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Evaluation of bioplastic films made from seaweeds and its polymers

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Abstract

Seaweeds grow and produce a significant amount of biomass per unit area, providing specialized phytochemicals for the manufacturing of bioplastics, which may otherwise less affordable. In this study, the brown seaweed *Sargassum denticulatum* and the red *Corallina officinalis* were used to prepare bioplastic films from both whole algal powder and their extracted polymers using two methods: conventional and ultrasound-assisted extraction. Ultrasound proved to be more effective for extracting polymers, as evidenced by the yield of alginate, which was higher than that of carrageenan in both methods. The functional compositions of the films were evaluated using Fourier-Transform Infrared (FTIR) spectroscopy. Additionally, key properties such as solubility, moisture absorption resistance, elongation at break, tensile strength, and degradability were assessed. The resulting bioplastic films exhibited varying physical and mechanical properties, making each suitable for distinct applications based on its characteristics. Notably, the whole-algae films demonstrated higher solubility, superior mechanical properties, and faster degradation compared to the polymer-based films. Therefore, bioplastic films made from seaweeds and their extracted polysaccharides hold great potential as viable alternatives to synthetic plastics, contributing to the reduction of environmental pollution caused by petroleum-derived materials.

Keywords: *Sargassum*. *Corallina*. Polymers. Bioplastic. Ultrasound

Introduction

Plastic is indispensable in our daily lives; it is used in appliances, electrical equipment, medical devices, building, cars and packaging, making our lives more comfortable. Plastics typically consist of high-molecular-weight polymers derived from crude and fossil fuels **(Rajendran et al. 2012)**.

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Maheswari and Ahilandeswari (2011) mentioned that plastics can be divided into two types depending on their physical properties: thermoplastic and thermosetting. Thermoplastic can be easily deformed upon heating and repeatedly remolded, whereas thermosetting plastics cannot be remelted or reshaped once cured. Around 359 million metric tons of plastic were produced worldwide in 2018, and by 2050, this is predicted to triple, making up a fifth of the world's oil consumption **(Chia et al. 2020).** During production, industrial plastics require a lot of fossil fuels and emit a lot of dangerous greenhouse gases (**Shamsuddin et al. 2017**). According to **Wang and Nomura (2010)**, plastic waste is one of the most significant environmental issues facing the world because

it takes thousands of years for it to break down naturally. It also contributes to pollution of the soil, water, and air, distorts the landscape and reduces soil fertility. Furthermore, **Reyna-Bensusan et al. (2019)** showed that open burning of mixed wastes contributes significantly to air pollution and has a number of negative consequences on the environment and human health in addition to global warming. Because there are numerous types of plastics and each one requires a unique recycling procedure, recycling these polymers is particularly challenging.

Bioplastics are plastics made from renewable biological sources, including bacteria, algae, and plants. Microorganisms like fungus and bacteria found in the soil can break them down without releasing any pollutants (**Rajendran et al. 2012**). The size of the worldwide bioplastics market is expected to grow daily, from its current value of $ 9.2 billion to $ 20 billion by 2026 (**Nanda et al. 2022**). Due to their many benefits, including their high output and capacity to thrive in a variety of conditions, algae are a great feedstock for the manufacturing of plastic (**Greg Stevens 2010**). Seaweeds are a cost-effective plant that grow in saltwater rather than freshwater and doesn't require land or pesticides. Red algae (Rhodophyta), brown algae (Phaeophyta), and green algae (Chlorophyta) are the three main kinds of seaweeds (**Ditchburn and Carballeira 2019**).

Because of its gelling and film-forming qualities, seaweed's biochemical composition, which was rich in hydrocolloids (polysaccharides) including agar, carrageenan and alginate, makes it suitable for the creation of bioplastics (**Schmidtchen et al. 2022).** Other biopolymers found in seaweed, such cellulose and starch, may be used as raw materials to make bioplastics (**Romero-Vargas et al. 2023**). The brown seaweed's cell walls contain alginate, which contributes to the seaweed's elasticity (**McHugh 2003).** *Ascophyllum, Durvillaea, Ecklonia, Laminaria, Lessonia, Macrocystis, Sargassum* and *Turbinaria* species are the primary commercial sources of alginate (**Bixler & Porse 2011**). One common usage for brawn algae was the making of films; **Lim et al. (2018)** employed *S. siliquosum* as a raw material to create bioplastic film. For usage in packaging films, researchers have studied *Sargassum* sp. (**Kanagesan et al. 2022**), *Sargassum natans* (**Mohammed et al. 2023**), *Saccharina latissima* (**Ayala et al. 2024**), *Padina pavonica* and *Rugulopteryx okamurae* (**Torrejon et al. 2025**). Additionally, *Laminaria digitata* and and *Jolinay laminaioides* have been investigated for their potential in hydrogels and biomedical devices (**Berglund et al. 2020; Rasheed et al. 2023).**

On the other hand, a galactose-rich carbohydrate polymer that is economically significant, carrageenan is derived from a variety of red seaweed species (**Rafiquzzaman et al. 2016).** To form bioplastic, **Phan et al. (2005)** and **Hollingworth (2010)** used red seaweed while **Machmud et al. (2013)** used *Eucheuma cottonii*. According to research by **Sebaaly et al. (2014)** and **Sudhakar et al. (2022)**, carrageenan obtained from *Corallina* sp. and *Kappaphycus alvarezii*, respectively, is appropriate for the creation of bioplastic films. The potential use of several species, such as *Gracilaria salicornia, Gracilaria verrucosa, Kappaphycus* sp*.,* and *Eucheuma* sp., in packaging and reusable bags, as well as their mechanical strength and biodegradability in the creation of bioplastic films, have been investigated (**Darni et al. 2019; Rizal et al. 2021; Ghobashy et al. 2023; Hanry 2023; Kadell and Callychurn 2023; Hanry and Surugau 2024; Leong et al. 2024).**

The present study aimed to extract two biopolymers -"carrageenan from *Corallina officinalis* and alginate from *Sargassum denticulatum*" - using two extraction methods.

Additionally, it involved manufacturing biodegradable plastic films from the algae as well as from the extracted polymers, followed by evaluating and comparing their physicochemical properties.

Materials and Methods

- **Collection and identification of seaweeds:**

One seaweed was collected from each of the following: red alga /*Corallina* sp. from Alexandria (31°12'18.2"N, 29°53'09.7"E) and brown alga /*Sargassum* sp. from Ras Sedr (29°37'20.4"N, 32°41'16.7"E) as shown in plates 1 and 2. To eliminate undesirable material, salts, and epiphytes, the seaweeds were thoroughly cleaned with distilled water. After being shade-dried for three days at room temperature and then dried in an oven at 60°C until the weight remained constant, the seaweeds were crushed and kept dry **(Sudhakar et al. 2021)**. The algae *Corallina officinalis* and *Sargassum denticulatum* were identification after verification through **Aleem (1978 and 1993)**; **Lipkin and Silva (2002)** by Prof**.** Wafaa S. Abou El-kheir.

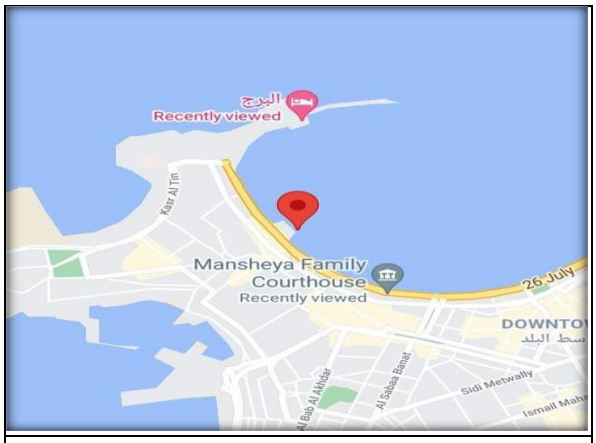


Plate 1: Google Earth image illustrating the collection site of *C. officinalis* in Alexandria.



Plate 2: Google Earth image illustrating the collection site of *S. denticulatum* in Ras Sedr**.**

- **Extraction of polymers from seaweeds:**

1- Conventional method:

1.1. Carrageenan extraction from *C. officinalis*: The algal powder was rehydrated for 12 h in distilled water to eliminate the water-soluble fraction , followed by immersion in a methanol / acetone (1:1 v/v) mixture to eliminate organic soluble fraction. The treated biomass was then placed in 150 mLof 1 M NaOH at 80 °C for 3 h (alkaline treatment) and the pH was adjusted to 8. Hot filtration was performed twice, after which the extracted polymer was dissolved in 60 ml of distilled water and subsequently precipitated by adding ethanol (96%) **(Pereira et al. 2003)**.

1.2. Alginate extraction from *S. denticulatum*: Alginate was extracted according to **Saji et al. (2022)** with a slight modifications. The dried algal biomass was soaked in 2% formaldehyde overnight. The following day, 1% HCl was added to the residue and the mixture was put in oven at 50°C for 2 h. Next, alginate was extracted using 4% Na_2CO_3 for 2 h in oven at 50°C. The liquid fraction was collected and precipitated with 95% ethanol. Finally, the resulting solid precipitate was dried in an oven at 60°C.

2- Ultrasound-Assisted method:

Carrageenan and alginate were extracted using ultrasonic LC 60 H, ELMA in the Desert Research Center (Mataria, Cairo, Egypt) **(Martín-del-Campo et al. 2021).**

- **Calculations of polymers yield:**

All polymers were calculated as a percentage, according to **Reis et al. (2011).**

$$\text{Yield of polymer (\%)} = \frac{Wc}{Ws} \times 100$$

Where Wc is the weight (g) of dried polymer and Ws is the initial weight (g) of dried seaweed used before extraction.

- **Characterization of the extracted polymers by FTIR analysis:**

FTIR measurements of carrageenan and alginate were done at the National Center for Radiation Research and Technology (NCRRT), Atomic Energy Authority (AEA), Egypt. Spectra were recorded on a JASCO FTIR -3600 spectrometer and data were collected at a resolution of 4 cm^{-1} over the wavenumber region of 400 to 4000 cm^{-1}. The FTIR spectra of the extracted polymers were compared with those of the corresponding commercial standards.

- **Bioplastic formation from seaweeds:**

Bioplastic films were prepared from algal biomass according to **Sabathini et al. (2018).** First, 2.8 g of *S. denticulatum* and *C. officinalis* powders were separately suspended in 60 mL of distilled water in a flask and mixed using a magnetic stirrer. In another flask, 2.8 g of polyvinyl alcohol (PVA) was dissolved in 60 mL of distilled water at 80°C for 30 minutes. The contents of both flasks were then combined into 1g of citric acid and 2.11 g glycerol under continuous heating at 90°C. The resulting solutions were cast into silicone molds and dried for 24 hours at room temperature. The dried films were carefully peeled from the molds to obtain the final bioplastic films.

- **Bioplastic formation from polymers:**

About 6.8 g of corn starch was added to 240 mL of distilled water and placed in a water bath (90 °C) with constant stirring. A fixed weight (1.2 g) of the previously extracted polymers (carrageenan and alginate) + 2 g of glycerol was added to the homogenized starch solution and stirred for 5 minutes. Each solution was poured into a silicone mold and allowed to dry at room temperature until the next day **(Wu et al. 2009)**.

- **Bioplastics characterization:**

The following properties were tested for each bioplastic films (from two algae and four polymers):

1. Physical & Optical properties

1.1. Color and Texture: The color and texture of each bioplastic films were assessed through visual inspection and tactile examination.

1.2. Solubility by solvent: The solvents used were acetone, ethanol and chloroform. The bioplastic films were cut into small pieces and 0.3 g of each piece was placed in test tubes containing 3 mL of each solvent. The test tubes were divided into two groups: one group was kept at 30°C, while the other was incubated at 60°C. The solubility of the films was assessed after one hour **(Tun and Mar 2020)**.

1.3. Moisture absorption: The bioplastic films were dried in a desiccator until a constant weight (W_1) was achieved. They were then exposed to normal atmospheric conditions for 24 h and reweighed (W_2) (**Tun and Mar 2020**). The moisture absorption percentage was measured at NCRRT, AEA, Egypt, and determined using the following formula:

$$\text{Moisture Absorption (\%)} = \frac{(W2-W1)}{W1} \times 100$$

2. Mechanical properties:

The tensile strength (TS) and elongation at break (EAB) of the bioplastic films were measured using a universal testing machine (Tinius Olsen H10KS, Horsham, PA, USA) at NCRRT, AEA, Egypt.

3. Structural & Morphological Properties

3.1 Scan electron microscope (SEM): SEM was used to assess the surface morphology of bioplastic films. The samples were viewed at 500x magnification using the JEOL-JSM-5400 scanning electron microscope at NCRRT, AEA, Egypt.

3.2 Fourier Transform Infrared Spectroscopy (FTIR): FTIR analysis of all bioplastic films were performed with the same equipment and procedures as previously mentioned.

3.3 X-Ray diffraction (XRD): X-Ray diffraction analysis was done at NCRRT, AEA, Egypt, to all the formed bioplastic films. Sample was used XRD- diffractometer with Cu target at 40 KV and 30 mA. The XRD runs drive axis over θ-2θ at a Scan speed 8.0000 (deg/min) sampling pitch 0.02.

4. Degradability

4.1 By soil: In the constant weight of clay and sandy soils, a constant weight of all bioplastic films were buried under the surface of it and noticed for 30 days **(Nguyen et al. 2016).** The following calculation was used to compute the films weight loss percentage (WL %).

$$WL\% = \frac{\text{initial wt.of the film} - \text{wt.of the film after degradation}}{\text{Initial wt.of the film}} \times 100$$

4.2 By water: According to **Naesa et al. (2019)**, the degradation test was conducted by placing a constant weight of all bioplastic films in different water media (distilled water, acidic water and alkaline water). Degradation occurred over 14 days and was measured every 7 days using the following equation:

$$WL\% = \frac{\text{initial wt.of the film} - \text{wt.of the film after degradation}}{\text{Initial wt.of the film}} \times 100$$

Statistical Analysis

All data were statistically analyzed three independent biological replicates for each treatment and are presented as mean ± SD (standard deviation). Differences between means were detected by one-way analysis of variance (ANOVA). Significant differences among variable treatments were identified using Duncan's Multiple Range Test (DMRT) conducted with Statistical Package for the Social Science (SPSS) software 16.0 (SPSS Inc., USA).The level of significance was determined at $p < 0.001$.

Results

Extraction polymers from seaweeds:

Extraction yields of biopolymers vary depending on the type of seaweed and the extraction method. **Figure 1** illustrates the difference in polymer yield (carrageenan and alginate) extracted from seaweeds by using conventional and ultrasound methods. The highest carrageenan rate extracted by using the ultrasound (U-carrageenan) was 6.6% compared to 3.7% by using the conventional method (C- carrageenan) for *C. officinalis*. A similar trend was observed in the extraction of alginate from *S. denticulatum*, where the polymer extraction rate by using ultrasound (U-alginate) was 51.8%, compared to 16.9% by using the conventional method (C-alginate).

Characterization of extracted polymers by FTIR analysis:

1. Carrageenan from *C. officinalis*:

A comparison was conducted between commercial, as a standard, (**Figure 2**) and extracted carrageenan (**Figure 3, a & b**) using FTIR analysis. Though the overall spectral pattern was similar with commercial carrageenan, some simple differences were observed in the height and shape of certain absorption bands. In **Figure 3 (a & b)**, the peaks region were found at 871 cm-^{1}, 872 cm-^{1} and 923 cm-^{1}. Some bands were observed at 1008 cm-^{1}, 1032 cm-^{1}, 1543 cm-^{1} and 1645 cm-^{1}. FTIR spectroscopy also revealed the presence of very strong absorption bands in the ranges of 2017 cm-^{1} to 2850 cm-^{1} and 3036 cm-^{1} to 3626 cm-^{1}.

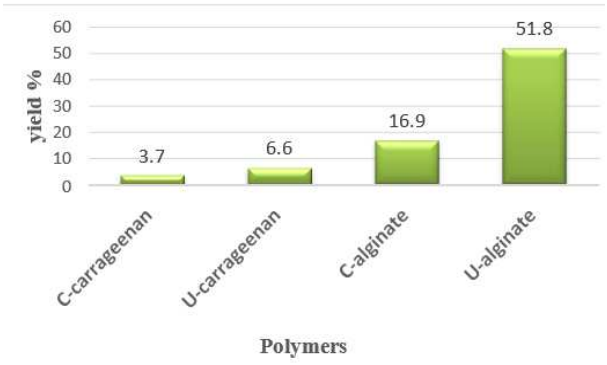


Fig. 1: Polymers content in 2 seaweeds

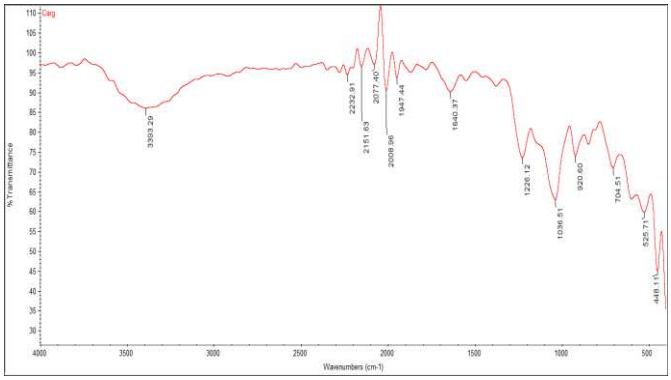


Fig. 2: FTIR spectrum of commercial carrageenan

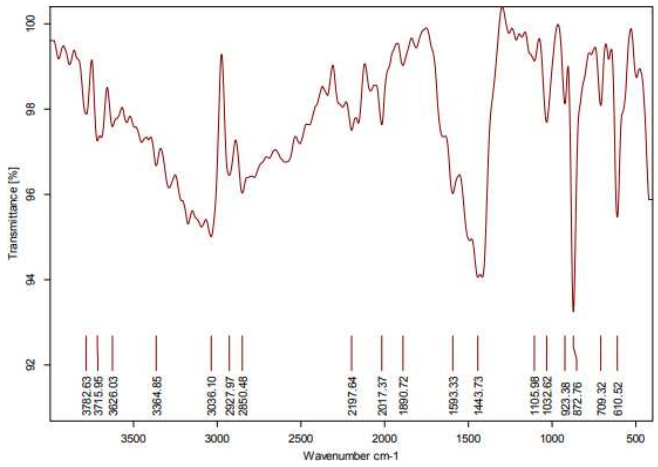


Fig. 3a: FTIR spectrum of C-carrageenan extracted from *C. officinalis*

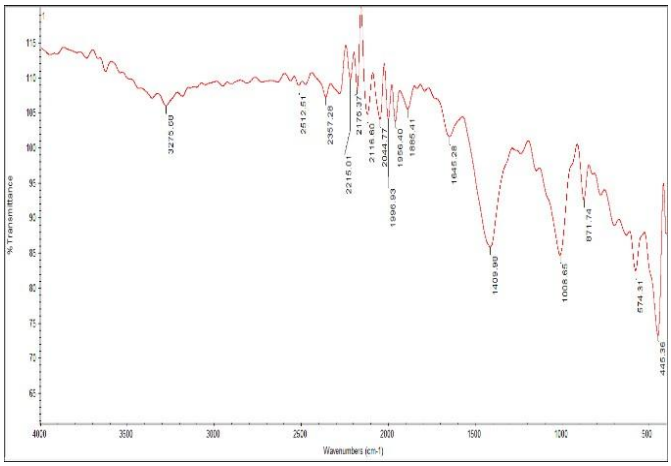


Fig. 3b: FTIR spectrum of U-carrageenan extracted from *C. officinalis*

2. Alginate from *S. denticulatum*:

A comparison of extracted alginate **(Figure 5, a & b)** and commercial alginate (**Figure 4),** as a standard, revealed some slight variations in the height and form of certain absorption bands, but overall the spectral pattern was similar. In **Figure 5 (a & b)**, a broad bands at 3211 cm-¹, 3347 cm-¹, 2826 cm-¹ and 2186 cm-¹ were stretched. Absorbance was also observed around 1500 cm-¹, 1300 cm-¹, 1020 cm-¹, 900 cm-¹ and 872 cm-¹.

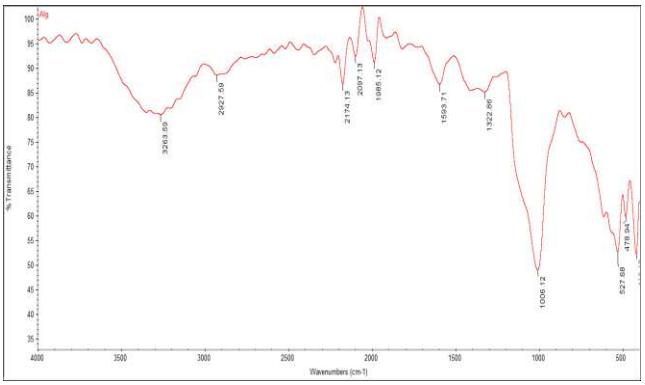


Fig. 4: FTIR spectrum of commercial alginate

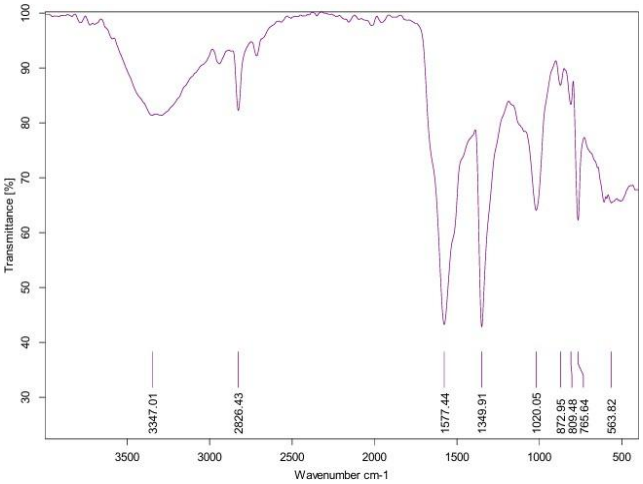


Fig. 5a: FTIR spectrum of C-alginate extracted from *S. denticulatum*

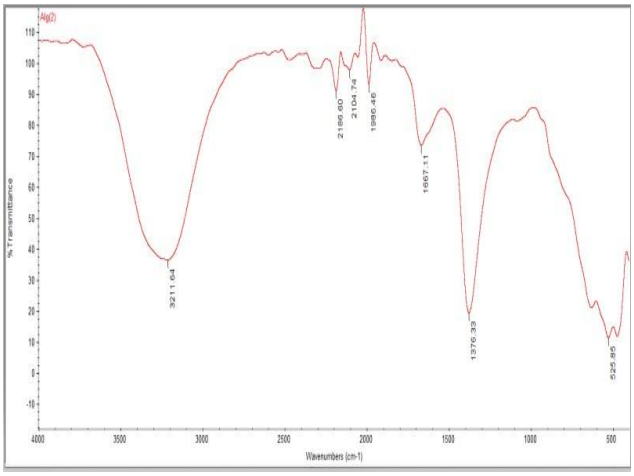


Fig. 5b: FTIR spectrum of U-alginate extracted from *S. denticulatum*

- **Bioplastic formation from seaweeds and polymers:**

  Bioplastic films were prepared directly from seaweeds and from the extracted polymers were produced. Films and its characterization were illustrated in **Table 1**.

- **Bioplastic characterization:**

1. Physical properties

1.1. Color and Texture: The color of the bioplastic films was visually assessed revealing that in *C. officinalis* film was pale yellow and in *S. denticulatum* film was brown. The C- carrageenan and U-carrageenan films were white, while the C-alginate film was brown and the U-alginate film was white. As for the texture, the films were between soft and rough, with some cracks appearing in some of them and smooth of rough surface as shown in **Table 1**.

1.2. Solubility by solvents: The solubility of bioplastic films in different solvents (acetone, ethanol and chloroform) at 30ºC and 60ºC after one hour was showing significant differences ($P<0.001$). The solubility ratios for each solvent were presented in **Figure 6**. The solubility of the films in acetone at 30ºC indicates that the films of *S. denticulatum* and *C. officinalis* were recorded values of 37.5% and 35.7%, respectively. The film of U-carrageenan showed a solubility at a value of 20% compared to C-carrageenan film (12.5%). Similarly, the films of polymers extracted by ultrasound, in both carrageenan and

alginate, showed a higher solubility compared to films of polymers extracted by the conventional method. Comparing at both temperatures, the solubility at 60ºC was higher than at 30ºC for all films. When examining the films' solubility in ethanol and chloroform, it was found to display a trend akin to that of acetone, but the chloroform had less effect than acetone and ethanol in both temperatures. Generally, the films prepared from algae were more soluble than films prepared from polymers.

Table 1: Observation of the visual properties of bioplastic films from different algae

Picture						
Observation	The film is soft, free of cracks , very thin with a rough surface and regular shape	The film is soft, free of cracks , thicker with a smooth surface and regular shape	The film is dry, crack-free and thin with a rough surface and irregular shape	The film exhibits softer, thinner , crack-free with a smooth surface and irregular shape	The film is soft and slightly thick. The surface is rough and irregular shape	The film is dry, crack-free , thin with a smooth surface and irregular shape
Film	*C. officinalis*	*S. denticulatum*	C- carrageenan	U- carrageenan	C-alginate	U-alginate

1.3. Moisture absorption: A significant difference (P<0.001) was observed in moisture absorbance as presented in **Figure 7**. The moisture content of the films was in the following descending order: *S. denticulatum* film (46.2 %), U- carrageenan film (14.3 %), C-alginate film (11.9 %), *C. officinalis* film (10.7%), C-carrageenan film (9.4 %) and the lowest was U-alginate film (2.6 %).

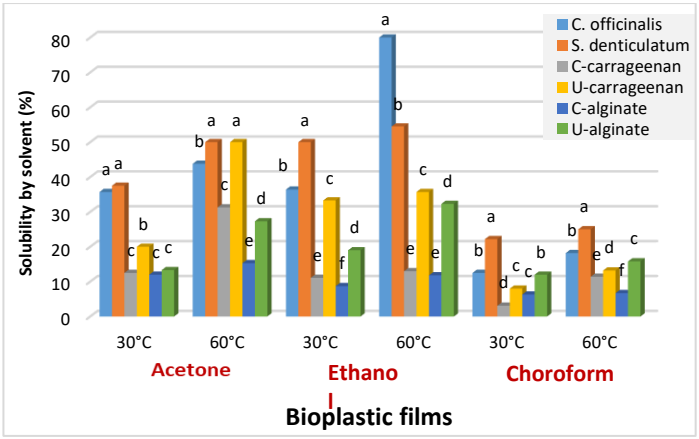


Fig. 6: Solubility of the bioplastic films in solvents

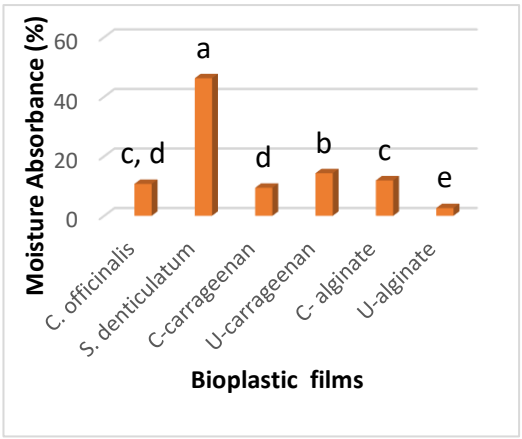


Fig. 7: Moisture absorption of bioplastic films

Values are presented as mean ± standard deviation. Significant at (p < 0.001). The values of identical letters within the same column are not significantly different based on Duncan's Multiple Range Test

2. Mechanical Properties:

Mechanical properties (TS and EAB) were presented in **Figure 8 (a & b)**. Tensile strength ranged from 0.15 to 119.2 MPa, with the highest value observed in *S. denticulatum* film and the lowest in *C. officinalis* film. In contrast, elongation at break ranged from 27.56 to 472 %, with the highest elongation at break was recorded in *C. officinalis* film and the lowest in U- alginate film.

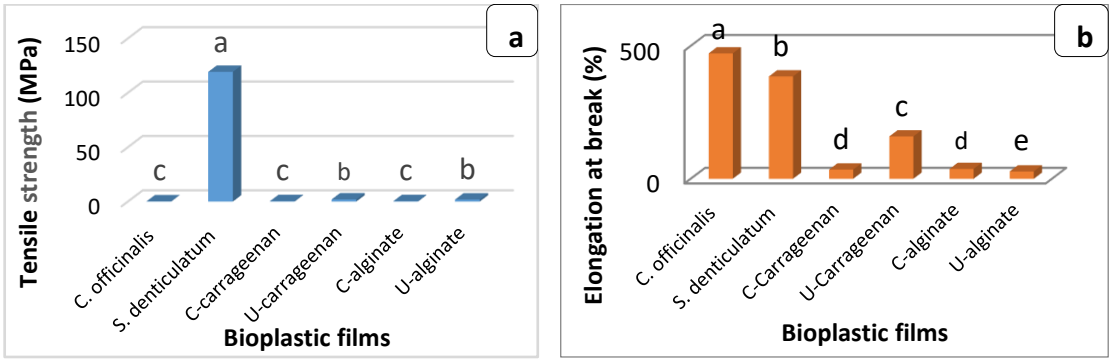
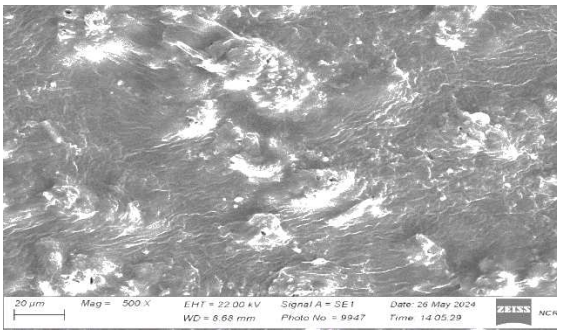


Fig. 8: Mechanical properties of different bioplastic films

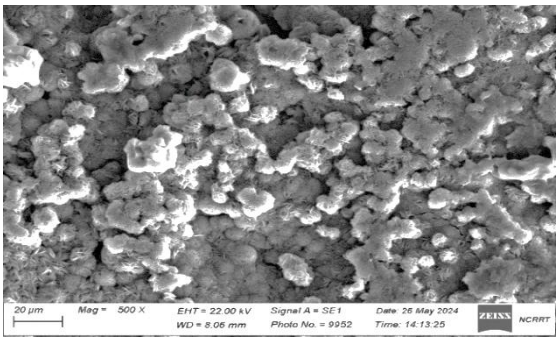
Values are presented as mean ± standard deviation. Significant at ($p < 0.001$). The values of identical letters within the same column are not significantly different based on Duncan's Multiple Range Test.

3. Structural & Morphological Properties

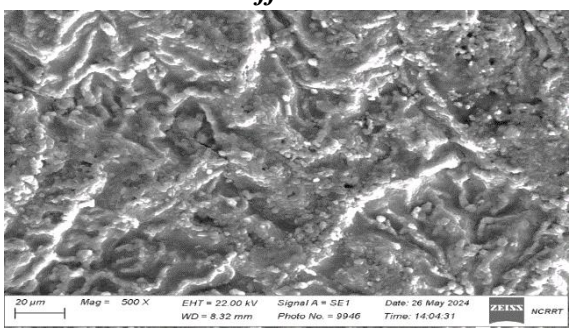
3.1 SEM: SEM analyses (**Figure 9**) of the bioplastic films obtained at the magnification of 500 x revealed a complex layered morphology with distinctive structural features. *S. denticulatum* and *C. officinalis* films exhibited more pores and the micrographs of both carrageenan films exhibited a predominantly laminar structure with multiple parallel layers. The layers display smooth regions interspersed with fine ripple- like patterns surfaces. U-alginate film was smoother and more cohesive than C-alginate film.



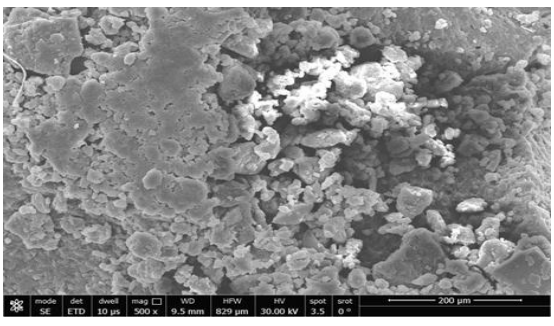
C. officinalis



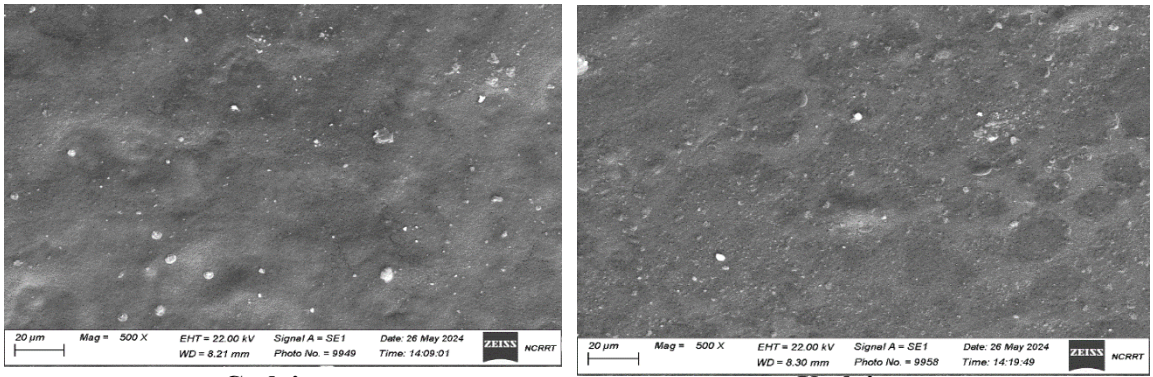
S. denticulatum



C-carrageenan



U-carrageenan



C-alginate **U-alginate**

Fig. 9: SEM images of different bioplastic film

3.2 FTIR

a) FTIR of bioplastic films obtained directly from algae:

When comparing the FTIR spectrum of *C. officinalis* powder (**Figure 10a**) with that of its bioplastic film (Figure 10b), it was observed that some peaks were absent in the *C. officinalis* film (3714 cm^{-1}, 2163 cm^{-1}, and 2018 cm^{-1}). On the other hand, some peaks were present in both the powder and in the film (3293 cm^{-1}, 2931 cm^{-1}, 1402 cm^{-1}, and 1122 cm^{-1}). The FTIR spectrum of the *S. denticulatum* film (**Figure 11b**), when compared to those of *S. denticulatum* powder (**Figure 11a**), revealed new peaks were presented in both the film and the powder at 3207 cm^{-1}, 2929 cm^{-1}, 2845 cm^{-1}, 1424 cm^{-1}, and 1033 cm^{-1}. Additionally, the peaks at 3692 cm^{-1} and 2018 cm^{-1} in the powder shift to 3584 cm^{-1} and 2122 cm^{-1}, respectively, in the film. Moreover, new peaks emerged in the *S. denticulatum* film at 1722 cm^{-1}, 1614 cm^{-1}, 1536 cm^{-1}, 1305 cm^{-1}, and 1253 cm^{-1}. These variations suggest changes in molecular interactions and structural configurations between the powder and film forms.

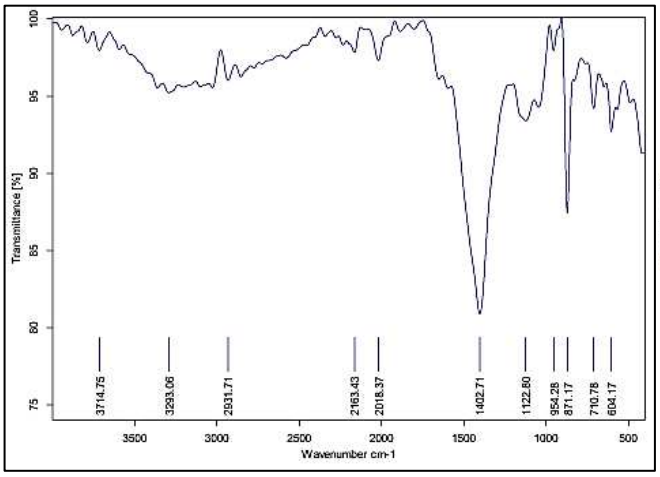


Fig. 11a : FTIR spectrum of *C. officinalis* powder

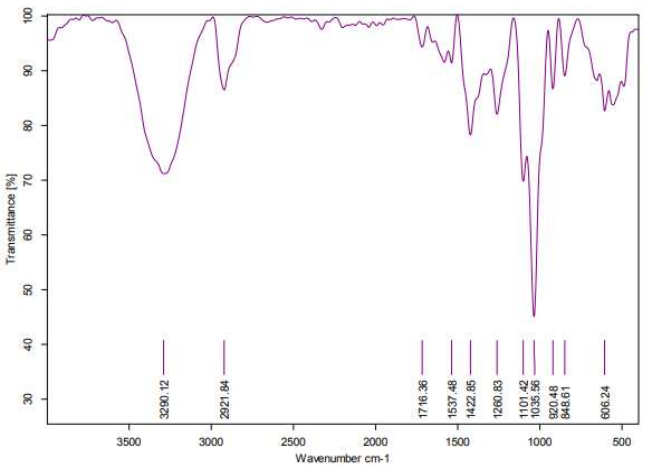


Fig. 11b: FTIR spectrum of *C. officinalis* bioplastic film

b) FTIR of bioplastic films from polymers:

When compared to the FTIR spectrum of the previous C-carrageenan biopolymer (**Figure 3a**) with the FTIR spectrum of C-carrageenan bioplastic film (**Figure 12a**), several peaks were absent in the film, specifically at 3782 cm^{-1}, 3715 cm^{-1}, 3626 cm^{-1}, 3036 cm^{-1}, 2850

cm^{-1}, 2017 cm^{-1}, 1890 cm^{-1}, 1105 cm^{-1}, and 1032 cm^{-1}. Additionally, peaks at 1593 cm^{-1} and 1443 cm^{-1} in the polymer shifted to 1645 cm^{-1} and 1375 cm^{-1}, respectively, in the film. Likewise, when we compared the FTIR spectrum of the previous U-carrageenan biopolymer (**Figure 3b**) with the FTIR spectrum of the U-carrageenan bioplastic film (**Figure 12b**), several peaks were absent in the film, specifically at 2512 cm^{-1}, 2357 cm^{-1}, 2215 cm^{-1}, 2175 cm^{-1}, 2116 cm^{-1}, 2044 cm^{-1}, 1996 cm^{-1}, 1956 cm^{-1}, and 1885 cm^{-1}. Moreover, new peaks emerged at 2925 cm^{-1}, 2890 cm^{-1}, 1336 cm^{-1}, 1241 cm^{-1}, 1151 cm^{-1}, and 1076 cm^{-1} in the FTIR spectrum of the film. FTIR of C-carrageenan and U-carrageenan films revealed distinct differences in absorption peaks. A broad peak at 3358 cm^{-1} in the C-carrageenan film shifted to 3297 cm^{-1} in the U-carrageenan film. A peak at 2969 cm^{-1} was observed in both films. In contrast, several new peaks emerged in the U-carrageenan film at 2890 cm^{-1}, 1417 cm^{-1}, 1241 cm^{-1}, 1151 cm^{-1}, 1105 cm^{-1}, 1076 cm^{-1}, and 1002 cm^{-1}.

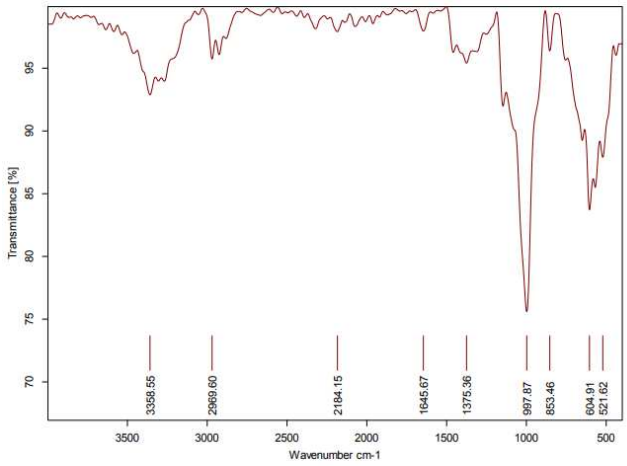


Fig. 12a: FTIR spectrum of C-carrageenan bioplastic film

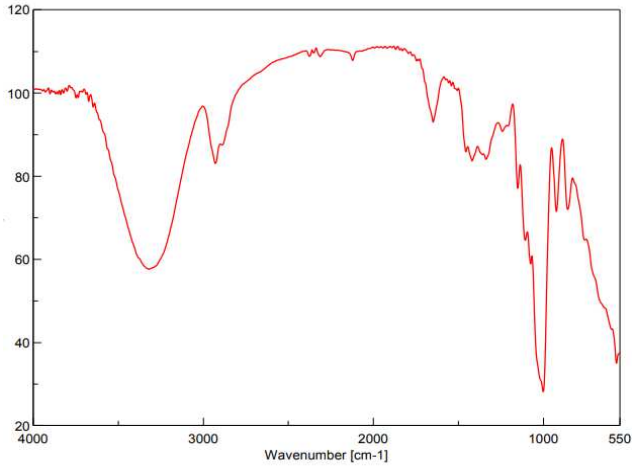


Fig. 12b: FTIR spectrum of U-carrageenan bioplastic film

In comparison to the FTIR spectrum of the previous C-alginate biopolymer (**Figure 5a**) with the FTIR spectra of the C-alginate bioplastic film (**Figure 13a**), a peak at 1032 cm^{-1} was absent in the film. Peaks at 3347 cm^{-1}, 2826 cm^{-1}, 1577 cm^{-1}, and 1020 cm^{-1} still exist in the film. Peak initially observed at 1349 cm^{-1} in the polymer shifted to 1260 cm^{-1} in the film, and new peaks emerged at 2916 cm^{-1}, 1466 cm^{-1}, and 1418 cm^{-1}. Compared to the FTIR spectrum of the previous U-alginate biopolymer (**Figure 5b**) with the FTIR spectra of the alginate bioplastic film (**Figure 13b**), the peaks at 2186 cm^{-1}, 2104 cm^{-1}, and 1986 cm^{-1} were absent in the film, but peaks at 3211 cm^{-1} and 1376 cm^{-1} still exist. Additionally, a peak initially detected at 1667 cm^{-1} in the polymer shifted to 1732 cm^{-1} in the film. Comparing the FTIR spectrum of the C-alginate and U-alginate films, a peak at 3309 cm^{-1} and 1260 cm^{-1} in the C-alginate film was shifted to 3298 cm^{-1} and 1149 cm^{-1}, respectively, in the U-alginate film. Peaks at 2916 and 1576 cm^{-1} were observed in both films and new peaks appeared in the U-alginate film at 1732 and 1399 cm^{-1}.

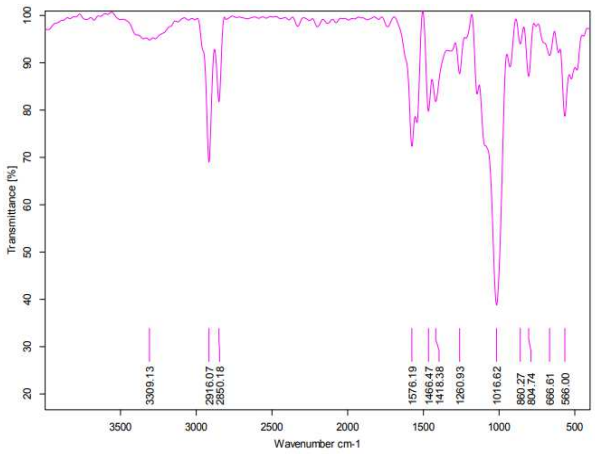


Fig. 13a: FTIR spectrum of C-alginate bioplastic film.

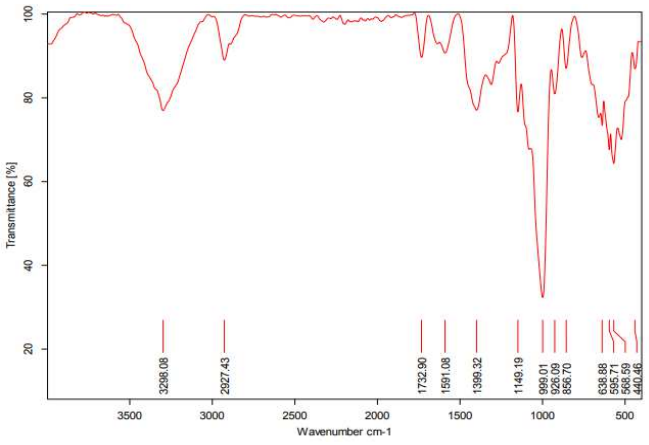


Fig. 13b: FTIR spectrum of U- alginate bioplastic film

3.3 X-Ray diffraction analysis (XRD): The diffract gram of all films shows multiple distinct diffraction peaks in the 2θ range of 10° to 80° (**Figure 14**). The XRD patterns of bioplastic film derived from *C. officinalis* indicate that the most intense peak appears at 2θ = 19.430° with a relative intensity of 100%. Several peaks were observed in the *S. denticulatum* film from 10° to 50° and the highest at 30° approximately. The X-ray diffraction pattern of the C-carrageenan film exhibits distinct peaks at 2θ = 32.000° with respective relative intensity of 100%. On the other hand, U-carrageenan film exhibits several characteristic peaks in the 2θ range of 10° to 80°, with the most prominent peaks observed between 15° and 35°. It was found that some similarity in XRD patterns in both alginate film. C-alginate film showed distinct peaks at 2θ = 22.209° and 20.023° with relative intensities of 100% and 96.1%, respectively. In contrast, the XRD analysis of U-alginate film revealed its most intense peak at 2θ = 17.287° with a relative intensity of 100%. This is followed by prominent reflections at 20.008° with a relative intensity of 95.1% and 22.150° with an intensity of 89.4%

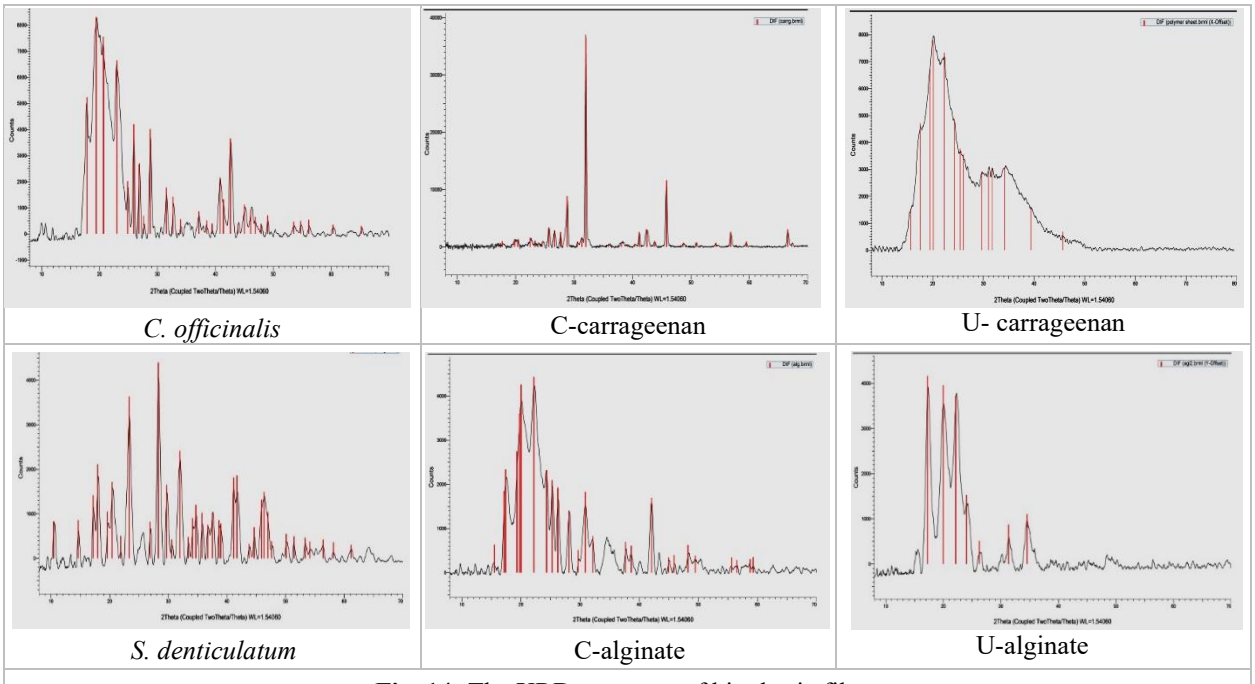


Fig. 14: The XRD spectrum of bioplastic films

4. Biodegradability:

4.1 Biodegradation by soil:

The degradation of bioplastic films in soil after 30 days was shown in **Figure 15**. In clay soil, the highest degradation rate was observed in *C. officinalis* film, reaching 27%, followed by U-carrageenan film (25.6%), *S. denticulatum* film (17.6%), C-alginate film (14.8%), U-alginate film (7.5%), and C-carrageenan film (4.6%). Degradation rates varied from 4.6% to 27%. In sandy soil, degradation rates ranged from 3.9% to 13.4%. The highest rate was observed in the U-carrageenan film, while the lowest was in the C-carrageenan film. The decomposition rate order was U-carrageenan film (13.4%), *C. officinalis* film (12.5%), C-alginate film (12.5%), *S. denticulatum* film (6.5%), U-alginate film (4%), and C-carrageenan film (3.9%).

4.2 Biodegradation by water:

All bioplastic films underwent assessment in acidic, distilled, and alkaline water for a duration of 7 days, revealing notable variations ($P<0.001$) as depicted in **Figure 16**. In acidic water, the *C. officinalis* film exhibited the highest degradation, achieving complete degradation (100%). The degradation values of the remaining films were as follows: C-alginate (95.3 %), U-alginate (76.8 %), U-carrageenan (64.5 %), *S. denticulatum* (58.3 %) and C- carrageenan (50 %). In distilled water, the *C. officinalis* film once again exhibited the highest degradation, achieving complete degradation (100%). The degradation rates for the other films were as follows: C-alginate (62.9 %), U-carrageenan (48.5 %), C-carrageenan (38.4 %), *S. denticulatum* (35.9%) and U-alginate (23 %). In alkaline water, the *C. officinalis* film demonstrated the highest degradation, achieving complete degradation (100%) while the C-carrageenan film showed the lowest degradation value of 4 %. The other degradation rates films were *S. denticulatum* (58.3 %), U- carrageenan (34.3 %) and U-alginate (22.7 %).

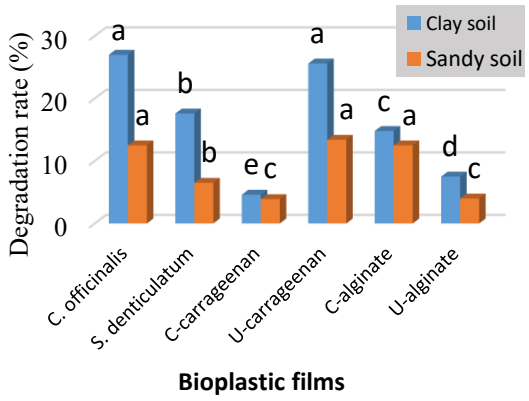


Fig. 15: Degradation of bioplastic films in soil after 30 days

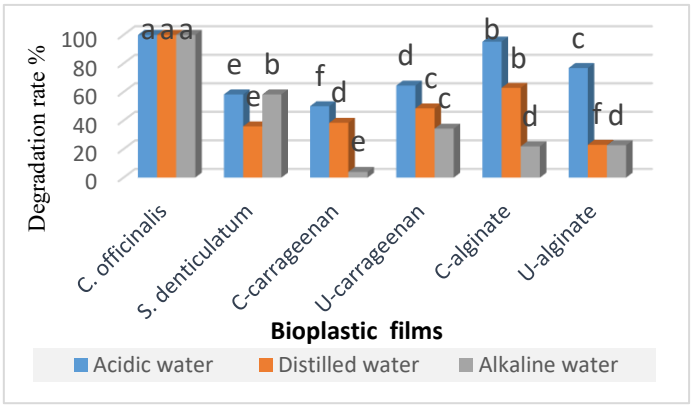


Fig. 16: Degradation of bioplastic films in water after 7 days

Values are presented as mean ± standard deviation. Significant at ($p < 0.001$). The values of identical letters within the same column are not significantly different based on Duncan's Multiple Range Test.

Discussion

A significant step toward more environmentally friendly plastic alternatives is the use of marine algae and other biomass resources to create bioplastics. In order to avoid reabsorbing moisture and contaminants, which could lower the feedstock's quality and have an impact on the process of producing bioplastics, seaweeds were dried and ground into a fine powder before use (**Kachaanun et al. 2022; Kanagesan et al. 2022; Hanry and Surugau 2024).** Polysaccharides (such as alginate and carrageenan) found in abundance in seaweeds can be used to manufacture bioplastics (**Venugopal 2016).** The manufacturing of seaweed-based bioplastics can directly use the entire biomass without separating its constituent parts or the extraction of particular biopolymers, which are then converted into bioplastics (**Torrejon et al. 2025**). The yields of biopolymers differ significantly based on the species of alga and the extraction technique utilized **(Abd El-Latif et al. 2025)**. In this study, alginate was extracted from *S. denticulatum* and carrageenan from *C. officinalis* using two methods: the conventional method and the ultrasound-assisted method. **Latifi et al. (2015)** demonstrated that extract alginate from *Sargassum* sp. yielded between 12% and 30% based on the extraction method. In the present study, alginate extracted from *S. denticulatum* using the conventional method was 16.8%, which falls within the range reported by **Belattmania et al. (2020),** who found that the yield of alginates in *S. muticum* reached up to 18%, while the yield from *S. filipendula* was 17%. Furthermore, according to **Dalal et al. (2021),** the alginate yield was 17.5% in *S. latifolium* based on dry biomass. **Hanry et al. (2022)** recorded that the average alginate yield from *Padina* sp. was 21.5 %. In case of carrageenan, according to **Ha et al. (2022),** the red algae *Eucheuma gelatinae* yielded 42.68% carrageenan. In comparison, the yields of carrageenan from *C. officinalis* in this study ranged from 3.7% to 6.6%. This is lower than the results of **Sebaaly et al. (2014),** who found an overall carrageenan yield of 10% isolated from the red alga *Corallina*. The difference in yields of hydrocolloids between different studies is due to the variability of seaweed properties, the season, harvest time, location and other variables (**Beata Łabowska et al. 2019**).

Recent methods, like ultrasound-assisted extraction, have greatly increased extraction yields and claims **(Saji et al. 2022).** According to **Cheng et al. (2022)** higher extraction yields, lower energy usage and shorter extraction times are key advantages of the ultrasonic extraction method. This is consistent with the present data, which showed that employing ultrasound-assisted extraction boosted the yield of carrageenan from *C. officinalis* from 3.7% to 6.6% and the yield of alginate from *S. denticulatum* from 16.8% to 51.8%. Ultrasound waves' cavitation effects, which break down cell walls and boost solvent penetration through cellular diffusion, are responsible for this improvement **(Flórez-Fernández et al. 2019; Martín-del-Campo et al. 2021; Gómez Barrio et al. 2022)**. Furthermore, the shock waves generated by ultrasonic waves cause cells to rupture and pores to widen, which facilitates material release and increasing extraction yields even further **(Wang et al. 2018).** The ultrasound technique minimizes negative effects on the environment while increasing the effectiveness of biopolymer recovery by using less energy, solvent, and processing time. These developments are essential to the more sustainable and profitable production of seaweed-based bioplastics **(Matos et al. 2021; Junior and Turan 2022).**

FTIR analysis of carrageenan in this study (**Figures 2 &3**) is consistent with the findings of **Martín-del-Campo et al.** (**2021**), where absorption bands in the region of 3200–3400 cm^{-1}, corresponding to –OH groups, were observed in both the C-carrageenan and U-carrageenan samples compared with commercial carrageenan. At around 1250 cm^{-1}, low-intensity peaks were observed in both the commercial and C-carrageenan samples, indicating the presence of sulfate ester ($O–SO_3$) groups. Strong characteristic C–O stretching bands of 3, 6-anhydrogalactose were observed at 1036, 1032, and 1008 cm^{-1}. Additionally, bands were identified in the 920–925 cm^{-1} region in commercial and C-carrageenan samples, corresponding to 3, 6-anhydro-D-galactose and D-galactose-4-sulfate (G4S), which are characteristic functional groups of carrageenan. These peaks are characteristic of carrageenan as reported by **Pereira et al.** (**2009** and **2013**).

In the present FTIR spectra of C- and U-alginate (**Figures 2 &3**) extracted from *S. denticulatum*, a strong broad peak around 3347 cm^{-1} was observed, corresponding to the O–H stretching vibrations. Additionally, weak peaks around 2826 cm^{-1} were attributed to C– H stretching vibrations. These results are consistent with the findings reported by **Sari-Chmayssem et al. (2016).** Peaks observed around 1667 cm^{-1} and 1376 cm^{-1} in the two extracted alginate correspond to the stretching vibrations of the carboxylate (COOH) groups, which are associated with the mannuronate and guluronate units in sodium alginate this was also found by **Rhein- Knudsen et al. (2017).** According to **Sinha et al. (2010)**, two distinct peaks at 1006 cm^{-1} and 1020 cm^{-1} detected in the commercial and C-alginate samples indicate the presence of β- mannuronic acid and α-L-guluronic acid residues, respectively. According to forgoing discussion the above, the carrageenan and alginate were verified in comparison with the commercial polymer.

All prepared films were softness and no cracks (**Table 1**) may be mainly due to sufficient cross-linking with additives (**Bouzenad et al. 2024**). Glycerol, PVA and starch were added to films made from the polymers or algae to improve the bioplastic structure which increases flexibility. Glycerol is an effective plasticizer because of its ability to soften the film structure increase the mobility of the biopolymer chain and improves mechanical properties in bioplastics **(Putri et al. 2023**). PVA able to form physically cross-linked hydrogels and exhibit a variety of qualities, including stability at room temperature, rubber-like flexibility, and great mechanical strength (**Ricciardi et al. 2004).** Also, adding starch improves the mechanical properties of films due to combine with molecules through hydrogen bonding and presence of hydroxyl groups in their molecular chains (**Cheng et al. 2022**).

The susceptibility of bioplastic films to damage is determined by testing their solubility in organic solvents. The results represented in Figure 6 indicate that bioplastics derived directly from algae exhibit the highest solubility in all solvents compared to films formed from polymers. This may be due to incorporation of PVA in film which enhances the properties of bioplastics, as supported by the findings of **El-Sheekh et al. (2024).** In case of carrageenan and alginate films, there are slightly soluble in organic solvents but soluble in water, as illustrated by **Cheng et al.** (**2022**) and **Rosiak et al.** (**2021**). However, the rate of solubility varied depending on the type of bioplastic, solvent and temperature.

The results of the moisture absorption test on the prepared bioplastic film (**Figure 7**) revealed that all biofilms demonstrated significantly high moisture resistance except *S.*

denticulatum film. The moisture absorption resistance of these films may be attributed to the strong adhesion between the algae and PVA, especially in case of *C. officinalis* film, which reduces the presence of O-H groups and consequently limits the diffusion of water molecules into the films (**Rahmadiawan et al. 2022**). The moisture absorption resistance was high when compared with the results obtained by **Siah et al. (2015)** and **El-Sheekh et al. (2024)**. The polymers films were lower moisture absorption according to alginate and other polysaccharides have a high affinity for water (**Bouzenad et al. 2024**). According to **Sudheesh et al. (2020)**, bioplastic films with higher relative crystallinity had reduced water penetration. **Santana et al. (2022)** observed that the higher the biomass content, the lower the water uptake capacity, this comment may explain the low moisture absorbance of *C. officinalis* film.

Mechanical properties (TS and EAB) are one of the most important indicators of the core performance of algae-based bioplastics **(Krishnan et al. 2024).** Adding glycerol is necessary to improve mechanical properties. This can be explained by the fact that glycerol is hydrophilic, has a large number of hydroxyl groups, and has a short molecular size, which results in homogenous penetration into the polymer matrix (**Guidara et al. 2020).** The mechanical property analysis of the prepared bioplastic films showed that the *S. denticulatum* film had the highest tensile strength (119.2 MPa). In this study, TS is low when compared with the previously obtained results (**Echeverria et al. 2017; El-Sheekh et al. 2024)** but revealed some agree with **Abdo and Ali (2019**) who synthesized bioplastics from *Microcystis* sp. (TS= 1.35 Mpa and EAB = 530 %). Whereas **Hanry et al. (2022)** found that TS of the alginate film extracted from *Padina* sp. ranged from 0.52 – 9.43 MPa and EAB ranged from 4.97 – 8.61 mm. The higher in TS in *S. denticulatum* could be explained by the improved interfacial adhesion with PVA, which creates a more uniform mixture. Furthermore, a stronger bond between *S. denticulatum* and the PVA matrix improved their compatibility and produced a higher TS, which is in line with **Dianursanti et al. (2019).**

Elongation values in algal films were range of elongation values of conventional LDPE (low-density polyethylene) based plastic (400 %) and less than the plastics international standard (ASTM 5336) in UK (above 500%) **(Belgacem and Gandini 2011**).The higher EAB observed in *S. denticulatum* and *C. officinalis* films can be ascribed to several cross-linking that impact the biofilm's pliability and stretch capacity. This cross−linking affects network formation and the biofilms' mechanical properties (**Paixão et al. 2019; Lee et al. 2023; Bouzenad et al. 2024**). **Paula et al. (2015)** found that the TS and EAB of the pure -carrageenan film were 2.5 MPa and 1.04%, respectively**,** while **Langit et al. (2019)** demonstrated TS was 0.84 MPa in alginate film. **Tarique et al. (2021)** observed TS ranging from 1 to 17 MPa, whereas **Sudhakar et al. (2021**) reported TS was 13.78 MPa and EAB over 6% of the carrageenan film. **Sudhakar et al. (2022)** employed nanocomposites containing carrageenan and reported TS ranging from 4 to 10 MPa and EAB of 1 to 6 %.

Scanning electron microscopy was used to examine the bioplastic films' cross-sectional area. The image analysis of the bioplastic films at 500x magnification is displayed in Figure 9. It was found that the alginate bioplastic film was the most homogeneous of all the bioplastic films. **El-Sheekh et al. (2024)** observed that the surfaces of biomass algal film may be rough which low component and plasticizer adherence may have caused that. According to **Abdillah and Charles (2021),** adding more

carrageenan to starch blended films makes the surface of the film rougher.

The formation of bioplastic film cause changes in functional groups of film due to the polymerization. Because of the plasticizing reaction between biomass and glycerol, the presence of each functional group of glycerol is bound to change or shift some peaks. (**Hanry et al. 2022)**. The interaction of the chemicals in the bioplastic film may be the cause of several peaks found at the lower wavenumber (**Kadell and Callychurn 2023)**. Since the number of peaks indicates the complexity of the sample, more peaks indicate more compounds, and numerous peaks frequently indicate the existence of different functional groups or chemical bonds (**Nandiyanto et al. 2019**). This is found greatly in U-alginate and U-carrageenan films in which new peaks were observed. This also found in FTIR of *S. denticulatum* film compared to FTIR of *C. officinalis* film for the first had more number sharp of peaks. The bioplastic sheets' FTIR examination revealed strong peaks between 3200 and 3650 cm^{-1} were seen in all films, suggesting the existence of O-H functional groups. Between 3000 and 2700 cm^{-1}, C-H and O-H stretching caused by COOH functional carboxylic acid groups was noted. Additionally, the FTIR spectra showed strong peaks for the sulfate ester group's S=O stretching in the 1336–1418 cm^{-1} range **(Khan et al. 2024).** N-H bond (Amines and some proteins) appear around 3300 cm^{-1} .In addition, the functional groups of ester group (1250-1720 cm^{-1} range) and amine group (1000-1250 cm^{-1} range) were found in all bioplastic films. These functional groups may have an impact on the biodegradability, water solubility, mechanical, optical, and physical characteristics of bioplastic films (**Filiciotto and Rothenberg 2021).** Carboxylic acid is essential for improving the chemical and mechanical characteristics of bioplastics, which makes them a competitive substitute for polymers derived from petroleum. Amines help bioplastics become more biodegradable by enhancing their strength, flexibility, and resistance to heat breakdown (**Nagaraj *et al.* 2023).**

XRD describes the physical characteristics and chemical makeup of various materials and can also be used to determine the degree of crystallization **(Zhang et al. 2019).** It is one of the most crucial properties to determine the mechanical properties of the plastic material; generally speaking, a higher degree of crystallization makes the material stiffer, stronger, but more brittle. The polymer's tactility, hardness, modulus, density and transparency may all be affected by the degree of crystallinity (**McAdam et al. 2020**). The XRD pattern of the *C. officinalis* and *S. denticulatum* films exhibit sharp and intense peaks around $2\theta \approx 20°$, indicating a high degree of crystallinity**. Ricciardi et al. (2004)** reported that strong intermolecular interactions between PVA chains through hydrogen bonding result in multiple diffraction peaks, indicating the presence of crystalline atactic PVA. XRD patterns of the U-carrageenan and U-alginate films exhibited sharp peaks, indicating a higher degree of crystallinity compared to the films produced using the conventional extraction method. This increased crystallinity may be attributed to the ultrasonic process, which can enhance molecular alignment and promote more ordered polymer structures during film formation (**Kumar et al. 2023)**.

In soil environments, the degradation process typically begins with the colonization of the bioplastic surface by diverse microbial populations. These microorganisms form biofilms on the polymer surface, initiating enzymatic degradation **(Tosin et al. 2019)**. Microbial communities secrete specific

extracellular enzymes capable of cleaving polymer chains into smaller monomers, which are further metabolized into CO_2, H_2O and other end products **(Adhikari et al. 2016, Chamas et al. 2020)**. Both bacterial and fungal communities play a pivotal role in the degradation of biodegradable plastics. Fungi may be interact with plastic surfaces, creating grooves, pits, and cavities **(Muroi et al. 2016).** In this study, within 30 days, the bioplastic films in both clay and sandy soil rapidly lost their original structural integrity and shape. Significantly variable weight loss ratios were found in the results, suggesting that varying polymer and type of plasticizer had a major impact. Additionally, because glycerol loves water, it may have promoted microbial development (**Marium et al. 2021).** *C. officinalis* film degraded at the fastest rate in clay soil (27%), whereas U-carrageenan film degraded at the fastest rate in sandy soil (13.4%). The biofilms in clay soil go through a process called chain scission, in which chemical hydrolysis breaks up polymer chains. Furthermore, the biofilms are further broken down by microbes into biomass, CO_2 and water (**Kyrikou and Briassoulis 2007).** All of the bioplastic films exhibited absorption peaks for oxygen-containing groups (ester, carboxylic, amine, etc.) in the FTIR study. It is well recognized that these functional groups improve biodegradability **(Khan et al. 2024).** In alginate, the primary reasons of weight loss are the breakdown of the chains and removal of the carboxylate group (**Bouzenad et al. 2024**). Average of degradation alginate films ranged from 7.5 to 14.8 %, this was slower than **Solak and Dyankova (2014**) which degraded 40-60% alginate films in 12 days and study of **Putri et al. (2023**) which degraded 37-64% in 10 days. **Langit et al. (2019)** also found that the higher the alginate concentration, the lower the biodegradation time due to the inhibition of water absorption and the viscosity of the solution. This is seen in U-alginate, which deteriorated more quickly than C- alginate. Degradation of carrageenan ranged from 4.6 to 27 %, according to **Djonaedi et al. (2023**), biodegradability is influenced by the composition and properties of the bioplastic films and polymers. The more hydrophilic a polymer will accelerate the degradation process (**Adhikari et al. 2016**). **MarathakaRani and Navinraj (2026)** stated that the decomposition time of bioplastics varies depending on their composition and the type of soil that factors such as microbial activity, moisture, and organic content had influence decomposition rates.

One important property of bioplastics that indicates their capacity to decompose is their solubility rate (**Arham et al. 2016**). The solubility of the film depends on a number of variables, such as free hydroxyl groups, relative crystallinity and film shape. The interactions between water molecules and free hydroxyl groups change a film's hydrophilicity **(Li et al. 2018**). During the analysis of all films, under alkaline conditions, degradation efficiency decreases according to changing the functional groups' ionization state and decreasing hydrophilicity, approved with **MarathakaRani and Navinraj (2026).** It was found that the water solubility of the film made from *C. officinalis* was completely degraded in acidic, distilled, and alkaline water within 7 days. This result slightly agreement with **Naesa et al. (2019**), bioplastic films have outstanding degradability in all water. Researchers found that films made from algae dissolve in different types of water to varying degrees (**Siah et al. 2015; El-Sheekh et al. 2024; Khan et al. 2024**). As shown in Figure 16, the solubility of all films increases in acidic water, conversely, distilled water, which has low ion concentrations, has high solubility because it interferes less with the molecular

interactions of the substance. It is important to note that solubility may also be impacted by the type of plasticizer used (**Müller et al. 2008**). The solubility of all forms of carrageenan in water depends on a number of variables, including the molecule's sulfate group and the amount of 3, 6-anhydrogalactose present (**Nguyen et al. 2014**). The low molecular weight, lower charge of the carrageenan **(Suryanto et al. 2019**) and hydroxyl groups (**Cheng et al. 2022**) enhance the solubility .Because alginates are highly hydrophilic so increasing the number of hydrophilic molecules leads to a significant increase in the percentage of film solubility (**Alba and Kontogiorgos 2018**). This is evident in that U-alginate film is soluble more than C-alginate film. The results of this study are in line with mounting evidence that seaweeds and their polysaccharide-based polymers offer a workable basis for creating bioplastics that are environmentally friendly. Previous studies have highlighted the need of utilizing renewable resources to address resource circularity and plastic pollution. The present research offers a solid basis for creating single-use, low-impact bioplastic products that support Egypt's 2030 objective. To maximize scalability and durability for industrial applications, more work is required.

Conclusion

In this study, polymers were extracted from seaweed using two methods (with ultrasound being the most efficient) to prepare bioplastic films from the seaweed itself and the extracted polymers while studying the physical, mechanical, morphological, and degradability properties of all films. The films were successfully prepared without any cracks with varying properties, making each suitable for a different application. Due to its high degradation rate in soil and water, the *C. officinalis* film is suitable for short-term applications such as packaging applications and facial masks. Meanwhile, the *S. denticulatum* film, with moderate degradation in soil and slow degradation in water but with high TS and EAB, is well-suited for packaging applications like shopping bags. Alginates had higher productivity than carrageenan, and polymer films offer excellent physical and chemical properties, including moisture resistance and mechanical strength, along with biodegradability in soil. This contributes to reducing the harmful environmental impact of conventional plastic and reducing fossil fuel consumption.

Author contribution: Hager Abd El-Latif, Shymaa Abd-el-Kader and Afaf H. Ali, carried out the experiments, constructed the figures and tables. Wafaa Abou El-kheir identified seaweeds, wrote the first draft of the manuscript and review the final. Afaf Ali shared the research idea, wrote, edited the manuscript, and revised the final manuscript.

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Data availability The current article contains all of the data created or examined during this investigation.

Declarations

Competing interests The authors declare no competing interests.

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