Characterization of bioplastics developed from Kappaphycus alvarezii crosslinked with commercial sodium alginate

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Research Article

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Posted Date: March 31st, 2023

DOI: https://doi.org/10.21203/rs.3.rs-2754347/v1

Additional Declarations: No competing interests reported.

Characterization of bioplastics developed from *Kappaphycus alvarezii* crosslinked with commercial sodium alginate

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Abstract
Plastic pollution has become one of the most concerning problems globally due to excessive use of one-time use plastics. However, bioplastics could be the answer to help combat this problem as they are readily biodegradable. Development of bioplastics was done by mixing seaweed biomass into distilled water at specific ratio, using glycerol as plasticizer. Bioplastics were developed at the ratio of 100:0, 75:25, 50:50, 25:75, and 0:100 *K. alvarezii* to commercial sodium alginate ratio. Characterization was done based on their appearance, mechanical, thermal and permeability properties, and biodegradability. Resulted data for their appearance showed that when more *K. alvarezii* was in the mixture there were more colour differences in comparison to white background and the same trend for the opacity due to the natural colour of whole *K. alvarezii*. As for their mechanical properties, tensile strength of the bioplastics decreased from 100:0 ratio to 0:100 ratio at 7.91 ± 0.45 MPa (100:0), 6.78 ± 0.31 MPa (75:25), 5.20 ± 0.37 MPa (50:50), 4.13 ± 0.17 MPa (25:75) and 3.76 ± 0.14 MPa (0:100), respectively. Same goes for their elastic modulus at 20.93 ± 0.61 MPa (100:0), 16.47 ± 0.99 MPa (75:25), 11.42 ± 0.53 MPa (50:50), 8.78±0.45 MPa (25:75) and 6.65±0.32 MPa (0:100), respectively. This shows that the addition of alginate enhances the elasticity but decreases tensile strength. As a conclusion, developed seaweed-based bioplastics resulted different properties at different mixture ratio show potential to be incorporated into the market as they are a greener option to fight single-use plastic wrappings such as saran wrap, beverages and food additive packets.

Keywords: Kappaphycus alvarezii, crosslinked biopolymer, algae, alginate, sustainable bioplastics

1.0 Introduction
Seaweeds are macroalgae that can grow wildly in the ocean or be commercially grown, each for their own purpose according to their type and species (Phang et al., 2019). Seaweed has been used in many industries, especially the food industry, either as a raw ingredient or as an additive. It has also been used as a supplement in medicine, as it has been shown to have many health benefits for its users (Buschmann et al., 2017). There are a lot of circumstances that affect the growth of seaweeds, which at the same time can alter their properties when conditions are changed (Sugumaran et al., 2022). Malaysia has a great sea environment for the growth of seaweeds, which has led to concentrated seaweed availability (Hussin & Khoso, 2017). For that reason, *K. alvarezii* is abundant even though it is in the group of commercialized seaweeds (Hamid et al., 2020). Since the price of seaweed is not controlled, seaweed growers are demotivated to continue and instead concentrate on other business sectors because the government does not care about their welfare, which caused the price of seaweed to increase as a result of imports from other ASEAN countries (Asri et al., 2021; Hussin & Khoso, 2017).

Seaweeds are also studied for their hydrocolloid extracts, especially alginate, carrageenan, and agar, which are used in a wide range of fields (Qin, 2018). Other than being in those mainstream industries, current studies have focused on trying to introduce seaweed as a biomass source for bioplastics (Rajendran et al., 2012; Zhang et al., 2019). This is because studies have shown that seaweed-based bioplastics have good protective qualities (Lomartire et al., 2022). There are a few types of seaweed-based bioplastics that differ in terms of their source of biomass, like using whole seaweed (Hanry & Surugau, 2020; Moey et al., 2018), agar (Hii et al., 2016), alginate (Castro-Yobal et al., 2021; Yupa et al., 2021), and carrageenan (Farhan & Hani, 2017; Nasution et al., 2019). However, the properties of seaweed-based bioplastics depend on a few factors, starting from the source of biomass until the process of developing bioplastics (Lomartire et al., 2022).

Before going deeper into this study, *K. alvarezii*- and alginate-based bioplastics were studied to understand their properties individually. *K. alvarezii*-based bioplastics are closely similar to semi-refined or refined carrageenan-based bioplastics but lack purity in terms of k-carrageenan since *K. alvarezii* is the raw material for k-carrageenan production (Rudke et al., 2020). According to research, bioplastics are frequently crosslinked. For instance, adding pectin and mica flakes to increase barrier qualities (Alves et al., 2010) or crosslinking with essential oils to improve antibacterial and antioxidant activities (Shojaee-Aliabadi et al., 2013, 2014). Previous research on alginate-based biofilms concentrated on using them in medicine as pill capsules, pads for treating wounds, and others (Zia et al., 2017). According to studies, the main reason why other elements were added to alginate-based films was to improve their mechanical characteristics (Abdullah et al., 2021).

The study focused on using both alginate and *K. alvarezii* to produce bioplastics in the hopes of getting the good properties from both as their individual qualities were proven by previous studies. It was concluded that both *K. alvarezii*- and alginate-based films individually each have their own pros and cons. Hanry et al. (2022) and Hanry & Surugau (2020) studies revealed that there was more room for the use of *K. alvarezii*- and alginate-based bioplastics with the goal of replacing single-use packaging. This study is a follow-up that aims to serve as a baseline reference for following research by comparing each characteristic to assess the effects of combining commercial sodium alginate with *K. alvarezii* at various ratios to produce seaweed-based bioplastics.

2.0 Materials and method

2.1 Materials

The raw materials used in this study were fresh *Kappaphycus alvarezii* from Semporna, Sabah and commercial sodium alginate (SYSTERM). Chemicals used include glycerol with purity of 99% (SYSTERM), magnesium nitrate hexahydrate (SYSTERM), silica gel (SYSTERM).

2.2 Development of seaweed-based bioplastics

The method was adapted from a study by Rahmawati et al., (2019) with slight modification after preliminary study. The biomass to water ratio was 1:60, where biomass used in this study is whole seaweed (*K. alvarezii*) (from this point referred as WS) and commercial sodium alginate (from this point referred as CA). The mixture ratio of WS to CA was at set at 100:0, 75:25, 50:50, 25:75, and 0:100 %(w/w), respectively. 2g of biomass was dissolved in 120mL of distilled

water while being heated and stirred continuously to 60°C. Solution was remained for 10 minutes once the temperature reached 60°C before adding 1% (v/v) glycerol. The mixture was blended for 1 minute to homogenize the mixture before being heated to 80°C and remained for 1 minute. Mixture was taken off the heating plate. By slowly swirling the solution, any bubbles in the solution were eliminated with a glass rod. Bioplastics were created using the casting method, in which the solution was poured onto a petri dish and dried for 24 hours at 50°C in a vented dryer. Samples were left to cool before removing them from the cast which resulted Figure 3.1.

2.3 Characterization

2.3.1 Functional Group Analysis

Functional groups of raw materials and samples were determined using ATR-FTIR (Agilent Technologies). All the samples were tested to compare the differences and intensity in each mixture, plus before any reaction (raw material).

2.3.2 Moisture Content

Moisture content was calculated using equation 2.1 after heating the sample in an oven at 105°C for 3 hours.

$$\text{Moisture Content } (\%) = \frac{Initial\ Weight - Final\ Weight}{Initial\ Weight}\ X\ 100\% \tag{2.1}$$

2.3.3 Appearances

Prior to any testing, the thickness of samples was measured using a digital caliper. For their opacity, they were scanned at 660nm using UV-Visible spectrophotometer (Agilent Technologies). Calculation for opacity was done using equation 2.2 with the absorbance resulted from the scans.

$$\text{Opacity (O)} = \frac{Absorbance}{Film\ Thickness} \tag{2.2}$$

For their colour determination, each of them was scanned using colour reader (Konika Minolta), which gave L* (brightness), a* (+ redness, - greenness), b* (+ yellowness, - blueness) readings for each sample. The resulted data was calculated using equation 2.3, L = 83.8 , a = 12.4 , b = -3.0. to determine the colour difference in comparison to the white background

$$\text{Colour Difference } (\Delta E) = \sqrt{((L - L*)^2 + (a - a*)^2 + (b - b*)^2)} \tag{2.3}$$

2.3.4 Thermal Stability

Thermal stability of samples was determined using a Thermal Gravimetric Analyzer (TGA) (Mettler Toledo) with a temperature range of 25 - 700°C, a ramp speed of 20°C/min, and a gas flow rate of 30mL/min. The TGA/DTG graph was obtained from the scan.

2.3.5 Mechanical Properties and Water Vapour Permeability

according to ASTM D-618 (ASTM, 2002), samples were subjected to initial conditioning for 72 hours at 53% RH using magnesium nitrate hexahydrate ($Mg(NO_3)_2.6H_2O$) before undergoing mechanical and water vapour permeability testing. For determination of mechanical properties, samples were cut into dumbbell shape in strips of 90mm x 15mm and fixed on using Universal Tensile Machine (GoTech Instruments) at 5mm/min with 50mm gauge.

For water vapour permeability tests, silica served as the 0% RH medium while water served as the 100% RH medium. Each sample was placed in a desiccator on a permeation cup, and the weight of the sample was measured hourly for 6 hours. The permeability result was calculated using equation 2.4. Note that the silica was heated at 100°C for 3 hours prior to use.

$$\text{WVP} = \frac{Change\ in\ weight\ x\ Thickness}{Area\ exposed\ films\ x\ Time\ x\ Vapour\ pressure} \quad (2.4)$$

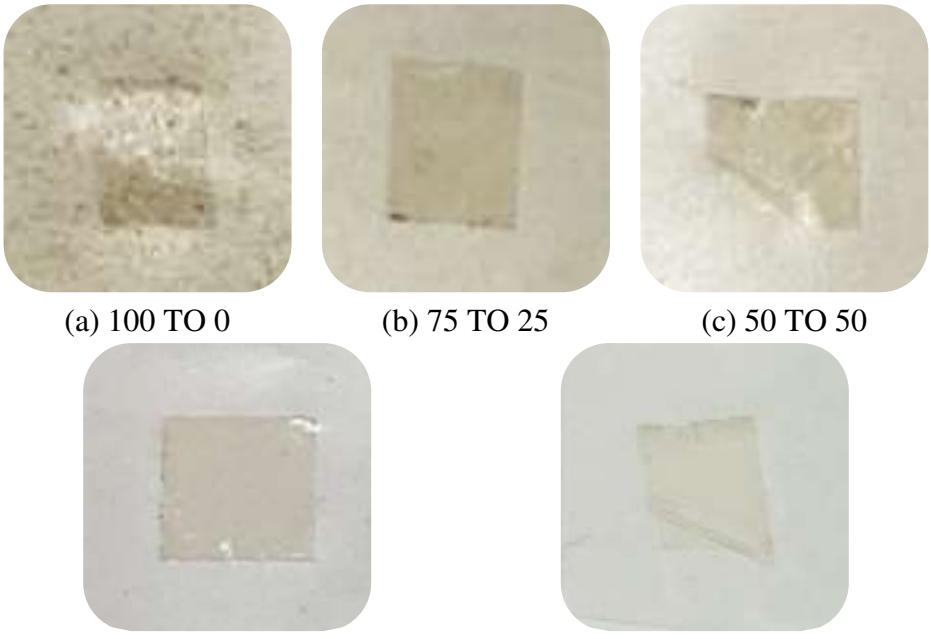
2.3.6 Biodegradability

The soil burial method was modified in relation to the method proposed by Chuensangjun et al. (2013) for sample biodegradability. Samples were cut into 5cm × 5cm squares and buried in the ground 10cm deep at 40% RH. Samples were monitored, and the number of days required for samples to decay completely was recorded.

2.4 Statistical Analysis

All samples were developed and tested in 3 replicates. The statistical analysis, Analysis of Variance (ANOVA) with Tukey HSD multiple comparison as post-hoc analysis was done using IBM SPSS Statistics 29.

3.0 Results



(a) 100 TO 0 (b) 75 TO 25 (c) 50 TO 50

(d) 25 TO 75 (e) 0 TO 100

Figure 3.1: Bioplastics at different WS to CA mixture ratio

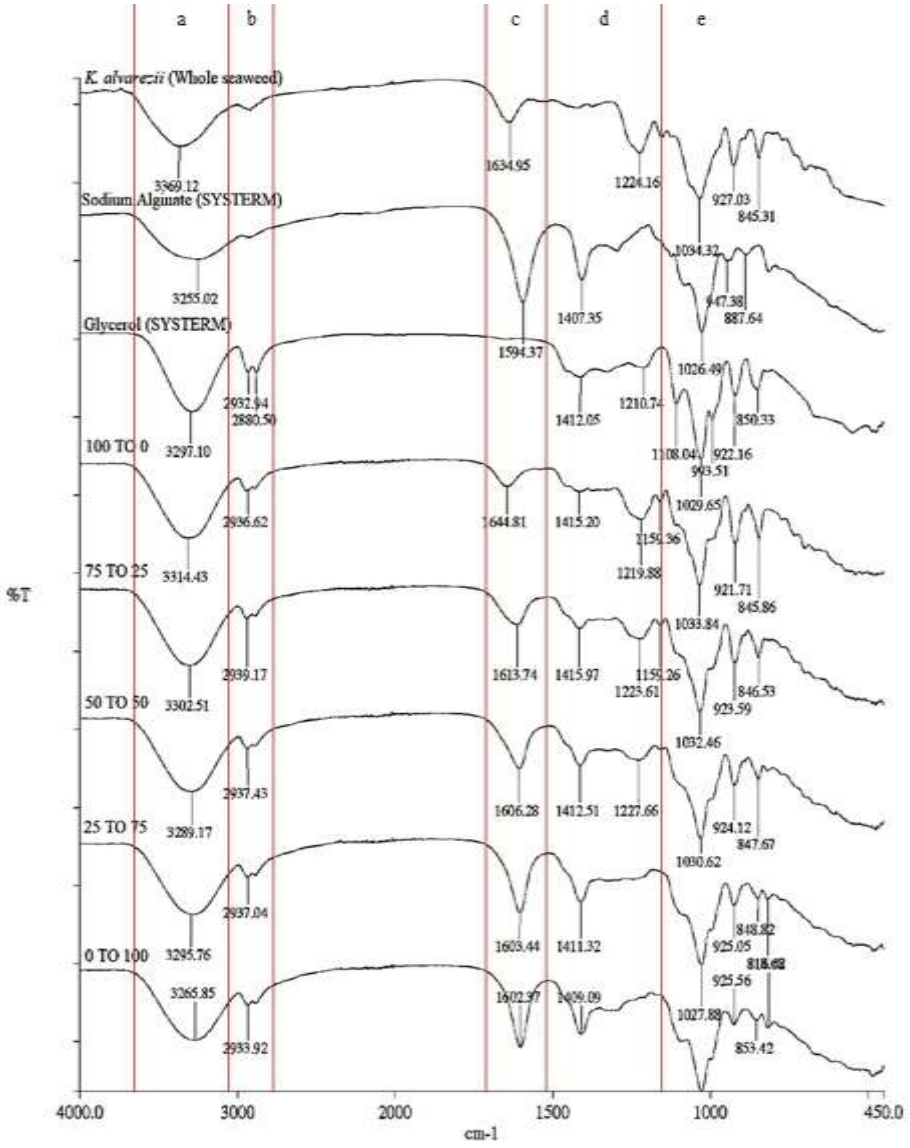


Figure 3.2: FTIR spectrum of *K. alvarezii,* sodium alginate, glycerol, and bioplastics at different WS to CA mixture ratio

Table 3.1: Moisture content, thickness, and opacity of bioplastics at different WS to CA mixture ratio

Sample	Characterization		
	MC (%)	T (mm)	O (cm^{-1})
100 TO 0	23.4 ± 0.6^{a}	0.054 ± 0.05^{a}	2.757 ± 0.001^{a}
75 TO 25	23.4 ± 0.4^{a}	0.052 ± 0.04^{a}	2.468 ± 0.001^{b}
50 TO 50	23.3 ± 0.3^{a}	0.050 ± 0.04^{a}	1.844 ± 0.008^{c}
25 TO 75	23.3 ± 0.4^{a}	0.053 ± 0.01^{a}	1.057 ± 0.001^{d}
0 TO 100	23.5 ± 0.5^{a}	0.052 ± 0.04^{a}	0.653 ± 0.001^{e}

MC: Moisture content, T: Thickness, O: Opacity; Data reported are mean ± standard deviation (n = 3), and values of different letters $^{a\text{-}e}$ of the same column are significantly different (P<0.05) from each other.

Table 3.2: Colour of bioplastics at different WS to CA mixture ratio

Sample	Colour			
	L*	a*	b*	ΔE
100 TO 0	75.57 ± 0.12[a]	15.57 ± 0.25[a]	5.53 ± 0.15[a]	12.3 ± 0.1[a]
75 TO 25	77.60 ± 0.10[b]	20.53 ± 0.21[b]	1.13 ± 0.06[b]	11.0 ± 0.2[b]
50 TO 50	78.63 ± 0.25[c]	19.43 ± 0.06[c]	0.87 ± 0.06[c]	9.5 ± 0.1[c]
25 TO 75	80.20 ± 0.10[d]	18.03 ± 0.06[d]	0.63 ± 0.06[d]	7.6 ± 0.1[d]
0 TO 100	82.43 ± 0.12[e]	17.17 ± 0.06[e]	0.27 ± 0.06[e]	5.9 ± 0.1[e]

L: Lightness (100)/ darkness (0), a*: redness (+)/ greenness (-), b*: yellowness (+)/ blueness (-), ΔE: Colour difference; Data reported are mean ± standard deviation (n = 3), and values of different letters [a-e] of the same column are significantly different (P<0.05) from each other.*

Table 3.3: Mechanical properties, water vapour permeability, and biodegradability of bioplastics at different WS to CA mixture ratio

	Characterization			
	TS (MPa)	EM (MPa)	WVP ($Kgs^{-1}m^{-1}Pa^{-1}$)	Biodegradability (days)
100 TO 0	7.91 ± 0.45[a]	20.93 ± 0.61[a]	4.69E-14 ± 1.46E-15[a]	15 ± 1
75 TO 25	6.78 ± 0.31[b]	16.47 ± 0.99[b]	3.80E-14 ± 7.82E-16[b]	15 ± 1
50 TO 50	5.20 ± 0.37[c]	11.42 ± 0.53[c]	3.33E-14 ± 1.00E-15[c]	15 ± 1
25 TO 75	4.13 ± 0.17[d]	8.78 ± 0.45[d]	2.64E-14 ± 1.21E-15[d]	15 ± 1
0 TO 100	3.76 ± 0.14[d]	6.65 ± 0.32[e]	2.22E-14 ± 6.57E-16[e]	15 ± 1

TS: Tensile strength, EM: Elastic Modulus, WVP: water vapour permeability; Data reported are mean ± standard deviation (n = 3), and values of different letters a-e of the same column are significantly different (P<0.05) from each other.

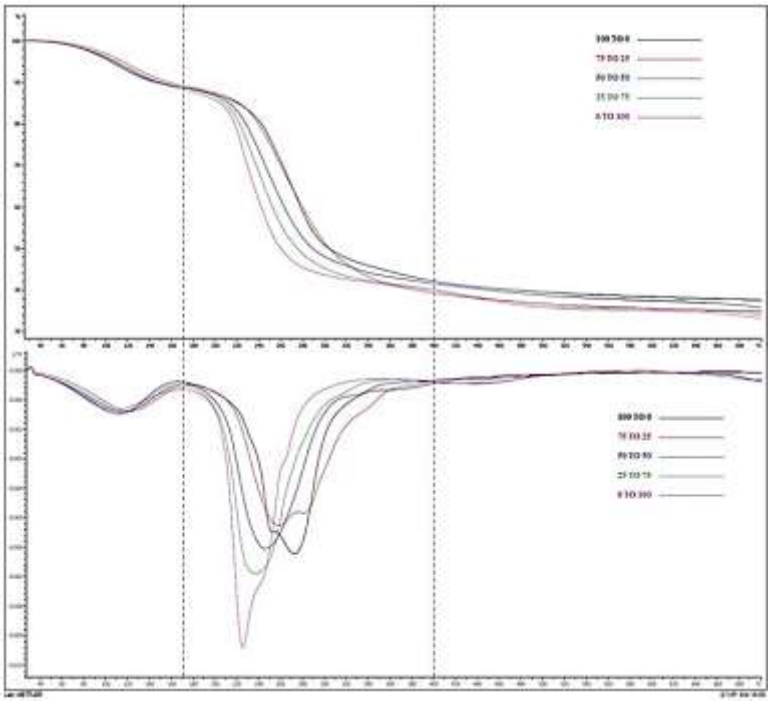


Figure 3.3: TGA/DTG graph of bioplastics at different WS to CA mixture ratio

4.0 Discussion

For the crosslinking of whole seaweed, *K. alvarezii* (WS), with commercial sodium alginate (CA), the ratio of WS to CA was set at 100:0, 75:25, 50:50, 25:75, and 0:100, respectively, in which bioplastics developed as shown in Figure 3.1, respectively. The biomass was dissolved in distilled water and glycerol was used as plasticizer. The bioplastics were developed using casting method.

4.1 Functional group

Functional groups of bioplastics and raw materials are important to identify changes before and after reactions and differences at varying WS/CA ratios, as shown in Figure 3.2. This is important as the addition of glycerol will cause plasticization, which will alter the peaks (Vieira et al., 2011). Figure 3.2 was analyzed and divided into a few sections with explanations after referring to multiple previous studies (Arzani et al., 2020; Dewi et al., 2015; Distantina et al., 2011; Naseri et al., 2019; Paula et al., 2015; L. Pereira et al., 2009).

Firstly, section (a) ranged between 3255.02 – 3369.12 is the hydroxyl (-OH) stretching, which was present for all the samples. Next, at section (b) is the -CH stretching which was present for glycerol at 2932.94 and 2880.50 but not intense in WS and CA. For the bioplastics, since all of them were added at the same concentration of glycerol, they each have the -CH stretching peaks which ranged between 2933.93 – 2939.17. At section (c), it shows peaks for COOH bonding, where it also showed that the intensity was more for CA and weaker for *WS* and no peak found on FTIR spectrum of glycerol. Bioplastics also showed that as more CA ratio, the more intense the peak.

Next at section (d), CA showed a peak at 1407.35 for the C-O-H bond, while glycerol also showed the same peak but with less intensity at 1412.05. Since WS showed no peak in that section, the 100:0 C-O-H peak was less intense than the rest but had a peak due to glycerol addition. Plus, as more alginate content was added, the peak also intensified, ranging from 1409.09 – 1415.97. For section (e), WS showed a peak at 1224.16 that represented the S=O bond, which was also present in glycerol at 1210.74 but not in CA. The peak was most intense at 100:0, which decreased with the addition of alginate which ranged from 1219.88 – 1227.66, and finally showed no peak at 25:75 and 0:100. Following that, the peaks in section (f) represent merging peaks of the C-O-C bond and mannuronic acid group for CA and the glycosidic bond for WS and glycerol. The presence of these groups was also found in the bioplastics' spectrum, which ranged from 1027.88 to 1033.84. Next, sections (g) and (h) represented the OH deformation and sulphate groups of the samples, respectively.

4.2 Moisture content, Thickness, Opacity and Colour

Data resulted from the study are tabulated in Table 3.1, which showed there were no significant differences on the moisture content and thickness of the bioplastics developed, which ranged between 23.3 ± 0.4% – 23.5 ± 0.5% and 0.050 ± 0.04mm – 0.054 ± 0.05mm, respectively, but their opacity was significantly different from each other. For their opacity, their trend is 100:0 > 75:25 > 50:50 > 25:75 > 0:100, their opacity decreases and becomes more transparent as they are lower in WS concentration and have higher CA content. The increase in transparency as more alginate is added is directly linked to the homogeneity of the mixture (Paula et al., 2015). On the other hand, since whole seaweed of *K. alvarezii* was used, there were more impurities, such as,

fibers, minerals, and others (Farah Nurshahida et al., 2020), which were directly linked to the opaqueness of the bioplastics with higher WS content.

Next, their colour can be described numerically according to CIE L*a*b* colour coordinate space, in which their coordinates can be translated to colour, where L represents lightness (100) and darkness (0), a* represents redness (+) and greenness (-), and b* represents yellowness (+) and blueness (-). To simplify, the bioplastics trend for L* and b* was 100:0 > 75:25 > 50:50 > 25:75 > 0:100, which means that going down the trend, the bioplastics were brighter and less yellow. Meanwhile, a* showed the trend of 75:25 > 50:50 > 25:75 > 0:100 > 100:0, meaning that 100:0 was less red compared to the rest, and as more alginate was added, the redness also decreased closer to 0. The colour difference in this study was in comparison to a white background, which had L*a*b* values of 83.8, 12.4, and -3.0, respectively.

For the colour difference (ΔE), the purpose was to compare if there were major differences compared to the white background, where the trend was similar as to their opacity. However, due to the transparency of the bioplastics, the colour difference resulted in the alteration of the white background colour when the respective bioplastics were placed on top of it. The mixing of biomass in the bioplastics' mixture causes deformity in the crystallization in the polymer matrix, which allowed more light penetration through the bioplastic and consequently resulted in lower opacity and colour difference (Farhan & Hani, 2017).

4.3 Mechanical, Water Vapour Barrier, Thermal and Biodegradability Properties

The outcome of the study on their mechanical, barrier, and biodegradability properties is tabulated in Table 3.3. The trends for tensile strength (TS), elastic modulus (EM), water vapour permeability (WVP), and thermal stability were 100:0 > 75:25 > 50:50 > 25:75 > 0:100. Their biodegradability properties, on the other hand showed no difference among them.

For their tensile strength, the outcome of this study of 7.91 ± 0.45 MPa for 100:0 was higher than the previous study on *K. alvarezii* films by Siah et al. (2014) at 6.82 MPa but lower than the studies by Sudhakar et al. (2020) and Ili Balqis et al. (2017) at 13.78 MPa and 69.69 MPa, respectively. However, the 0:100 resulted at 3.76 ± 0.14 MPa was much weaker than past studies that develop alginate-based bioplastics due to lack of additives, where with calcium chloride resulted 31 MPa (Paşcalau et al., 2012), with addition of pectin at 22.5 – 42.3 MPa (Galus & Lenart, 2013), and others. The 100:0 were the strongest in terms of load bearing strength, and as alginate was added, the tensile strength decreased. This addition of alginate into the *K. alvarezii* mixture disrupted the homogeneity, which led to the decrease in tensile strength following the trend (Ching et al., 2017). There were some previous studies that proved alginate-based films themselves are very homogenous and strong, but only with the addition of calcium chloride or with other enhancer (Abdullah et al., 2021), in which this study did not add any.

As for the elastic modulus (EM), higher value of the elastic modulus means higher rigidity. The most elastic among the bioplastics was 0:100 which resulted at 6.65 ± 0.32 MPa and the least was 100:0 at 20.93 ± 0.61 MPa. As for the rest, with more alginate into the film, their elasticity was indeed improved which is due to the disrupted homogeneity in the polymer matrix from the different type of biomass that reduces the rigidity among the particles (Paşcalau et al., 2012)`. Next, their water vapour permeability also improved when more alginate

concentration was in the mixture. This is because although they are all hydrophilic by nature, alginate is more hydrophobic compared to other hydrocolloids from seaweed (Castro-Yobal et al., 2021). Hence, bioplastics with higher alginate content showed better protection from water vapour.

For thermal stability, the resulting graph is shown in Figure 3.3. The first section dip at 25 – 120°C represents the loss in weight for evaporation of moisture absorbed, followed by the next section, which represents the degradation of glycosidic bonds in cellulosic parts, decarboxylation, decarbonylation, and hydration of alginate, which was around 170 - 400°C (Azucena Castro-Yobal et al., 2021; R. Pereira et al., 2013). This shows that 100:0 is most stable among them, with a degradation peak at 270°C, and 0:100 is the least stable at a lower temperature degradation peak at 223°C. This resulted in that trend because the structure became less compact as more alginate was added, which decreased the material's stability (Paşcalau et al., 2012). From Figure 3.3, there is a slight shift between the overlapping graphs, the graph shifting towards the right indicates higher thermal stability (Doh & Whiteside, 2020). The outcome of thermal stability helped supported the outcome of the mechanical and barrier properties.

On the other hand, the biodegradability of the bioplastics showed no difference between them as they were highly degradable. All the bioplastics were fully degraded between 14 -16 days when buried in soil with 40% RH. This showed that although all the other properties, in terms of mechanical, barrier, and thermal properties, were significantly different among them, their biodegradability was not affected.

Conclusion

To conclude, this study focused on developing WS (*K. alvarezii*) to CA (commercial sodium alginate) at different ratio at 100:0, 75:25, 50:50, 25:75, and 0:100. To summarize the outcome, the opacity, colour difference, tensile strength, elastic modulus, water vapour permeability, and thermal stability resulted in a trend of 100:0 > 75:25 > 50:50 > 25:75 > 0:100. The addition of alginate to the mixture disrupted the stability of the *K. alvarezii*-based bioplastics and vice versa. They still showed their potential to replace single-use plastic wrap at the right ratio. Low water vapour permeability indicates improvement in its barrier properties, which are equivalent to its protecting ability. The suggested future work from this point forward includes studying the possibility of improving more on their barrier and mechanical properties as they will be applied in the food industry, which will directly help extend the food preservation time.

Declaration

Funding

This study was funded by the Ministry of Higher Education of Malaysia (MoHE) through FRGS0562-1/2021 (Project ID:20518; Reference code: FRGS/1/2021/STG04/UMS/02/2)

Authors Contribution

Eunice Lua Hanry is the postgraduate student, who conducted the research and wrote the first draft. Noumie Surugau is the supervisor of this study, who was in charge of revising the draft and approved the final version of the paper before it is submitted for publishing.

Acknowledgement
We would like to thank Universiti Malaysia Sabah for providing the laboratory facilities and instruments to conduct this study.

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