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ECO-FRIENDLY BIOPLASTIC MATERIAL DEVELOPMENT VIA SUSTAINABLE SEAWEED BIOCOMPOSITE

Abstract: The study focused on the development of an environmentally friendly bioplastic material using sustainable seaweed-based biocomposites. Algal biomass (*Gracilaria edulis*) was processed and combined with starch, glycerol, glacial acetic acid, and chitosan to create flexible, homogenous biopolymer films. These films exhibited comparable physical properties to commercial plastics and retained their inherent colour post-processing. Spectroscopic analysis revealed intense UV-Vis peak points aligned with seaweed composition. Mechanical testing demonstrated adequate strength and flexibility, similar to starch-based bioplastics, with a tensile strength of 3.383 MPa and lower elongation strength of about 31.90 %. Material migration tests indicated a preference for water, suggesting suitability for low-moisture foods. The bioplastic film displayed notable biodegradability and compostability, showcasing its potential as a sustainable alternative for food packaging. This innovative contribution advances eco-friendly bioplastic material, addressing plastic pollution and promoting biocomposite use.

Keywords: *Gracilaria edulis*, chitosan, bioplastic film, food packaging, microscopy, spectroscopy, biodegradability, compostability

Introduction

There is a growing problem of contamination in the world, which needs to be addressed by the circular economy approach and better management [1, 2], as well as remediation of existing polluted areas [3]. In the face of the pressing challenge posed by excessive plastic waste, researchers have been diligently seeking effective strategies to mitigate its impact. India, a significant contributor to a staggering annual volume of approximately 3 tonnes (tonne = 10⁶ g = Mg) of plastic waste, confronts a pressing issue with the proliferation of persistent microplastic pollutants [4, 5]. These insidious pollutants

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not only disrupt aquatic and terrestrial ecosystems but also exert far-reaching consequences on human well-being [6]. In response, concerted efforts are being directed toward exploring advanced interventions for waste management and remediation [7], which include innovative biological degradation techniques [8, 9].

In the realm of eco-conscious alternatives to conventional single-use plastics, bioplastics derived from diverse sources such as potatoes, corn, vegetable oils, and seaweeds have exhibited considerable promise [8, 9]. Seaweeds, characterised by their abundant availability and capability to thrive in seawater environments, boast unique advantages, notably their potential for reducing greenhouse gas emissions and their edibility [10-12]. Scientists have discerned the latent potential of seaweeds in the domain of food packaging, paving the way for the creation of edible cups, pouches, and similar innovations [10, 11]. However, the utilisation of seaweed in bioplastics necessitates careful consideration of extraction methodologies, with supplementation proving crucial for bolstering mechanical strength and water resistance within seaweed films [10-12].

Within the scope of producing seaweed-based bioplastics, various methods have been classified into chemical, mechanical, and biological extraction approaches, each bearing its own advantages and drawbacks [10]. The production process is influenced by a myriad of factors, including the type of seaweed species employed, solvent selection, pre-treatment techniques, cost considerations, temporal aspects, environmental implications, and the intricacies of scaling up the production [10-12]. In isolation, seaweed films exhibit limitations such as compromised mechanical properties and a lack of robust water resistance [10]. Consequently, there arises a compelling necessity to diversify the applications of seaweed materials by integrating supplementary components that can fortify the resulting bioplastic films.

To this end, the present study embarked on the creation of seaweed-based composite films, harnessing the potential of supplementary materials like starch and chitosan. Starch, renowned for its capacity to yield translucent films and introduce flexibility [13], synergises with chitosan, celebrated for its multifaceted properties encompassing antimicrobial action, bioadhesion, biocompatibility, and biodegradability [14]. Notably, these individual components bear certain limitations when employed in isolation or combination; starch contributes to excessive water vapour permeability and chitosan, when utilised in its bare form, introduces challenges associated with structural integrity and settling issues [13, 15]. However, the amalgamation of these materials with the extract of *G. edulis* seaweed serves as an astute strategy to ameliorate these inherent drawbacks. This innovative fusion effectively addresses the individual weaknesses of these components, offering a promising avenue for the sustainable development of bioplastic materials with enhanced properties.

Materials and methods

Pre-treatment and processing of algal biomass

Algal biomass (*G. edulis*) in dried form was purchased from AK Seaweeds, Ramanathapuram, Tamil Nadu, India. The received biomass is sorted and thoroughly washed with distilled water to remove dust and debris. Then, washed biomass (1000 g) was soaked for 24 h in distilled water (5000 mL) for swelling. A lemon slice was added to the container to remove the grassy odour. Swelled biomass stored at 4 °C and used whenever it is necessary.

Preparation of algal biomass-starch based bioplastic films

Swollen biomass (100 g) in 500 mL distilled water was subjected to sonication using a bath sonicator (SKE-5S, Labnet scientific services, India), operated at a temperature of 70 °C for 45 min and allowed for heat treatment using an autoclave (121 °C for 15 min). Now, autoclaved biomass was homogenised using a blender and the suspension was filtered using a muslin cloth (~100 µm) to remove the coarse particles that were not homogenised. To this mixture, starch (50 g) was added and the suspension was boiled at 95 °C with shaking until the mixture becomes homogenous dispersion. Later glycerol (15 g), glacial acetic acid (6 g) and chitosan (4 g) were added and mixed. Heating was stopped once the dispersion became thick and was finally cast onto a Teflon plate. This mixture was kept in a ventilated room at ambient conditions (30 °C), and bioplastic films were recovered after 48 h of drying. All the stated chemicals in this section were purchased from SRL, India.

Microscopy, physical and mechanical properties; spectroscopic analysis; moisture content analysis and material migration test; biodegradability and compostability assessments were done to the obtained biofilm and were detailed in the Supplementary Materials (ESI).

Results and discussion

Bioplastic films

Consistent free-standing films were produced upon casting the water-based solutions into Teflon moulds and drying them at ambient conditions. Figure 1a shows the photograph of the prepared solution and obtained biopolymer film, which is homogenous and flexible. The colour of the solution was preserved on drying, indicating that mild processing conditions have no adverse effect on the inherent colour properties. The microstructure of the obtained bioplastic film was investigated by an inverted metallurgical microscope. Figure 1b shows the surface morphology of the film at 100x magnification which states that the bioplastic material produced was continuous and exhibited an amorphous-like matrix structure.

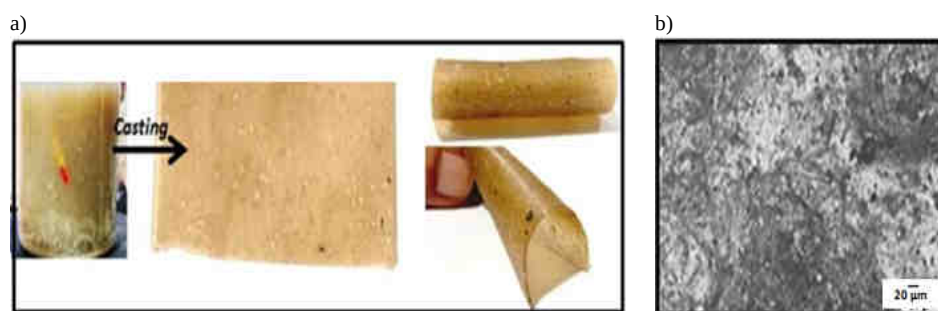


Fig. 1. a) *G. edulis* extract with starch and chitosan supplements, casted bioplastic film with folded and bent pictures, and b) microscopic image of the dried film at 100x magnification

The key physical parameter of the prepared bioplastic film is given in Table 1. Key roughness parameters, arithmetical mean deviation R_a and root mean square R_q values are calculated and compared with the commercially available plastic sheet. In brief, R_a is the

average roughness deviation of all points from a plane to the test portion surface and R_q is the average of the measured height variations taken within the evaluation of area, measured from the mean linear surface. These values from the prepared bioplastic film are very near to the commercial plastic film (Fig. S1, Supplementary Materials).

Table 1

Physical properties of the produced bioplastic film

Properties	Observed values
Colour	Copper brown
Porosity [%]	10.2 ± 1.7
Thickness [mm]	0.83 ± 0.04
Roughness parameters	
R_a	177.29 ± 0.32
R_q	196.01 ± 0.14
Moisture [%]	10.2 ± 0.8

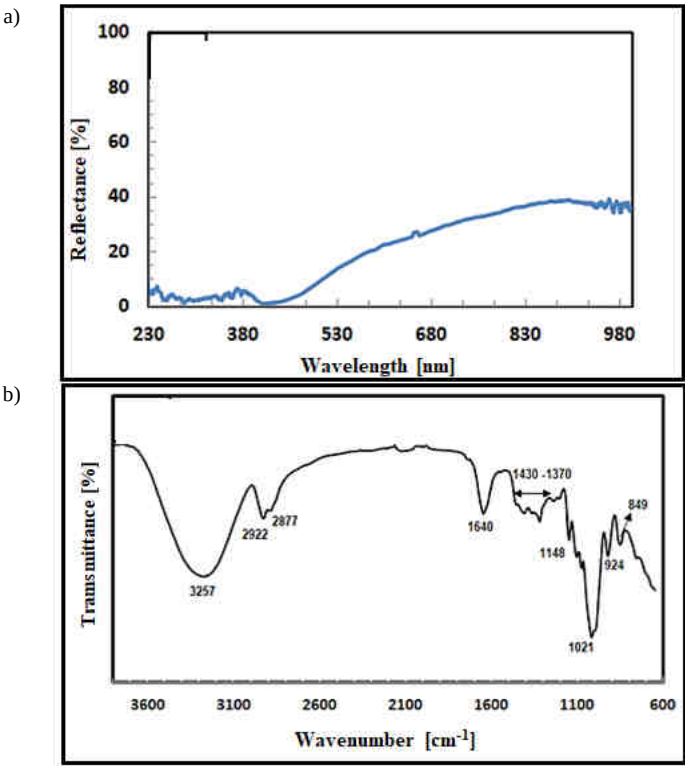


Fig. 2. a) UV-Vis-NIR spectrum and b) FTIR spectrum of the obtained bioplastic film

Figure 2a shows the UV-Vis-NIR reflectance spectra of the produced bioplastic film. As seen in Figure 2a, the overall shape of the spectra indicates that the bioplastic film has more intense peak points in UV-Vis region than in NIR region. This behaviour is expected as the bioplastic film is based on seaweed and supplements (starch, chitosan, and glycerol), which absorb light below 500 nm. Furthermore, the absorbance and reflectance of light in

NIR spectral range (~700 nm to 2500 nm) for plastic samples are due to overtone or combination vibrations of C-H, O-H, N-H and C-O in resin molecules, and the spectra are independent of colour in this region [16]. As presented in Figure 2, the spectral peak points above 940 nm to 1050 nm are weak and resemble commercial resin-based plastic material samples [16].

The colour of the bioplastic film was also identified by Commission Internationale de l'Eclairage values (CIE $L^*a^*b^*$) where L^* is lightness, a^* is redness and b^* is yellowness (details are given in SI, spectroscopic analysis). While * denotes the range of values; L , a and b are the observed values from the prepared bioplastic film. The colour measurements recorded are $L = 50.1$, $a = 5.1$ and $b = 38$. These indicate the film produced was having good lightness with reduced redness and more yellowness. The white index (WI) value was calculated to be 37.07. This is an optical value that indicates how white a material should be. WI of the produced bioplastic film is equal to and better than the plastic materials such as bio-based high density polyethylene (HDPE), bio-based low density polyethylene (LDPE) and petrol-based LDPE that have comparable thickness with the produced film in this study [17].

FTIR spectra (Fig. 2b) showed the complex nature of the bioplastic film with prominent peaks at (3257, 2922, 2877, 1640, 1319, 1148, 1103, 1021, 1013, 924, and 849) cm^{-1} . The broad strong peak at 3257 cm^{-1} is due to the merging of -OH, -NH stretching groups from *G. edulis*, starch, chitosan and glycerol which are the ingredients of the film [18-21]. Peaks at 2922 cm^{-1} and 2877 cm^{-1} are due to stretching vibrations of the C-H bond, usually from starch, glycerol and chitosan [19, 21]. Furthermore, from Figure 2b, the peak at ~1640 cm^{-1} is attributed to the asymmetric C=O stretch of COOH, and the peak at 1021 cm^{-1} is due to C-O bands dominant in *G. edulis* [18]. There are some peaks at ~1430 cm^{-1} to ~1370 cm^{-1} (due to $-\text{CH}_2$ stretching) and ~1148 cm^{-1} and 1013 cm^{-1} (due to $-\text{CO}$ and $-\text{C}-\text{O}-\text{C}$ groups) owing to starch and chitosan's presence in the prepared bioplastic film [20, 21]. Peaks at 924 cm^{-1} , 849 cm^{-1} attributed to the C-O-C bridge of 3,6 anhydrogalactose and D-galactose-4-sulfate which are common groups of a polymeric polysaccharide from *G. edulis* extract [22].

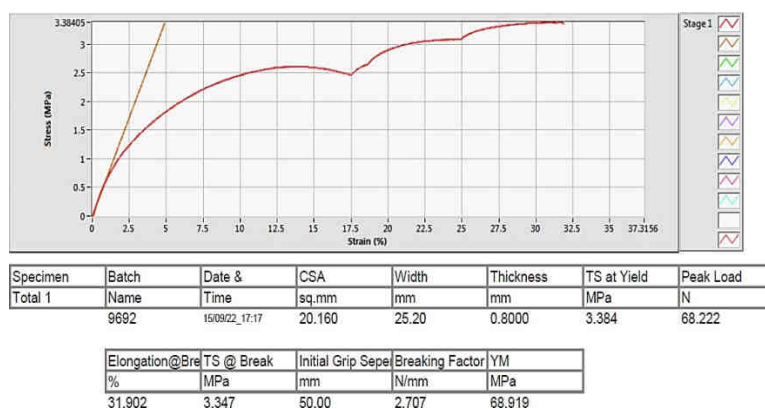


Fig. 3. Mechanical properties of the produced bioplastic film based on *G. edulis* extract

Figure 3 shows the mechanical properties of the produced bioplastic composite. The results indicate the composite has decent mechanical strength with good flexibility. The maximum force the film can withstand during its pull i.e., tensile strength, was observed to be 3.384 MPa and the stretching potential amount measured to be 31.902 %. With a difference in tensile elongation, the observed tensile strength values of the composite are very near to the starch-based bioplastics [23]. Reported studies infer that a decrease in tensile elongation reduces the flexibility of the bioplastic by enhancing the interactions within the composite and facilitating the free volume reduction between the biopolymer chains. For instance, starch-based bioplastics with more tensile strength and decreased elongation are preferred to produce more robust bioplastic materials. Thus, additional ingredients like TiO_2 are intentionally added to attain an anti-plasticiser effect [23]. But in this study, the as-prepared bioplastic composite material displayed desired characteristics like increased tensile strength with suitable lower elongation strength than the reported studies without additional fabrication processes, additives, or inorganic supplements.

Material migration test

Chemical compounds may be transferred to the food if the obtained bioplastic material is used as a packaging material [24]. Hence, the potential migration assessment of material from bioplastic film will give us an idea regarding its applicability to various foods. Three solvents were used to estimate the bioplastic material migration from the prepared bioplastic film sheets. Table 2 indicates the weight loss followed the order water > acetic acid > ethanol. This behaviour indicates that the material has more affinity to water, which is a polar solvent and is prone to dissociate in 96 h. More dissociation in water helps these materials to be bioavailable to the microorganisms and enable prompt degradation in the environment after disposal. With the migration results, it is stated that the developed bioplastic material could be more beneficial for foods with less moisture.

Table 2

Weight loss from bioplastic film in various concentrations of specific food stimulants

Parameter	3% Acetic acid	10% Ethanol	Distilled water
Weight loss [%]	8.69	5.88	10.2

Biodegradability and compostability

Biodegradation represents the breakdown of organic materials by microorganisms such as bacteria and fungi in abiotic and biotic environments [25, 26]. During the biodegradation process, microorganisms grow by utilisation of polymers through deterioration, fragmentation, mineralisation, etc., and convert organic carbon to CO_2 under aerobic conditions and CH_4 in anaerobic conditions [25]. For biodegradation investigation, *E. coli*, *R. rubra* and *F. solani* are selected as they are commonly found in air, soil and water environment [27-29]. As shown in Figure 4a, microorganisms exhibited good growth in defined media. However, it is observed that bioplastic solutions as sole substrates displayed promising growth with the tested microorganisms. The differences in the optical density O.D values from controls are observed to be ~38 %, 35 % and 34 % for *E. coli*, *R. rubra* and *F. solani*. This states that the performance order for degradation is in the order *F. solani* > *R. rubra* > *E. coli*. Thus, bioplastic solutions as sole substrates took

a significantly short time interval for the biodegradation process, which states its common behaviour under aerobic environments with ambient conditions [25].

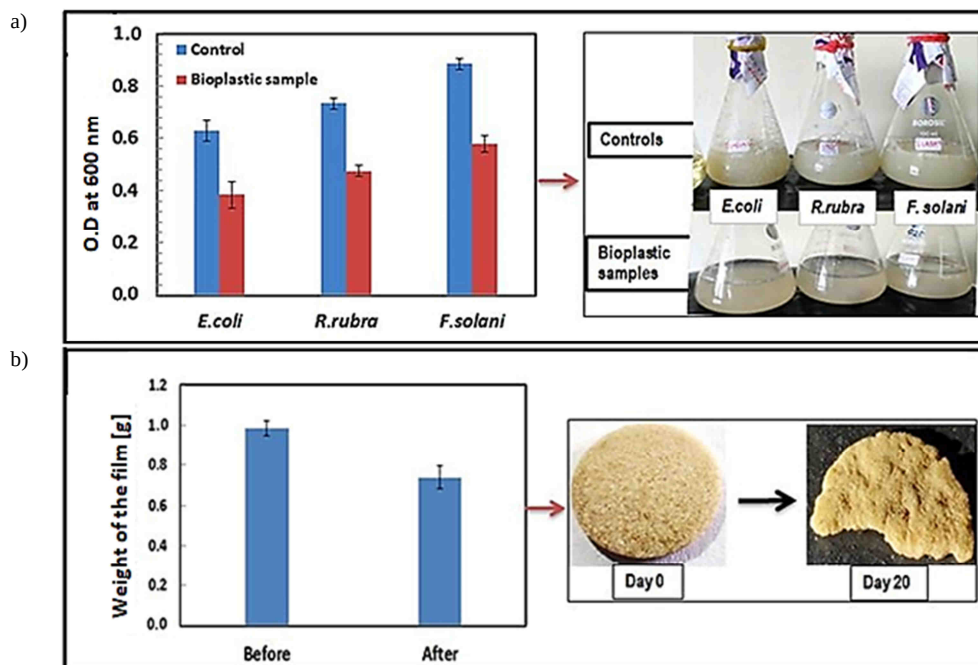


Fig. 4. a) Biodegradability and b) compostability assessment of bioplastic film prepared by *G. edulis* extract. Incubation time for samples for biodegradation tests are 48 h for *E. coli* and *R. rubra* and 72 h for *F. solani* (a) and it is 20 days for compostability tests (b)

In general, the composting of bioplastic films is influenced by nutrient availability, microbial consortia and soil temperature. Composting is routinely considered as an aerobic biodegradation process in a solid phase under controlled conditions [25]. Weight loss and physical appearance are direct inferences pertaining to compostability and are observed in this study. Figure 4b shows the weight loss from the bioplastic film after 20 days, and the change in its physical appearance was noticeable. Figure 4b illustrates that the composting conditions resulted in texture loss of the produced biofilm, which is a favourable sign for compostable polymers [25, 30]. This behaviour might be a result of the common biodegradation activity of various microorganism classes like bacteria, fungi, and yeast that have been known to aid in the biodegradation of polymers under composting conditions [25]. Thus the obtained *G. edulis* based bioplastic material has promising scope to be used as a common plastic alternative for food packaging and may solve the issues regarding the chemical modification of starch and cellulose-based plastic materials that are relatively expensive and possess a high impact on biodiversity.

Conclusion

In conclusion, this study introduces a pioneering approach in the realm of sustainable materials with the development of a novel bioplastic derived from *G. edulis* extract. This work comprehended various aspects, from preprocessing of algal biomass to the preparation of composite films using starch and chitosan supplementation. Notably, the resulting bioplastic films exhibited a remarkable resemblance to commercial plastic sheets in terms of microscopy and colour values. The structural stability of the composites was confirmed through infrared analysis, while mechanical tests showcased decent strength and flexibility akin to conventional starch-based bioplastics. Moreover, the material migration study highlighted its affinity for water and potential use in low-moisture food packaging. Equally promising was the film's behaviour upon exposure to microorganisms, displaying favourable biodegradation (within 48 h - 72 h) and composting (within 20 days). This eco-friendly bioplastic film, rooted in seaweed's renewable properties and benefiting from an uncomplicated production process, emerges as a compelling contender for large-scale, sustainable material production.

Acknowledgements

The authors thank Department of Biotechnology (DBT) for their financial support under Marine Bioresource and Biotechnology Network Programme, reference number: BT/PR40444/NDB/39/747/2020 and also the Vel Tech Rangarajan Dr. Sagunthala R&D Institute of Science & Technology, India for providing the Centre for Interfaces & Nanomaterials laboratory facility to carry this research work. The work is also supported by Amrita School for Sustainable Futures (ASF), Amrita Vishwa Vidyapeetham, Amrita University, Kollam, Kerala, India.

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