65).

Preparation of Na-Alginate Capsule Shell Films

A total of 100 mL of distilled water was heated, after which sodium alginate from *Sargassum* was added at concentrations of 3, 4, and 5 g, respectively. The mixture was stirred until smooth, then glycerol was added and stirred until completely smooth. After that, the solution was heated to about 55–60 °C and poured into petri dishes to make capsule shells. To prevent clumping, the solution was poured into petri dishes and then rotated or spread evenly. It was then dried in an oven (Mettler) at 60 °C for 3–4 hours [12]. The formulation used for preparing sodium alginate capsule-shell films from brown seaweed is presented in Table 1.

Table 1. Formulation for Na-Alginate *Sargassum* Capsule Shell Films

Formulation	Na-alginate <i>Sargassum</i> (%)	Pouring volume (mL)
F1-1	3	10
F1-2	3	15
F2-1	4	10
F2-2	4	15
F3-1	5	10
F3-2	5	15

Characterization of Capsule Shell Films

The characterization of the capsule shell films included the evaluation of organoleptic properties, film thickness, film weight, and moisture content [12]

Results and Discussion

Brown seaweed was collected from Pangandaran Beach, West Java, and taxonomically identified at the Plant Taxonomy Laboratory, Department of Biology, Faculty of Mathematics and Natural Sciences, Universitas Padjadjaran, Jatinangor. The identification letter No. 27/HB/12/2022 confirmed that the brown seaweed used in this study belonged to the species *Sargassum crassifolium*.

The extraction process was done to free alginate from the cellulose matrix and separate it from other parts. This made sodium alginate (Na-alginate). This study found that the yield of sodium alginate from *Sargassum crassifolium* was 26.21%, with a moisture content of 12%. These results were higher than the sodium alginate yields obtained from other *Sargassum* species, such as *Sargassum muticum* (13.57%), *Sargassum polycystum* (15.85%) and *Sargassum natans* (19.00%) [13], [14], [15].

Functional group identification using IR spectrophotometry (Figure 1) showed that the spectrum of Na-alginate extracted from brown seaweed exhibited absorption bands consistent with those of the standard Na-alginate. The extracted Na-alginate was subsequently used as the raw material for the preparation of capsule shell films.

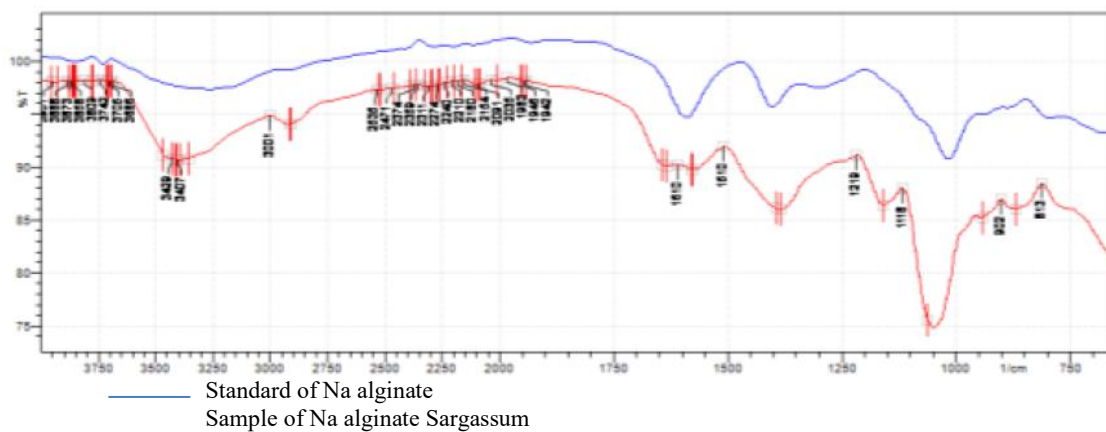


Figure 1. FTIR Spectra of reference Na-alginate and Sargassum-derived Na-alginate

The sodium alginate spectrum obtained from *Sargassum crassifolium* showed absorption bands identical to those of the standard sodium alginate. These included the hydroxyl group, carbonyl group, C–O–H bond, and C–O–C bond, appearing respectively at 3202–3419 cm^{-1} , 1636–1853 cm^{-1} , 1258–1393 cm^{-1} , and 993–1085 cm^{-1} . Based on these results, it can be concluded that the brown seaweed *Sargassum crassifolium* and the standard Na-alginate possess the same functional groups.

Table 2 summarises the organoleptic properties,

thickness, weight, and moisture content of the capsule shell films. The organoleptic evaluation showed that the films were brownish and slightly opaque. This visual trait is closely linked to the inherent pigment of the Na-alginate powder obtained during the extraction process, which imparts a brown colour to the final product. The films' opacity could also be linked to the distribution of alginate particles in the matrix and to the presence of residual particles that scatter light.

Table 2. Characterization Results of Na-alginate Sargassum Capsule Shell Films

Formulation	Organoleptic			Thickness (mm)	Weight (mg)	Moisture content (%)	
	Color	Odor	Clarity				Form
F1-1	Slightly brown	Odorless	Murky	Paper-like form	0.03	59.00	8.00
F1-2	Brownish	Odorless	Murky	Paper-like form	0.06	69.00	13.00
F2-1	Slightly brown	Odorless	Murky	Paper-like form	0.18	67.00	12.00
F2-2	Brownish	Odorless	Murky	Paper-like form	0.20	245.00	17.00
F3-1	Brownish	Odorless	Murky	Paper-like form	0.16	167.00	21.00
F3-2	Brownish	Odorless	Murky	Paper-like form	0.26	396.00	24.00

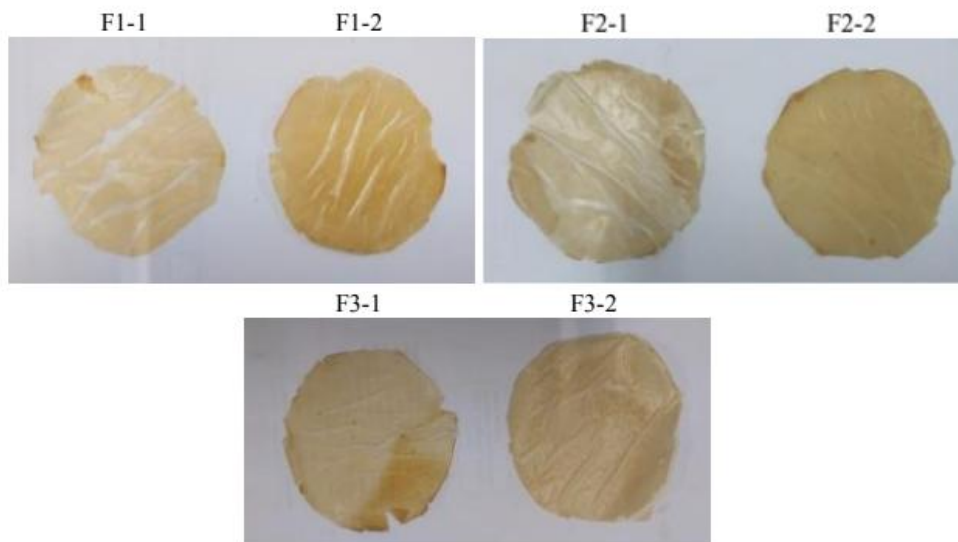


Figure 2. Results of Na-alginate Sargassum capsule shell film

The concentration of Na-alginate in the formulation affected the texture, physical integrity, and appearance of the capsule films. Films with higher polymer concentrations were usually thicker, denser, and stiffer. This is because more polymeric networks are formed within the matrix. On the other hand, lower concentrations produced thinner, less compact films that were also more flexible. These differences show how important the amount of polymer is in determining the physical and mechanical properties of capsule films. To get the right balance of strength, flexibility, and uniformity, the films must be optimized [16], [17].

The thickness of the capsule films was measured using a calliper to ensure precise detection of even slight variations across formulations. This parameter is essential because the structural integrity and mechanical performance of capsule films are strongly influenced by their thickness. The physicochemical correlation between capsule shell thickness and weight is attributable to the accumulation of total polymer solids, such as sodium alginate, throughout the processes of capsule film production and drying [18]. A consistent thickness indicates uniform polymer distribution and optimal film formation during casting and drying. Any irregularities in thickness may indicate inconsistencies in the polymer solution, uneven spreading, or variations in drying rate, all of which can affect the film's usability and overall quality [19], [20].

Meanwhile, the weight of each film was determined using an analytical balance with high sensitivity to capture minor differences between samples. The purpose of recording film weight was to establish its correlation with film thickness and evaluate how polymer concentration affected the total solid content. Films with greater thickness consistently showed higher weights, as they contained a

larger amount of dried Na-alginate matrix following solvent evaporation. This relationship highlights how increasing the alginate concentration leads to denser and more substantial films, emphasizing the importance of optimizing formulation parameters to achieve films with desirable mechanical strength and uniformity [17].

F3-1 had the thickness of the capsule film that was closest to that of commercial capsule shells (0.107 mm), with a measured thickness of 0.16 mm. The F1-2 formulation had a higher film weight than the F2-1 and F2-2 formulations, which were both higher than the F3-1 formulation. This aligns with the relationship between Na-alginate concentration and film mass. An increase in capsule thickness directly contributes to an increase in capsule weight [17]. The weight standard for size 0 hard capsules from PT. Kapsul Indo ranges from 87 to 107 mg. Products derived from organic materials are generally susceptible to mold and fungal growth when their moisture content exceeds 20–60%, and bacterial contamination becomes more likely when moisture levels exceed 60%. Based on the commercial standard set by PT. Kapsulindo Nusantara, capsule shells must have a moisture content between 12.5% and 15% [21].

Research on natural polymer capsules reveals that combinations of matrix components, such as alginate and starch, that result in denser, more stable film structures are associated with higher weight values and increased wall thickness, aligning with the principle of enhanced total solids in polymer solutions post-drying [20]. The results of this study show that films made with Na-alginate concentrations between 3% and 5% had moisture contents of about 8% to 24%. Variations in moisture content were affected by drying temperature, humidity, and drying duration [22].

Conclusion

Na-alginate derived from brown seaweed (*Sargassum crassifolium*) can be used to make capsule shell films compatible with all tested formulations. Formulation F3-1 produced the film thickness that was closest to that of commercial capsule shells, while formulation F1-2 produced the moisture content that was acceptable for commercial capsules.

Author's Contribution

B. V. E. Viera contributed to the experimental design, conducted the extraction and characterization of sodium alginate, performed capsule shell films fabrication, and carried out data collection, analysis and revised the manuscript. S. Saepudin contributed to the research process, refined the methodology, validated the analytical results, contributed to data interpretation, and revised the manuscript. L. I. A. Saidah conducted the extraction and characterization of sodium alginate and performed the fabrication of capsule shell films.

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