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Supercritical CO_2-Dried Alginate/Soy Protein Isolate Beads Aerogels for Natural Oil Absorption

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Oil spills negatively impact ecosystems and groundwater supplies, harm human and animal welfare, and indirectly contribute to global warming and climate change. Natural oil absorbents, such as aerogels, have gained attention for being low-cost, environmentally friendly, biodegradable, and sustainable. This study synthesized natural oil absorbents from a combination of soy protein isolate and alginate using the sol-gel method. These absorbents underwent surface modification and were then dried using supercritical CO_2 at 110 ± 5 bar and 40 °C for 4-5 h to produce alginate/soy protein isolate aerogels. Aerogels are porous solid materials with a huge surface area; thus, their potential as oil absorbents is investigated. The physical characteristics of the oil absorbent are characterized via Fourier transform infrared (FTIR) to identify the interaction of functional groups in the hybrid aerogel, contact angle analysis to evaluate the changes on the surface functionalization characteristics, and oil absorption capacity to measure the ability of aerogel to absorb oil. The composition of the alginate and soy protein isolate was found to influence the hydrophobicity of the aerogels. The alginate/soy protein isolate aerogel with a ratio of 1:1.5 was demonstrated as the optimum formulation of natural oil absorbent with nearly superhydrophobic characteristics with 142.8^o of contact angle and 76% oil absorption capacity.

1. Introduction

Current recovery methods for oil spills are often questioned due to their limitation in completely removing the spilled oil. The approaches include in-situ burning, bioremediation (microbes), chemical (dispersant, solidifiers), and physical (booms, skimmers, sorbent). Among these methods, sorbents perform best which can effectively absorb oil. Sorbents are insoluble materials that could remove oil via adsorption, absorption, or both (Negreiros et al., 2022). However, current sorbents that are mainly made from synthetic material (such as polypropylene) and natural material (such as recycled cotton, and paper) have poor degradability, and low oil selectivity, respectively. This causes the sorbent to sink to the bottom and lead to other environmental issues. Hence, there is considerable interest in finding advanced materials with affordable, sustainable, and efficient oil absorbents to overcome this limitation. Natural organic sorbents like cellulose, starch, and glycogen are primarily made of carbon, hydrogen, and oxygen, making them hydrophilic. Numerous studies have reported on the utilization of biopolymers as an alternative to synthetic absorbents, for example, cellulose (Zhou and Xu, 2020), cotton (Lee et al., 2016), chitosan (Li et al., 2018), and alginate (Wang et al., 2022).

Alginate, a natural anionic polysaccharide originating from seaweeds, offers the benefits of biocompatibility, low cost, and abundant availability (Wang et al., 2022). Alginate is good at absorbing water due to the hydroxyl groups (OH) on its surface, which limits its use in cleaning oil. Nevertheless, the surface of the alginate could be altered to become hydrophobic by replacing the OH groups. Wang et al. (2022) successfully proved that after a hydrophobic alteration, superhydrophobic absorbents can be made from initially hydrophilic alginate foams. Alginate possesses a simple synthesis process, and good gelation capability, and could serve as the framework

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of a sorbent. However, a single biopolymer often suffers from poor mechanical strength and oil selectivity. Thus, a combination of the alginate with other resources such as biopolymer making as hybrid materials could enhance its mechanical and functional properties. In the environmental benefits, hybrid biopolymer oil absorbent could be the solution to current limitations while being cost-effective and less straining to the environment (Huang et al., 2022). Soy protein isolate (SPI) is a natural polymer that has been extensively investigated for biomedical application, delivery of bioactive compounds, and absorption. SPI has been considered an excellent material for numerous applications due to its good biocompatibility, highly soluble water, natural abundance, biodegradable, anti-carcinogenic, and non-immunogenic properties. SPI, a plant protein produced cheaply, abundantly, and sustainably, consists primarily of the 7s (-conglycinin) and 11s globular protein component fractions (glycine). Compared to several biodegradable polymers and natural proteins, SPI exhibits important qualities for an oil absorbent such as water resistance and stability. These characteristics have made SPI an intriguing starting material in biotechnology and biomedical uses (Prusty et al., 2019). The primary challenge lies in modifying these materials to be hydrophobic while retaining each of their capabilities. In the presented work, sodium alginate and soy protein isolate are combined producing hydrogels, followed by surface modification to alter the surface functional characteristics from hydrophilic to hydrophobic.

2. Methodology

Soy protein isolate (SPI) was supplied from Radiant Code Sdn. Bhd whereas sodium alginate (SA) was purchased from Signa Aldrich. The chemical reagents of ammonium persulfate (APS), and N, N-methylene bisacrylamide (MBAm) for synthesis oil absorbent and methyltrimethoxysilane (MTMS) as an agent for preparing the surface modification process were obtained from Sigma Aldrich. Calcium carbonate ($CaCO_3$), calcium chloride ($CaCl_2$), and ethanol were purchased from R&M Chemicals whereas 99% purity of liquefied carbon dioxide (CO_2) for supercritical drying was supplied by SR Focus Trading.

2.1 Synthesis of alginate/soy protein isolate absorbent

Alginate/soy protein isolate hydrogel was prepared by adding approximately 1-2 g of sodium alginate (SA), 3 g of soy protein isolate (SPI), and 0.6-1 g of ammonium persulfate (APS), N, N-methylene bisacrylamide (MBAm), and calcium carbonate ($CaCO_3$) into 100ml of distilled water. The mixture was stirred until it became homogenized by using a high-speed homogenizer (model FSH-2A). To initiate the gelation process, alginate/soy protein hydrogel was transferred into a syringe to produce a bead shape of the hydrogel. The hydrogel bead is pumped using a syringe pump (NE 300 Infusion Syringe Pump) into a calcium chloride ($CaCl_2$) solution at the rate of 10 ml/min. The hydrogels are then covered with parafilm and left in a freezer at -4 ^oC for one day.

To replace the water content in the hydrogel, solvent exchange must be performed before supercritical drying. In the solvent exchange stage, the hydrogels are submerged in ethanol/water solution (10, 30, 50, 70, 90, and 100% v/v) every 24 h, which finally produces alcogels after completing the solvent exchange procedure.

2.2 Preparation of surface modification for alginate/soy protein isolate

Before the drying process, the alginate/SPI alcogels that had been completed immersion in ethanol/water exchange were subjected to a surface modification using methyltrimethoxysilane (MTMS) to modify the functional groups from hydrophilic to hydrophobic. The alcogels were immersed in the MTMS solution for 48 h at room temperature. The modified alcogels were then wrapped in filter paper and placed in an extraction chamber of supercritical drying setup.

2.3 Supercritical CO_2 drying

The alcogels were dried using supercritical carbon dioxide (CO_2) at 110 ± 5 bar and 40 ^oC for 4-5 h. Briefly, the $SCCO_2$ extracts the liquid ethanol from the alcogels, and the ethanol-$SCCO_2$ stream flows into the separator column and is separated from the $SCCO_2$ by precipitating in the separator column that was set at 40 bar and 80 ^oC, to convert the $SCCO_2$ into a gaseous state before was recycled to the CO_2 chamber. This drying process was continuously conducted every hour, the extracted ethanol was collected and taken out from the system. After 4-5 h, depressurization of the extraction chamber took place at a rate of 3 bar/min to prevent the aerogels from rupturing.

2.4 Characterization Analysis

2.4.1 Physical and thermal properties

The characteristics of the synthesised aerogels were analysed using Fourier transform infrared spectroscopy (FTIR) (Model Perkin Elmer Spectrum One) to determine the functional group and its interaction in the aerogels. The thermal stability of the aerogels was measured using the thermal gravimetric analysis (TGA, Model Mettler Toledo) analysis, where the measurement was performed at a temperature range from 20 to 400 °C at 10 °C

/min of nitrogen gas. Meanwhile, the contact angle analysis was performed using the CVA3000 model to measure the hydrophobic and wettability of the aerogel.

2.4.2 Oil absorption analysis

The absorption ability of the aerogels was performed to determine the effectiveness of the aerogel as an oil absorbent and its capacity. The oil absorption capacity is to analyse the tendency of aerogel to absorb oil. To determine the oil absorption capacity, a sample of waste engine oil was used as a pollutant model. The weight of aerogel before and after were measured, and the percentage of the oil absorption capacity was calculated as the equation below:

$$OAC = \frac{W - W_o}{W_o} \times 100\% \tag{1}$$

where *OAC* is the oil absorption capacity, W_0 is the weight of the aerogel before absorption, and *W* is the weight of the aerogel after absorption. The analysis was repeated more than three times and the data presented are the average values with standard deviations.

3. Results and Discussion

3.1 Alginate/SPI aerogels functional groups

The Fourier transform infrared spectroscopy (FTIR) analysis in Figure 1 shows that the spectra of sodium alginate reveal carboxyl vibrations at 1608 cm^{-1} and 1417 cm^{-1}, corresponding to asymmetric and symmetric stretching vibrations of carboxyl groups, respectively (Li et al., 2022). The overlapping peak vibration of the -OH group stretching in sodium alginate was attributed to absorption bands at 3200 cm^{-1} and 3600 cm^{-1} (Xiao et al., 2022) and demonstrated the alginate characteristic and the hydrogen interaction between alginate polysaccharides and protein (Hannah Sofiah et al., 2023).

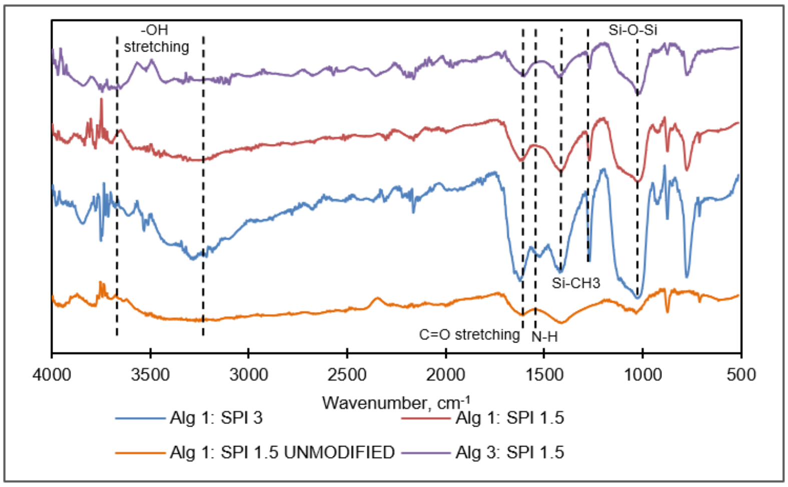

Figure 1 FTIR spectra alginate/SPI aerogels at different ratios

The spectra also indicate the presence of both alginate and soy protein isolate. Soy protein isolate peaks appear at several peaks absorption, mainly 1640 cm^{-1} for amide I which represents the C=O stretching, 1550 cm^{-1} for amide II attributed to the N-H bending, and nearly 1238 cm^{-1} for amide III stretching i.e., C-N and N-H in-phase bending) (F. Liu et al., 2023). In the modified aerogels, the absorption peak intensities are significantly higher at 1025 cm^{-1} which corresponds to the -Si-O asymmetric stretching in -Si-O-Si-, and at 1271 cm^{-1} and 1411 cm^{-1}, attributed to $-CH_3$ asymmetric stretching and $Si\text{-}CH_3$ groups bending, compared to the unmodified aerogels (Yang et al., 2012). Furthermore, the C-H stretching vibration becomes more apparent after undergoing surface modification. Therefore, the alginate/soy protein isolate aerogels with surface modification have high peaks compared to unmodified alginate/soy protein isolate aerogel (Meng et al., 2018). It was explained that unmodified alginate/soy protein isolate with a ratio of 1:1.5 was hydrophilic and other aerogels were hydrophobic.

3.2 Thermal stability

Figure 2 presents the thermal stability of the alginate/SPI aerogels at different composition ratios. It is shown that the unmodified alginate/SPI 1:1.5 exhibited the lowest thermal stability which lost more than 55% of its weight compared to modified aerogels. The modified aerogels with hydrophobic properties have more methyl groups than the unmodified aerogels may contribute to the higher thermal stability due to the inert nature of the methyl groups. In comparison, the 1:1.5 aerogel loses its initial weight of 70% over 400 °C, which is lower compared to the other composition ratios. This indicates that the 1:1.5 aerogel has higher stability that may attributed to the strong interaction through covalent bonding between alginate and SPI functional groups. Nevertheless, as the SPI increased to 3 wt%, its thermal stability dropped with 62% weight loss. This is probably due to weaker bonding between the alkyl chain in the SPI and the alginate backbone. Furthermore, increasing the alginate content from 1 to 3 wt% has shown the apparent reduction of its weight to 85% at the temperature of 120 to 220 °C. At this stage, the depolymerization of the alginate has occurred (Mustapa et al., 2016) and the high ratio of alginate has made it have less interaction with the alkyl chain of the SPI, hence decreasing its thermal stability.

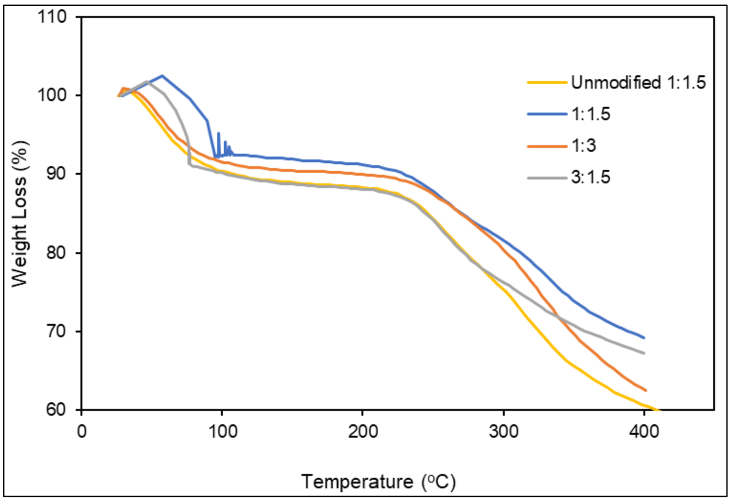

Figure 2 Thermal stability of the alginate/SPI aerogels at different ratios

3.3 Contact angle and wettability

The contact angle of each aerogel is examined and presented in Figure 3. Results demonstrated that the aerogels with ratios of 1:1.5, 1:3, and 3:1.5 had readings of more than 90°, indicating the success of the surface modification to hydrophobic from hydrophilic using MTMS reagent. The MTMS has been reported to act effectively as a silane coupling agent to improve the hydrophobicity of substance (Lee et al., 2019). The methyl group in MTMS enables the long alkyl chain in alginate/SPI aerogel to create a well-ordered structure, hence explaining the hydrophobic increasing action. The silanes of MTMS were deposited onto the smooth surface of alginate/SPI aerogel to diminish the background of the rough surface micro-structures (Ou et al., 2020). The gluing of MTMS to the aerogel was responsible for the improved durability. This situation indicates that the MTMS was deposited onto the aerogel, and the interfacial connection between the alginate/SPI was strengthened through MTMS interlinking. On the other hand, the unmodified aerogel of 1:1.5 alginate/SPI possesses hydrophilic characteristics, indicated by 0° of the contact angle. This unmodified alginate/SPI aerogel apparently can absorb both oil and water as shown in Figure 3A (see in the wettability section), showing that the 1:1.5 (unmodified) does not have absorption selectivity between oil and water.

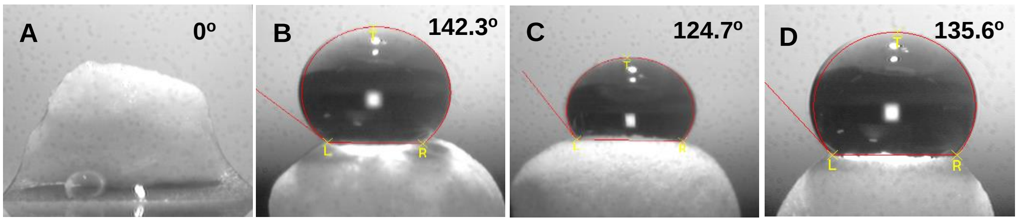

Figure 3: Water droplet on (A) unmodified alginate/SPI aerogels with ratio 1:1.5, modified alginate/SPI aerogels with ratio (B) 1:1.5, (C) 1:3, and (D) 3:1.5

Figure 3 (3A, 3B 3C, and 3D) shows the contact angle for each alginate/soy protein isolate with different ratios by using CVA3000 model equipment. The sample of 1:1.5 unmodified aerogel surface (see Figure 3A) shows that the water drops did not create any contact angle on its surface, indicating its hydrophilic characteristics. On the other hand, for the modified alginate/SPI aerogels (Figure 3B, 3C, 3D), the water droplet had developed a specified contact angle on the aerogel surfaces due to the surface tension between water-solid, demonstrating the hydrophobic properties of the aerogels. The contact angles had increased from 124.7° to 142.8 ° as the SPI reduced from 3 to 1.5 wt% at constant 1 wt% alginate, showing that increases in the SPI to 3 wt% did not enhance the hydrophobicity of the alginate/SPI. On the contrary, the values were found to decrease from 142.8º to 135.6º when the alginate content increased from 1 to 3 wt% at a constant SPI content of 1.5 wt%. These findings suggest that the hydrophobic characteristics of the alginate/SPI aerogels rely on the composition of the alginate and SPI. The SPI content dominates the alginate in governing the hydrophobic surface due to the presence of alkyl groups functional groups.

Selective wettability of alginate/SPI aerogels was determined by dropping water and waste engine oil droplets (yellow colour) on the aerogel (Liu et al., 2023). It exhibits that for the modified alginate/SPI aerogels (Figure 4B, 4C, and 4D) when water droplets (purple coloured-water) came into touch with it, they remained on the surface to retain the spherical shape, indicating the hydrophobic surface of the aerogels. The figures also depicted the selective absorption of the aerogels on waste engine oil, where the modified alginate/SPI aerogels (Figure 4B, 4C, and 4D) completely absorb waste engine oil, meanwhile, the unmodified aerogel (Figure 4A) absorbs water since it does not undergo the surface modification phase.

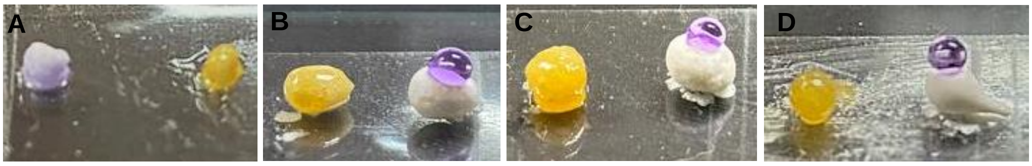

Figure 4: Wettability analysis of oil and water on (A) unmodified alginate/SPI aerogels with a ratio 1:1.5, modified alginate/SPI aerogels with ratio (B) 1:1.5, (C) 1:3, and (D) 3:1.5 (Yellow colour: engine oil, purple colour: water)

3.4 Oil absorption capacity

Oil absorption capacity was measured by soaking alginate/SPI ratios of 1:1.5 (unmodified), 1:1.5, 1:3, and 3:1.5 aerogel in waste engine oil. As shown in Table 1, the oil absorption capacity of 1:1.5 was found as the highest at 76%, while the lowest oil absorption capacity of 58% was obtained for the alginate/SPI ratio of 3:1.5. This can be explained by the blend variation of the biopolymers that attributed to the dominant characteristics on the hybrid materials. Alginate with high soy protein isolate has a large particle size which may be caused by the dense network structure by the formation of soy protein and sodium alginate through covalent bond interaction (Chen et al., 2022). The excessive amount of alginate in the ratio 3:1.5 reduces oil absorption ability compared to soy protein isolate. The sodium alginate had a compact microstructure, which reduced the porosity and roughness of the aerogel, preventing oil from reaching the samples (Li et al., 2023).

Table 1: Oil absorption analysis

Ratio (Alginate/soy protein isolate)	Oil Absorption Capacity, %
1:1.5 (unmodified)	70 ± 0.9
1:1.5	76 ± 1.2
1:3	73 ± 1.5
3:1.5	58 ± 0.2

4. Conclusion

Utilizing natural materials such as alginate and SPI shows their promising potential as natural oil absorbents. Variation composition of the alginate and SPI influences the hydrophobicity of the aerogel absorbent. A higher alginate content of up to 3 wt% and increased SPI to 3 wt% reduces the hydrophobic characteristics of the alginate/SPI aerogels, hence the decreased absorption capacity. The alginate/SPI aerogel 1:1.5 has been found as the optimum formulation of natural-based absorbent for oil absorption application. It has the highest oil absorption capacity of 76% and a contact angle of 142.8º which is nearly to the superhydrophobicity of greater than 150º. A high contact angle with nearly superhydrophobic and high oil absorption on the alginate/SPI aerogels indicates the successful surface modification of turning the hydrophilic alginate/SPI to hydrophobic in a simple functionalization process. The use of monomeric MTMS with a combination of ethanol solution is recommended to enhance the hydrophobicity characteristics of the alginate/SPI aerogels.

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