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Optimization of biomass-to-water ratio and glycerol content to develop bioplastics from whole seaweed, Kappaphycus alvarezii

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Abstract

Many reports have described the development and properties of biodegradable plastics made from seaweed hydrocolloids, namely carrageenan, alginate, and agar. Although there is much potential, these seaweed extracts are expensive and require lengthy chemical treatment. In this study, the development of bioplastics using the whole biomass of red seaweed (*Kappaphycus alvarezii*) as base material, focusing on the optimization of the ratio of biomass (B) to water (W) and plasticizer (glycerol) concentration. The ratios (B: W) were 1:50, 1:60, 1:70, 1:80, and 1:90 (w/v), whereas glycerol was 1, 2, 3, 4 and 5% (v/v). The casting method was used for producing the bioplastics, and their physicochemical properties were tested using standard methods. As the water volume increases, tensile strength, colour intensity and opacity decrease while elongation-at-break and water vapour permeability increase. Similar trends were observed for the effect of increasing glycerol. All the bioplastics were fully degraded via soil burial within two weeks, and their shelf life lasted more than 14 weeks. Interestingly, the bioplastics developed at varied B: W ratios and glycerol concentration exhibit varied features that offer various applications, particularly food packaging.

1.0 Introduction

Seaweed-based bioplastics include those made from seaweed derivatives, either whole seaweed or extracts such as alginate, carrageenan, agar, and others. Seaweeds as a resource for bioplastic production are gaining popularity due to their many advantages, such as short cultivation and harvesting time, not posing land-use issues with other crops and being rich in nutrients (Balina et al., 2017). Most of the previous works utilized hydrocolloids from seaweed, namely carrageenan (Praseptiangga et al., 2022; Shojaee-Aliabadi et al., 2014), alginate (Mushollaeni & Rusdiana, 2011; Pereira et al., 2013), and agar (Elhefian et al., 2018; Hii et al., 2016; Rhim & Wang, 2013). However, using these hydrocolloids cost more as they go through a long process of treatment and purification compared to whole raw seaweed (Rhein-Knudsen & Meyer, 2017). There are some reports using whole seaweed by Siah et al. (2014), Sudhakar et al. (2021), Hettiarachchi et al. (2020) and a few others but still limited.

The properties of bioplastics are usually determined by their base material and type of plasticizer. For instance, bioplastics made of protein and polysaccharides exhibit good gas barrier properties but not for vapour (Wang et al., 2007). Other examples include past studies on seaweed hydrocolloid-based bioplastics, which showed good lipid and gas barriers but poor moisture barriers. They showed that they help prevent lipid retardation and preserve food moisture by sacrificing their own, which helps enhance food flavour, texture and batter adhesion (Lacroix & Tien, 2005). They have also been applied as antimicrobial packaging because they can transport antimicrobials and incorporate antioxidants, which also help prevent off-flavour during storage (Cha et al., 2002; Dussault et al., 2015; Lacroix & Tien, 2005).

In previous studies, the ratio of biomass to water and glycerol concentration was varied. For instance, B: W ratio suggested by Sudhakar et al. (2020) was 3–5g in 100.0 mL, Rahmawati et al. (2019) used 1.5% (w/v), and many others. As for the glycerol concentration, Tong et al. (2013) used 7–25% (w/w), Asria et

al. (2015) used 1–4% (v/v), and many others. Each study varied in the mixture and resulted differently, the same as other studies not mentioned. Hence, optimizing the mixture is essential to further the study. Preliminary studies using the 1:50 B: W ratio mixture (suggested by other studies) were too viscous to develop bioplastics by casting method. Glycerol concentrations suggested by each study were all different and varied in the method of concentration calculation, causing a dilemma as to which study to refer to in furthering the study. This study aims to determine the optimum B: W ratio and glycerol concentration to develop *K. alvarezii*-based bioplastics by comparing each characteristic affected by the B: W ratio and glycerol content, which will be the reference for future studies.

2.0 Materials And Methodology

2.1 Materials

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

2.2 Development of seaweed-based bioplastics

As Rahmawati et al. (2019) suggested, the development method was adapted and modified to fit this study. The optimization of glycerol content (1–5% (v/v)) and B: W ratio (1:50–1:90) was conducted. 1g of *K. alvarezii* was weighed and dissolved into 50.0–90.0 mL of distilled water (according to the 1:50–1:90 (w/v) ratio). The solution was heated to 60°C and remained for 30 minutes with continuous stirring. Glycerol was added at 1–5% (v/v), respectively, while continued being heated to 80°C. The mixture was stirred for one minute for bubble removal before being cast in a petri dish and dried in a ventilated drier at 50°C for 24 hours. Dried samples were carefully peeled from the cast resulting in bioplastics in Figs. 3.1 and 3.2 for different glycerol content and B: W ratio, respectively, and labelled as listed in Table 2.1

Table 2.1
Labeling for bioplastics developed at different glycerol content and B: W ratio

Different glycerol content, developed at 1:50 B: W ratio	Different B: W ratio developed at 1% (v/v) glycerol content
WS-1%	WS-1:50
WS-2%	WS-1:60
WS-3%	WS-1:70
WS-4%	WS-1:80
WS-5%	WS-1:90

2.3 Characterization

2.3.1 Functional Group Analysis, Moisture Content and Physical Properties

Firstly, functional groups were determined using ATR-FTIR (Agilent Technologies).In the characterization of bioplastics, the thickness was initially measured using a digital calliper. The moisture content was determined using Eq. 2.1 after being heated in the oven at 105°C for 24 hours, while their appearance, in terms of opacity and colour difference, was determined using a UV-Visible spectrophotometer (Agilent Technologies) and colour reader (Konika Minolta) and calculated using Eq. 2.2 and 2.3, respectively.

Moisture Content (%) = $\frac{InitialWeight-FinalWeight}{InitialWeight} X100\%$ (2.1)

Opacity (O) = $\frac{Absorbance}{FilmThickness}$ (2.2)

Colour Difference (ΔE) = $\sqrt{((L-L*)^2+(a-a*)^2+(b-b*)^2)}$ (2.3)

Where, L = 85.7, a = 6.4, b = -2.1

2.3.3 Mechanical and Barrier Properties

As for the mechanical and barrier properties, samples were conditioned 72 hours prior to use to 53% RH using magnesium nitrate hexahydrate ($Mg(NO_3)_2.6H_2O$) according to ASTM D-618 (ASTM, 2002). For mechanical properties, the samples were cut into strips with measurements of 90mm x 15mm and tested with a 5 mm/min pulling rate with a 50mm gauge on the Universal Tensile Machine at (GoTech Instruments).

The water vapour permeability test was carried out using the apparatus depicted in Fig. 2.2. The bioplastic was placed on the permeation cup in a desiccator with two relative humidity (RH) mediums. Water in the desiccator is 100% RH, and silica in the cup is 0% RH. Prior to use, the silica gel was heated for three hours at 100°C. The weight change data were noted hourly for 6 hours and calculated using Eq. 2.4.

WVP = $\frac{Changeinweight x Thickness}{Areaexposedfilms x Time x Vapourpressure}$ (2.4)

2.3.4 Biodegradability and Sustainability

Biodegradability was tested using the soil burial method suggested by Chuensangjun et al. (2013). The samples were checked daily to observe the days taken to complete degradation. The bioplastics were cut into a uniform size of 50mm x 50mm and buried 80mm into the soil, where the condition was set to 40% RH. The sustainability test was performed to see changes in the appearance of the films over six months (24–26 weeks) at predetermined conditions. In this study, the bioplastic films were stored in three conditions, namely: (i) inside an air-tight container, (ii) inside a closed folder and (iii) exposed on a table at 25^{o}C and 56% RH.

3.0 Results

Table 3.1: Moisture content, thickness, opacity, and colour difference of bioplastics at different glycerol content and B: W ratio

Sample	Characterization			
	MC (%)	T (mm)	O (cm^{-1})	ΔE
WS-1%	27.9 ± 0.3^{a}	0.071 ± 0.008^{a}	4.19 ± 0.84^{a}	30.6 ± 0.1^{a}
WS-2%	28.4 ± 0.0^{a}	0.074 ± 0.010^{a}	3.91 ± 0.33^{a}	28.4 ± 0.1^{b}
WS-3%	30.9 ± 0.5^{ab}	0.086 ± 0.007^{a}	3.47 ± 0.01^{a}	24.3 ± 0.0^{c}
WS-4%	33.2 ± 0.3^{bc}	0.089 ± 0.014^{a}	2.75 ± 0.68^{ab}	23.1 ± 0.1^{d}
WS-5%	35.7 ± 0.4^{c}	0.100 ± 0.012^{a}	2.07 ± 0.20^{b}	20.9 ± 0.1^{e}
WS-1:50	27.9 ± 0.3^{a}	0.071 ± 0.008^{a}	4.19 ± 0.84^{a}	30.6 ± 0.1^{a}
WS-1:60	31.4 ± 0.3^{ab}	0.076 ± 0.010^{a}	4.14 ± 0.45^{a}	29.6 ± 0.1^{b}
WS-1:70	32.6 ± 0.2^{b}	0.076 ± 0.011^{a}	3.80 ± 0.54^{ab}	27.2 ± 0.1^{c}
WS-1:80	35.5 ± 0.3^{c}	0.078 ± 0.012^{a}	3.17 ± 0.46^{bc}	23.4 ± 0.1^{d}
WS-1:90	38.4 ± 0.2^{d}	0.091 ± 0.014^{a}	2.43 ± 0.01^{c}	22.8 ± 0.1^{e}

MC: Moisture content, T: Thickness, O: Opacity, 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 3.2: Mechanical and barrier properties, biodegradability, and shelf life of bioplastics at different glycerol content and B: W ratio

Sample	Characterization						
	TS (MPa)	EAB (%)	WVP ($Kgs^{-1}m^{-1}Pa^{-1}$)	B (days)	Shelf life (Weeks)		
					E	SE	C
WS-1%	7.2 ± 0.9^{a}	14.7 ± 1.5^{a}	$3.85E\text{-}14 \pm 1.32E\text{-}15^{a}$	14.3 ± 0.6	14.0	15.0	26.0
WS-2%	5.4 ± 0.7^{b}	17.9 ± 2.0^{a}	$4.69E\text{-}14 \pm 1.46E\text{-}15^{ab}$	14.3 ± 0.6	15.0	16.0	26.0
WS-3%	4.8 ± 0.3^{b}	19.5 ± 2.5^{b}	$6.03E\text{-}14 \pm 2.23E\text{-}15^{b}$	14.3 ± 0.6	16.0	16.0	26.0
WS-4%	3.5 ± 0.5^{c}	20.9 ± 2.1^{b}	$7.58E\text{-}14 \pm 9.37E\text{-}16^{c}$	14.3 ± 0.6	16.0	16.0	26.0
WS-5%	2.6 ± 0.5^{c}	21.9 ± 1.1^{b}	$8.91E\text{-}14 \pm 8.65E\text{-}16^{d}$	14.3 ± 0.6	16.0	16.0	26.0
WS-1:50	7.2 ± 0.9^{a}	14.7 ± 1.5^{a}	$3.85E\text{-}14 \pm 1.32E\text{-}15^{a}$	14.3 ± 0.6	15.0	16.0	26.0
WS-1:60	5.3 ± 0.9^{b}	16.2 ± 1.5^{a}	$4.47E\text{-}14 \pm 7.37E\text{-}16^{b}$	14.3 ± 0.6	15.0	16.0	26.0
WS-1:70	3.7 ± 0.1^{c}	16.9 ± 1.1^{a}	$4.76E\text{-}14 \pm 6.02E\text{-}16^{c}$	14.3 ± 0.6	15.0	16.0	26.0
WS-1:80	2.8 ± 0.4^{cd}	17.1 ± 2.3^{a}	$6.21E\text{-}14 \pm 4.65E\text{-}16^{d}$	14.3 ± 0.6	15.0	16.0	26.0
WS-1:90	2.2 ± 0.7^{d}	17.7 ± 0.7^{a}	$7.65E\text{-}14 \pm 1.49E\text{-}15^{e}$	14.3 ± 0.6	15.0	16.0	26.0

TS: Tensile strength, EAB: Elongation at break, WVP: water vapour permeability; 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.

4.0 Discussion

This study focused on the optimization of the development mixture of seaweed-based bioplastic. The commonly used method involves mixing 50.0 mL of deionized water per gram of biomass and optimizing the plasticizer content according to the raw material used (Daza et al., 2018; Nazarudin et al., 2018; Rahmawati et al., 2019). Instead of solely focusing on optimizing the glycerol content, this study also optimized the B: W ratio, which on a large industrial scale could help save costs on development while giving approximately the same quality bioplastics. The primary raw material, *K. alvarezii*, was handled carefully to ensure the quality of the seaweed used in the development of bioplastics.

4.1 Optimization of glycerol content and B: W ratio of bioplastic development

The glycerol content must be optimized so there is no anti-plasticization effect on the bioplastic produced due to excess glycerol. Based on preliminary experiments (data not shown), the glycerol content tested ranged between 1–5% (v/v), where developed bioplastics are shown in Fig. 3.1. Following that, the B: W

ratio also needs to be optimized for a more favourable industrial development of seaweed-based bioplastics, where developed bioplastics are as shown in Fig. 3.2.

4.1.1 Functional groups

Compound functional groups are critical in determining the interaction during any reaction. Figure 3.3 is the FTIR spectrum from glycerol, which shows a peak at 3290.74 cm^{-1}, indicating hydroxyl (OH) stretching (a), followed by CH stretching (b) at 2932.61 cm^{-1} and 2879.48 cm^{-1}, respectively. The carboxyl group of C-OH bending was assumed to be 1415.76 cm^{-1} (c). The primary alcohol C-O stretch indicator is 1108.14 cm^{-1} (d). However, the C-O stretching of primary (c) to secondary (d) alcohol was indicated at 1415.76 cm^{-1} to 1108.14 cm^{-1}, respectively (Kongjao et al., 2016). Peaks at 993.36 cm^{-1}, 922.47 cm^{-1}, and 850.99 cm^{-1} (e) indicate the presence of C-C skeletal vibration (Guerrero et al., 2010).

The spectrum for *K. alvarezii* (Fig. 3.4) showed a broad peak of hydroxyl (OH) stretching at (a) 3351–3384 cm^{-1} and CH stretching at (b) with peaks between 2905–2920 cm^{-1}. Carbonyl groups (COOH and C-OH) (c) were detected at 1634–1639 cm^{-1}, while sulphate esters (d) were detected at 1227 – 1221 cm^{-1}. At 1060–1066 cm^{-1} and 922–924 cm^{-1}, the C-O-C bond of 3,6-anhydrogalactose rings (e) was shown. The glycosidic linkage of C-O-C and C = O of galactose (f) was observed in samples 1030–1037 cm^{-1} and 968–969 cm^{-1}. Peaks for galactose-4-sulphate (g), C = O-S, were observed at 844–845 cm^{-1}, indicating the presence of k-carrageenan. This proves that the whole seaweed has all the functional groups as k-carrageenan, to act as the base material for bioplastic development. This is supported by previous studies where k-carrageenan functional groups are aligned with the study's outcome (Arzani et al., 2020; Dewi et al., 2015; Distantina et al., 2011).

Figure 3.4 depicts peaks shifting at OH and CH stretch, COOH, C-OH, and C-O-C bonds in bioplastics spectrums. The S = O bond, second C-O-C peak, glycosidic link, and C = O-S bond remained in the same range before and after adding glycerol. Compared to their raw materials, there was a slight shift in the OH and CH stretch, C-OH bond, S = O bond, and C-O-C glycosidic link between those with and without glycerol. With seaweed, water also acts as an extracting solvent and a plasticizer, which explains the shifting of peaks between raw materials and bioplastics without adding glycerol (Naseri et al., 2019; Vieira et al., 2010). The presence of each glycerol functional group caused shifting because they are bound to change or shift some peaks due to the plasticizing reaction between biomass and glycerol (Vieira et al., 2010).

4.1.2 Moisture content and appearance

Table 3.1 shows that increasing the glycerol content increases the moisture content and thickness of the bioplastics. The moisture content of each bioplastics group increased with increasing glycerol content, from 1–5%. The outcome agreed with a study by Farhan & Hani (2017) on the impact of using different plasticizers and concentrations of glycerol and sorbitol. Also shown in Table 3.1, 1:50 to 1:90 also showed an increase in moisture content and thickness of bioplastics while decreasing their opacity and colour difference when a higher biomass-to-water ratio was used. Higher water molecule dispersion and

lower biomass in the film matrix loosen polymeric chain bonding, requiring more foreign molecules to fill the interstitial polymeric gaps, completing the film-forming matrix using water and glycerol molecules (Vieira et al., 2010). The thickness increased due to swelling from the increasing hydrophilic properties of bioplastics (Farhan & Hani, 2017). Then, the water molecules reduced the scattering of light, allowing higher penetration of light and producing lighter colour bioplastics (Zuraida et al., 2012).

The increase in water parts in the mixture resulted in bioplastics with higher moisture content for each group. When more water is added to the bioplastic mixture, the water interacts with the polymeric chain. It binds to bioplastics, increasing the amount of water in the bioplastic film-forming matrix (Ili Balqis et al., 2017). As with glycerol, adding water causes bioplastics to swell, increasing their thickness. Although the thickness of bioplastic increases as the proportion of water to bioplastic increases, its opacity decreases. This result is consistent with the findings of Farhan & Hani (2017) on carrageenan-based bioplastics and Daza et al. (2018) on edible films derived from ulluco starch. The colour difference follows the same pattern as the opacity, with a higher glycerol concentration resulting in less colour change, directly related to the bioplastics' opacity. Higher glycerol concentrations allowed for more significant deformation of crystallization of the bioplastic matrix, allowing for greater light transmission through the matrix, resulting in less opacity and colour difference (Farhan & Hani, 2017). This characteristic is crucial for aesthetic appeal and may affect the commercial value of seaweed bioplastic films.

4.1.3 Mechanical and Barrier properties

Table 3.2 summarizes the mechanical properties of the developed bioplastics films. When there was higher glycerol content, the tensile strength decreased while elasticity increased—the same for when a higher B: W ratio was used. The polymeric bond integrity weakens due to less polymeric material in the bioplastic matrix as more water is used in the bioplastics' production. The interaction that was supposed to be between polymeric chains was replaced by polymeric-to-water/glycerol molecule interaction instead (Farahnaky et al., 2013). The resulting data are in agreement with previous studies on the development of bioplastics using carrageenan (Rahmawati et al., 2019), semi-refined carrageenan (Ili Balqis et al., 2017) and corn starch (Gujar et al., 2014). Since there are higher glycerol and water content, the crystallinity of the film matrix was disrupted, weakening the polymeric chain bonding, which resulted in flexible but weak bioplastics (Abdullah et al., 2018; Vieira et al., 2010). The flexibility of bioplastics increases as polymeric chain bonding weakens because higher water parts in the film-forming mixture disrupt the bioplastic interstitial space, allowing water molecules to interact with polymeric chains and weaken their bond (Van Soest & Knooren, 1997).

Water vapour permeability (WVP) is critical in food packaging because higher WVP reduces the shelf life of bioplastic-preserved food. As a result, lower permeability is preferred in the food packaging industry because it allows for more extended storage periods (Avila et al., 2022). As shown in Table 3.2, increasing the glycerol content increases the permeability of the water vapour of bioplastics same goes for increasing the B: W ratio. Two significant factors could explain the study's findings. To begin with, adding glycerol to the bioplastic increases the void between the polymeric chains, lowering the binding force of the bioplastic in the film-forming matrix (Ili Balqis et al., 2017). This reduces their ability to prevent water

vapour molecules from passing through the bioplastics (Khazaei et al., 2014). Second, this phenomenon occurred due to the hydrophilic nature of the raw materials used, including seaweed, water, and glycerol, which means they will attract water if water is present nearby (Galus & Lenart, 2013). As a result, osmosis played an important role in water absorption during the WVP test, where the two opposite mediums induced the water transfer.

4.1.4 Biodegradability and Sustainability

As shown in Table 3.2, the increase in glycerol content and B: W ratio was insignificant in their biodegradability property, as for their shelf life which was observed at three different conditions, as shown in Table 3.2. The result shows that mould growth was faster on WS-3% < WS-4% < WS-5% < WS-2% < WS-1% in days. This happened because of the moisture content and the glycerol reaction after the bioplastics were developed. However, it is also highly dependent on their storage condition, as those stored in containers lasted more than the observation period of 26 weeks, while those exposed showed mould growth as soon as 14 weeks.

4.1.5 Selection of Bioplastic by characteristics

The optimal glycerol content and B: W ratio were chosen based on the evaluation of each studied characteristic. Figures 3.6 summarise the properties of each bioplastic group developed at various glycerol content (left) and B: W ratios (right). The data used to plot the graphs were converted from their actual value unit to percentage (%) to compare each characteristic objectively. Each graph plotted included moisture content, film thickness, colour difference, opacity, tensile strength, elongation at break, water vapour permeability, biodegradability, and shelf life for exposed, semi-exposed, and covered conditions, which aids in determining the desired properties. Figure 3.5(a) clearly shows that the bioplastic of 1% line produced the best and most balanced results for tested properties. Meanwhile, based on their summarized characteristics data in Fig. 3.5, the 1:60 ratio line was optimal for their B: W ratio (b). Because of the slight margin difference from 1:50, less biomass would be used in the production line to achieve the same quality.

4.2 Potential Application of Seaweed-based Bioplastics

This study aims to produce the optimum mixture to produce greener bioplastics at a lower cost, which can be the substitution of conventional petroleum plastics in hopes that it will help improve our environmental health. Figure 4.1 shows the current packaging of our daily-use products, an example of single-use plastics. Since they do not require greater mechanical properties, current study products are perfect for replacing these wrappers, as demonstrated in Fig. 4.2.

Conclusion

In conclusion, this study focused on developing seaweed-based bioplastics at different glycerol content and B: W ratio to determine the best mixture to develop seaweed-based bioplastics in advancing with this study. It was found that the optimum condition for the development of seaweed-based bioplastic was at 1:60 seaweed B: W ratio with 1% (v/v) glycerol, which resulted in good characteristics in terms of

appearance, mechanical properties, water vapour permeability, biodegradability and shelf life, to function for its selected purpose. Developing *K. alvarezii*- based bioplastics was an excellent alternative to replace one-time-used plastic wrappers requiring less tensile strength. Also, this is an alternative in helping the world fight global plastic pollution by going into a lower cost, greener, chemical- and waste-free bioplastic production. Further studies into enhancing mechanical and barrier properties also study incorporating antimicrobial properties as the way forward for this study.

Declarations

Data Availability

The authors declare that the data supporting the findings of this study are available within the paper. Should any raw data files be needed in another format they are available from the corresponding author upon reasonable request.

Conflict of Interest

The authors named in this manuscript, Eunice Lua Hanry and Noumie Surugau, is currently a postgraduate student and senior lecturer, respectively, at Universiti Malaysia Sabah. This study is funded by Ministry of Higher Education, Malaysia through grant FRGS0562-12021. However, listed authors are not involved in any organization or entity with any financial interest or non-financial interest in the subject matter discussed in this manuscript.

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Authors Contribution

Both authors conceived the concept. Eunice Lua Hanry generated the data and wrote the first draft of the main manuscript text. Noumie Surugau revised and edited the draft, proofread and approved the final version of the paper before it was submitted for publishing.

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Figures

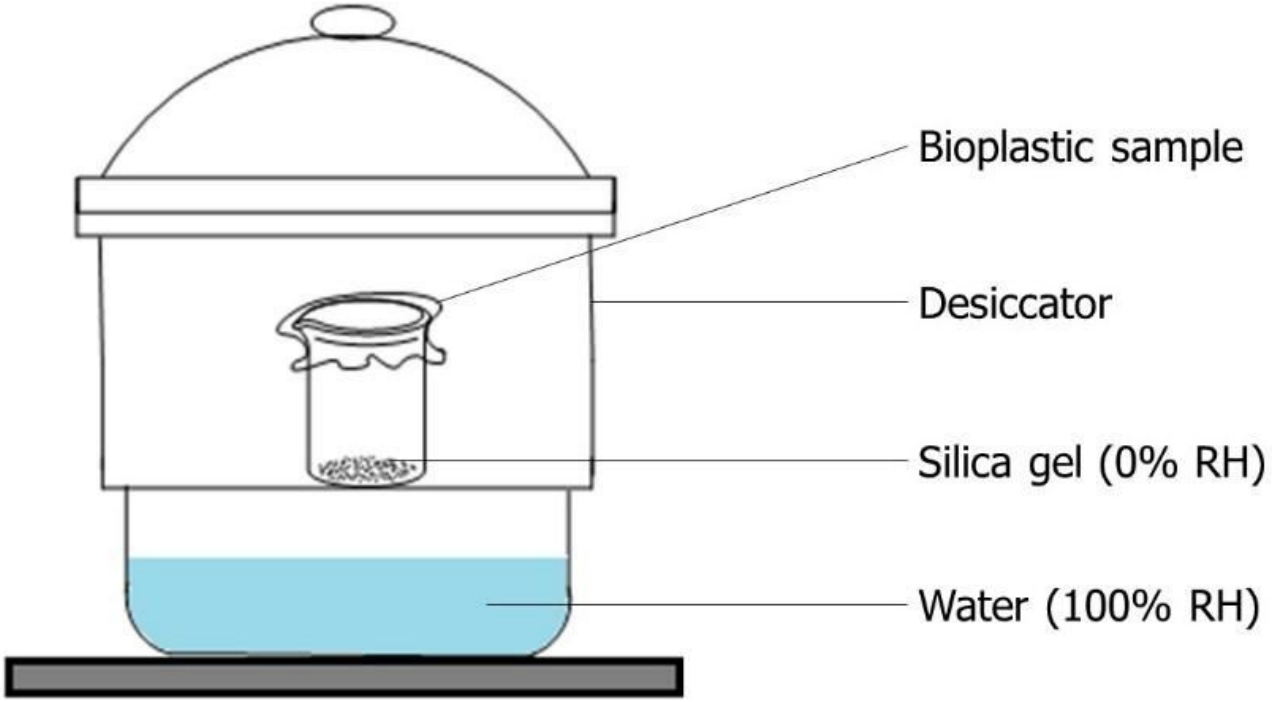


Figure 1

Water vapour permeability analysis set up.

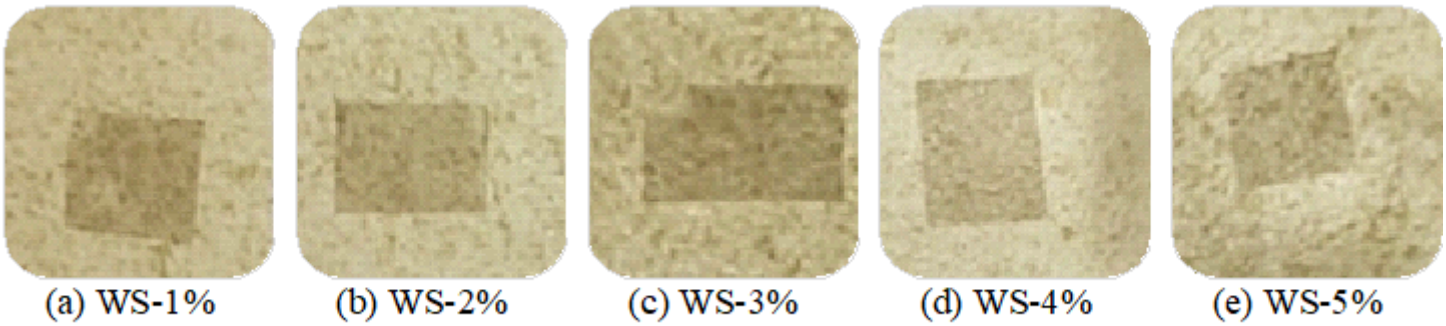


Figure 2

Bioplastics of 1:50 (w/v) B: W ratio at different glycerol content



Figure 3

Bioplastics with 1% (v/v) glycerol at different B: W ratio

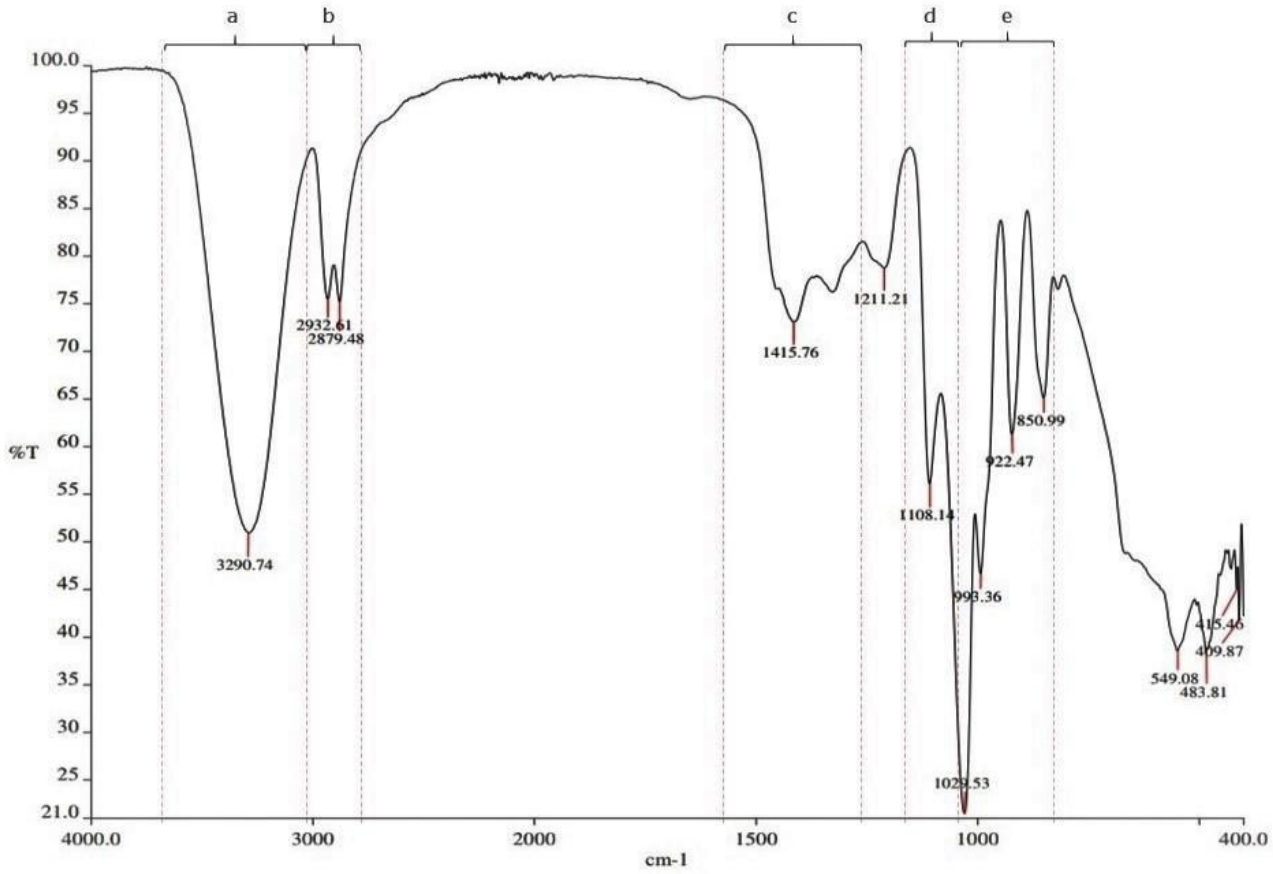


Figure 4

FTIR Spectrum of glycerol

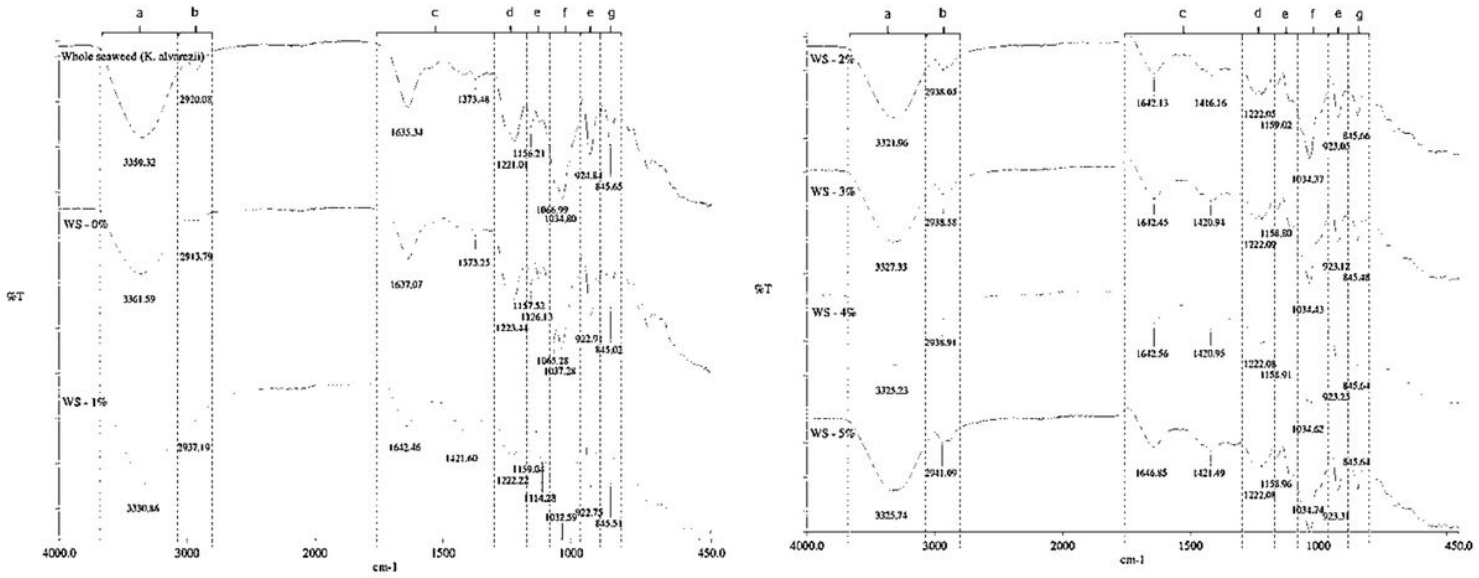


Figure 5

FTIR spectrum of *K. alvarezii* and bioplastics with 0 – 5% (v/v) glycerol

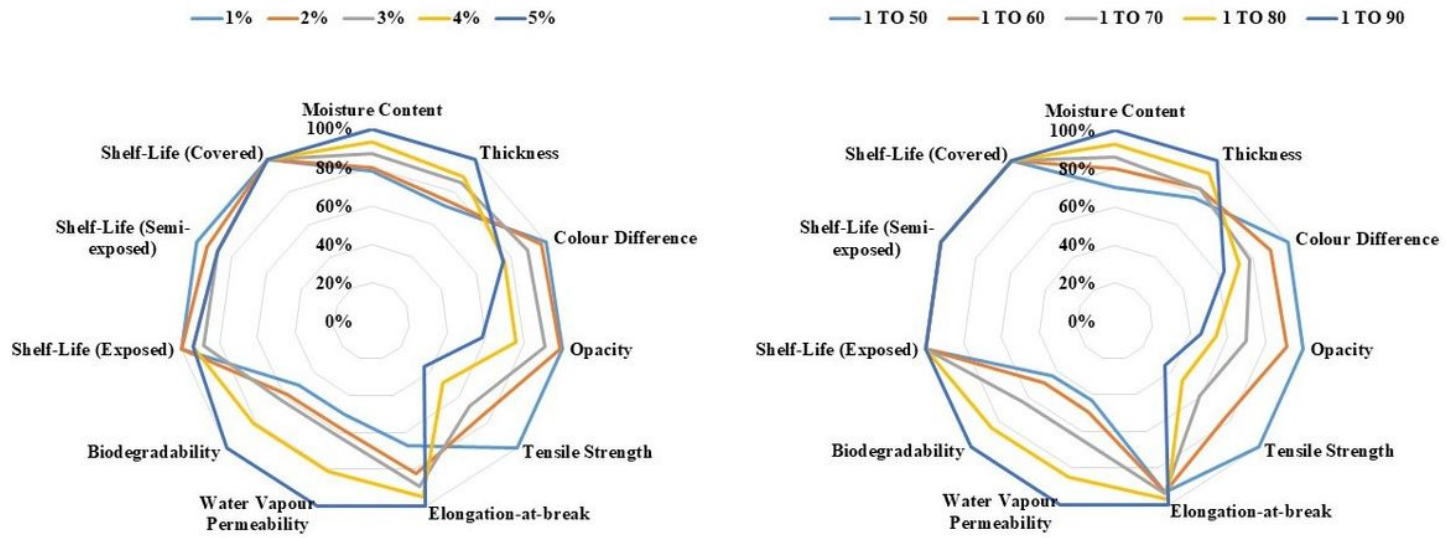
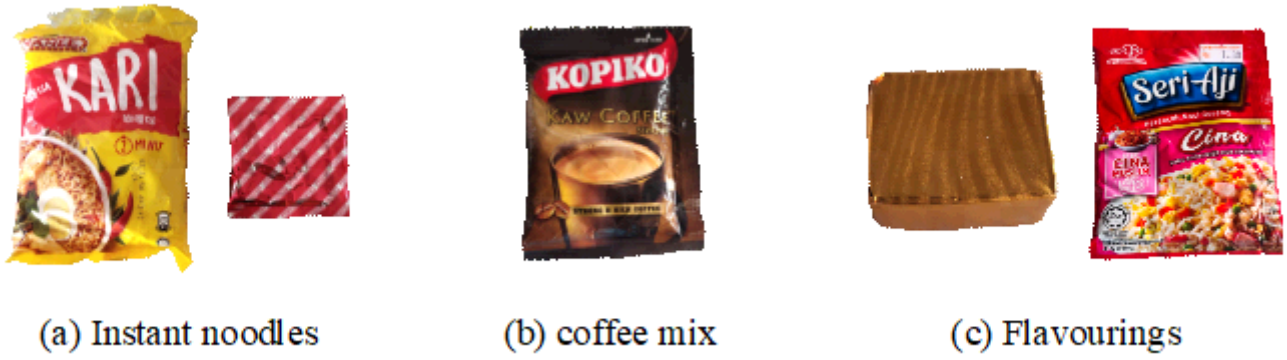


Figure 6

(a) Comparison of different glycerol content

(b) Comparison of different B: W ratio

Overview comparison of bioplastics' characteristics by percentage



(a) Instant noodles (b) coffee mix (c) Flavourings

Figure 7

Current one-time-use packaging



(a) Instant noodles (b) coffee mix (c) Flavourings

Figure 8

Demonstration of re-packaging using WS-bioplastics.