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Development of a pH-Responsive Mineral–Biopolymer Hybrid Bioplastic Derived from Eggshell Waste and Tamarind Seed Polysaccharides

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Abstract

Globally, the amount of non-biodegradable plastic waste is increasing, posing a serious threat to the environment. Conventional plastics made from petroleum resources pollute soil and water habitats and linger in the environment for long periods of time. Biodegradable materials made from recyclable and waste-derived materials are becoming more popular as sustainable solutions to this problem. In this study, tamarind seed kernel was used as the main polymer source to create a biodegradable bioplastic. Natural polysaccharides found in tamarind seeds, a common agro-industrial by-product, can be used to make polymeric films. Citric acid was used as a crosslinking agent to increase structural stability, and glycerol was used as a plasticizer to increase the polymer matrix's flexibility. To further strengthen the composite material, calcium carbonate made from eggshell waste was added as a reinforcing filler. Butterfly pea flower extract, which is abundant in natural anthocyanin pigments, was utilized to provide pH-responsive colour behaviour to further illustrate functional qualities. Solution casting was used to create the bioplastic film, which was then completely dried. The sustainable method for turning food and agricultural waste into biodegradable polymer materials that could be used in eco-friendly packaging and intelligent indication systems is demonstrated in this paper.

Keywords: *Butterfly pee Flower; Biopolymer Composite; Eggshell Calcium Carbonate; Sustainable Materials; Tamarind Seed Polysaccharide*

1. Introduction

Plastics made from crude oil are a big problem for our planet. They don't break down easily and last for a very long time, causing harm to our environment and health. When we use too much of these plastics, they build up in our land and water, leading to long-term damage to our ecosystems. They even break down into tiny pieces called microplastics, which are bad for animals and people. To fix this issue, scientists are working hard to create new materials that can break down naturally and are made from renewable resources like plants. One idea is to use waste from farms to make biodegradable materials that can replace traditional plastics in packaging and disposable products, which would be much better for the environment (Chen et al., 2025) . So, when we think about natural polymers, one type that really stands out is polysaccharide-based materials. They're great because they can form films, are abundant, and work well with eco-friendly processing methods. Let's take tamarind seed kernel polysaccharide, for example. It's a natural hydrocolloid that comes from the seeds of the Tamarindus indica plant and is mainly made up of galactoxyloglucan chains with lots of hydroxyl functional groups. These groups are useful because they allow us to modify the polysaccharide chemically, cross-link it, and even combine it with other materials. This makes tamarind seed polysaccharide a great candidate for developing biodegradable films. Recent research has shown that this polysaccharide is not only biocompatible but also has antioxidant properties and is good at forming strong films. Because of this, it has a lot of potential applications, such as food packaging, biomedical materials, and biodegradable polymer systems. This is exciting because it could help us move away from materials that harm the environment and towards more sustainable options. The fact that tamarind seed polysaccharide can be used in so many ways makes it a very promising area of study. As we continue to learn more about this natural polymer, we may discover even more ways it can be used to benefit our planet (Raj & Lee, 2024). Recent research studies have shown that tamarind seed-derived materials, such as tamarind seed starch and polysaccharides, can successfully be used as matrices for the development of sustainable packaging materials and biodegradable films. The valorization of tamarind seed waste is of great significance, as the processing industries of tamarind fruits generate a huge number of seed by-products, which are often not utilized properly. The use of tamarind seed waste for the production of biodegradable materials is a major step forward for the sustainable management of resources (Jiang et al., 2026). Although the advantages of polysaccharide-based bioplastics are many, the disadvantages, which are generally common with bioplastics, are that they are not mechanically strong, are highly sensitive to moisture, and lack structural strength compared to conventional synthetic plastics. To overcome the problems, the use of reinforcing fillers and cross-linking agents with biodegradable polymers has been researched. The addition of mineral fillers can improve the mechanical strength, thermal properties, and barrier properties of biopolymer films (Chen et al., 2025). Eggshell waste has also attracted recent attention for use as a green calcium carbonate source for reinforcement in polymers. Generally, eggshells possess over 90 % calcium carbonate along with minor percentages of proteins from the organic matrix. The food processing sector and domestic consumption contribute to a vast amount of waste from eggshells. The disposal of such waste causes environmental pollution. Recent studies have revealed that calcium carbonate obtained from eggshells can be effectively utilized for enhancing the mechanical, thermal, and barrier properties of biodegradable polymers while at the same time helping in waste reduction (Piras et al., 2024). Some recent research has also found that eggshell fillers can greatly improve the structural properties of biodegradable materials. The improvement of the properties of biodegradable materials with the help of calcium-based materials derived from eggshells has been found to enhance stiffness, as well as the tensile strength of the materials. The functional properties of the materials were also found to be improved with the help of eggshell fillers (Ormazábal et al., 2024). In view of these aspects, in the present study, an attempt is made to develop a biodegradable bioplastic based on tamarind seed kernel polysaccharide and calcium carbonate derived from eggshell wastes. Some studies show that adding Butterfly Pea extract can indicate pH by changing the colour in material, which can be incorporated in the material. In addition, bioplastic films were fabricated by employing a solution-casting technique with a plasticizer like glycerol and a cross-linking agent like citric acid. In addition, characterization of prepared bioplastic films was carried out using Fourier Transform Infrared Spectroscopy (FTIR), X-ray diffraction, ultraviolet-visible spectroscopy, and microscopic studies. The aim of the present study is to assess the possibility of utilizing agricultural and food waste as a source for developing eco-friendly biodegradable bioplastics.

2. Method

2.1.Materials

Tamarind seeds, belonging to the Tamarindus indica species, were sourced from food processing waste. Eggshell waste was sourced from kitchen waste. Butterfly Pea flower was sourced from local flower shops was added, Citric[1] acid and glycerol, both analytical grades, were sourced from standard chemical vendors. Distilled water was used throughout the experimental process. Tamarind seed kernels are known to contain galactoxyloglucan polysaccharide with film-forming properties, which can be used to synthesize biodegradable polymers (Gamage et al., 2024).

2.2.Extraction of Tamarind Seed Kernel Polysaccharide

Washed tamarind seeds were then dried at room temperature using distilled water to remove impurities. The tamarind seeds were then roasted slightly at 110°C for 15-20 minutes to remove the outer seed coat. The kernels were then separated and ground into coarse particles. The kernels were then soaked in water and then heated at 90-95°C for 60 minutes with constant stirring to extract the polysaccharide content. The solution was then filtered using muslin cloth to remove any impurities. The filtrate, which contains tamarind seed polysaccharide[2], was then concentrated by heating at 70-80°C to obtain a uniform solution. Similar extraction methods were used to obtain tamarind seed polysaccharide in recent studies on biopolymer research (Ren et al., 2022).

2.3.Preparation of Eggshell Calcium Carbonate

Eggshells were cleaned with the use of distilled water to remove any impurities. They were then dried at 80 °C for 2 h. The inner membrane was removed manually, and the shells were crushed into fine powder with the use[3] of a mechanical grinder. A 100-mesh sieve was used to sieve the powder to obtain calcium carbonate particles. Eggshells contain more than 90 % calcium carbonate[4]. Eggshells have been used as natural fillers in biodegradable polymer composites (Azarian & Sutapun, 2022).

2.4.Bioplastic Film Preparation

The extracted tamarind seed polysaccharide solution was then mixed with glycerol, which acted as a plasticizer, in approximately 10% (w/w) content with respect to the total content of the polymer. In addition, citric acid (2–3% w/w) was mixed with the solution to act as a cross-linking agent to improve the intermolecular bonding between the polymers. Next, calcium carbonate powder derived from eggshell (5–10% w/w) was slowly mixed with the solution with continuous stirring. The solution was then subjected to heat treatment at 80–90°C for 20 minutes to improve cross-linking between the polymers, resulting in a homogeneous solution. Then the Butterfly Pea Flowers[5] were boiled with distilled water at 80oC for 10 minutes to extract the pigments which act as a natural pH responsive material (Hasanah et al., 2023). The solution was then poured onto petri dishes to produce bioplastic films using the solution casting method. Similar methods have been used to produce biodegradable polysaccharide-based bioplastics (Mathew & Oksman, n.d.)

2.5.Characterization of optimized and control bio plastic films

2.5.1. Physical properties analysis

Thickness A digital outside micrometer (Insize, SL-M, India) was[6] used for the measurement of the thickness of the bio-plastic as per the method described as (Oluwasina et al., 2018) specified by Eq. 1. The average of ten measurements taken at different points was determined.

$$\text{Thickness} = \frac{\text{Sum of Measured Values}}{10} \qquad (1)$$

Moisture Content The initial weight (W1), as obtained from the digital weighing scale, of each film sample was used to determine the moisture content of the film samples[7]. After drying the film samples for 24 hours at 100°C in an oven, each film sample was re-weighed to obtain the final weight (W2). The moisture content (MC) of each film sample can be computed by applying Eq. 2 (Hazrati et al., 2021).

$$\text{Moisture Content}\,\% = \frac{W1-W2}{W1} \times 100 \qquad (2)$$

Biodegradability test In order to calculate the percentage of biodegradation, the method of slight modifications as shown in Eq. 3 was used. To test for the biodegradability of bioplastic[8], pieces of 2 cm2 of film were first weighed before composting (W1). The weight of residues was cleaned and then re-weighed for the final weight (W2) after 1 month of storage period at room temperature (Campos et al., 2025).

$$\text{Biodegradability}\,\% = \frac{W1-W2}{W1} \times 100 \qquad (3)$$

2.5.2. Mechanical properties analysis

Tensile strength (MPa) The texture analyzer (AMETEK, Texture Analyzer TA1, USA) was used for determining the film's mechanical properties using methods described by (Yadav et al., 2023). The film sample of dimensions close to 1.5 cm by 5 cm was fixed using tensile grips. During stretching, the force (N) and deformation (mm) were recorded. The force (N) and deformation (mm) were recorded during stretching. The tensile strength (TS) of each sample was calculated[9] using the maximum force and dividing it by the specimen's original average cross-sectional area. The resulting values were reported using mega pascal (MPa) using the formula shown below and reported to three significant digits as shown in Eq. 4.

$$\text{Tensile Strength (MPa)} = \frac{\text{Fmax}}{\text{A}} \qquad (4)$$

where A is the sample's cross-sectional area (mm2) and Fmax is the maximum stress (N) required to tear it apart.

Absorption of Water To find the absorption of water by the bio-plastic samples, the bio-plastic samples are dried in an oven at 100°C for 24 hours to find the initial dry weight of the bio-plastic samples (W1). Then the bio-plastic samples are left in a beaker with 50 mL of distilled water at room temperature for 24 hours. After that, the water is filtered and the weight of the bio-plastic sample is found (W2). The amount of water absorbed by the bio-plastic film is found using Eq. 5.

$$\text{Water absorption \%} = \frac{W2-W1}{W1} \times 100 \qquad (5)$$

2.5.3. Thermal properties analysis

Differential scanning calorimeter (DSC) analysis bio plastic film Thermal characteristics of the biofilm were evaluated according to the procedure of (Tarique et al., 2021). By heating the sample at a rate of 10 °C per minute, the differential scanning calorimeter was utilized to examine the results of the DSC test. Next, the sample pan was heated in an atmosphere of nitro gen from 20 to 250˚C at a rate of 10˚C each minute.

2.5.4. Functional group, structural and morphological analysis

XRD analysis The XRD analysis was carried out according to the procedure given by (Vilas Dhumal et al., 2019). For this purpose, a 40 kV X-ray diffractometer with a 40 mA current was used. The film samples were kept in a saturated relative humidity chamber overnight before performing the XRD analysis. The samples were then subjected to scanning within a 5° to 60° 2θ range using X-ray radiation with a wavelength of 1.57 Å, i.e., Co Kα.

FTIR Analysis The samples were pressed into pellets and placed on the sample holder. IR light was applied, and data was collected at 32 resolutions and 16 scans. The 400–4000 cm−1 range was used to record the spectra[10 – 15].

UV-Vis Spectroscopy The optical properties of the prepared bioplastic films were investigated using a UV-Visible Spectrophotometer. The test was conducted over a wavelength range of 250-750 nm. The prepared bioplastic film was cut into uniform sizes and placed in the sample holder of the spectrophotometer[16]. The test was conducted using a blank. The spectrum of the prepared film was recorded over the selected wavelength range. The results were used for determining the UV absorption and transparency of the film (Jafarzadeh & Jafari, 2021).

3. Results And Discussion

3.1. X-ray Diffraction (XRD) Analysis

X-ray diffraction analysis was carried out to identify the structural characteristics of the developed bioplastic film. Figure 1 shows the XRD pattern obtained for the film. The diffraction pattern shows a broad peak in the region between 20° to 30° (2θ), with a maximum intensity peak at an angle of around 23-25°. This suggests that the film is mostly in an amorphous state[17]. This is a characteristic feature of a olysaccharide-based biodegradable polymer matrix. The absence of sharp crystalline peaks in the diffraction pattern suggests that the polymer chains in the polysaccharide-based film are disordered in nature. This is because of the disordered arrangements of polysaccharide chains in the film. Although there are minor intensity changes in the diffraction range, there are no strong crystalline peaks, suggesting that the eggshell-derived calcium carbonate particles are well distributed in the polymer matrix or that they are present in small amounts. The amorphous characteristics of the particles are advantageous in the formation of biodegradable films, as they provide flexibility and regularity in the film structure. In conclusion, the XRD pattern confirms that the prepared material is an amorphous biopolymer composite reinforced with minerals, typical of biodegradable polysaccharide-based plastic films.

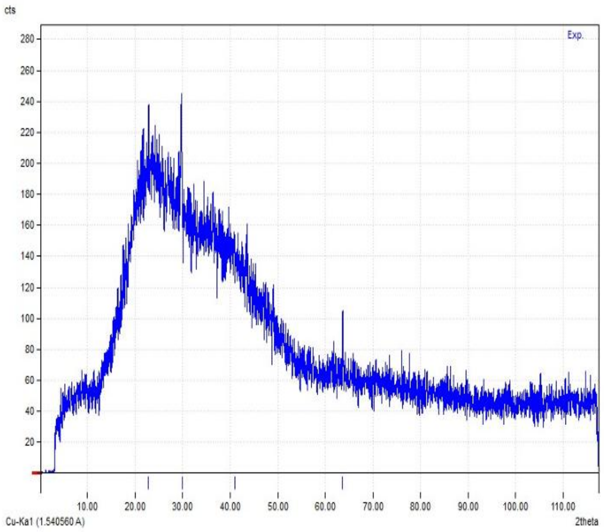

Figure 1 **X-ray Diffraction (XRD) Analysis**

3.2.FTIR Analysis

The FTIR spectroscopy method was utilized to identify the functional groups and intermolecular interactions within the prepared bioplastic film. The FTIR spectrum (Figure 2) shows various characteristic peaks related to the polysaccharide structure and cross-linking interactions. The broad and strong absorption peak observed between 3200-3400 cm^{-1} (Peak 1) relates to the O-H stretching vibrations, which are characteristic for hydroxyl groups present within the polysaccharide structure. The broadness of this peak indicates the high degree of hydrogen bonding within the tamarind seed polysaccharide structure. The peak observed near ~2900 cm^{-1} (Peak 2) relates to the C-H stretching vibrations, which are characteristic for aliphatic carbon present within the polysaccharide structure. The weak peak observed near ~2100-2200 cm^{-1} (Peak 3) might be related to the minor contributions from intermolecular interactions or functional groups present within the composite material.The peak observed near ~1600-1650 cm^{-1} (Peak 4) relates to the C=O stretching or the presence of bound water, which might be related to the interaction between the citric acid and the polysaccharide chains. The peaks present at ~1400 cm^{-1} (Peak 5) are generally related to the C-H bending vibrations. The strong bands of absorbance, appearing between ~1000 and 1150 cm^{-1} (Peak 6, 7, and 8), relate to the presence of C-O-C and C-O stretching vibrations, generally related to the glycosidic bonds present in the polysaccharide structure. The peak present at ~900 cm^{-1} (Peak 9) might be related to the structural vibrations of carbohydrate rings. The results of the FTIR analysis confirm the presence of the polysaccharide groups and the successful formation of the polymer network in the bio plastic.

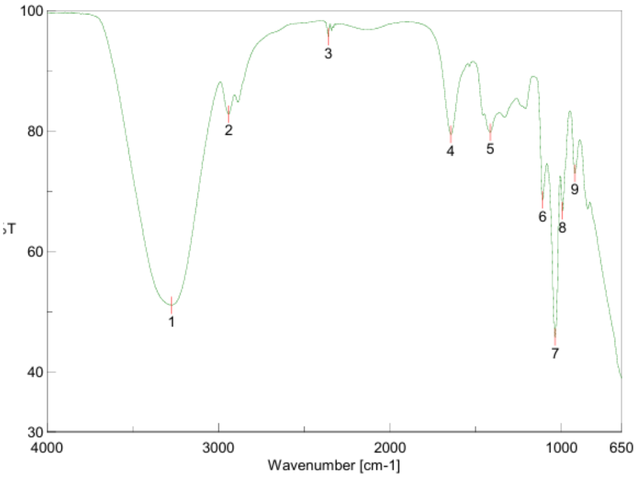

Figure 2 **Ftir Analysis**

3.3.UV–Visible Spectroscopy Analysis

The optical property of the developed bioplastic film was examined using UV-Visible spectroscopy in the range of 250-750 nm. The UV-Vis spectrum of the composite film (Figure 3) shows high values of absorbance in the ultraviolet region below 320 nm, then gradually decreases with increasing wavelength. The high values of absorbance in the range of 260-280 nm indicate that bioplastic film can absorb ultraviolet radiation. This may be attributed to the electronic transitions of the polysaccharide groups and the interaction with the minerals in the composite material. As the wavelength increases to move towards the visible region, the absorbance decreases gradually. The values of absorbance are low after 600-700 nm. This shows that the film has a certain degree of transparency to visible light. The observed property of absorption of UV light by this film suggests that this prepared bioplastic film has the

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potential to act as a UV shield.

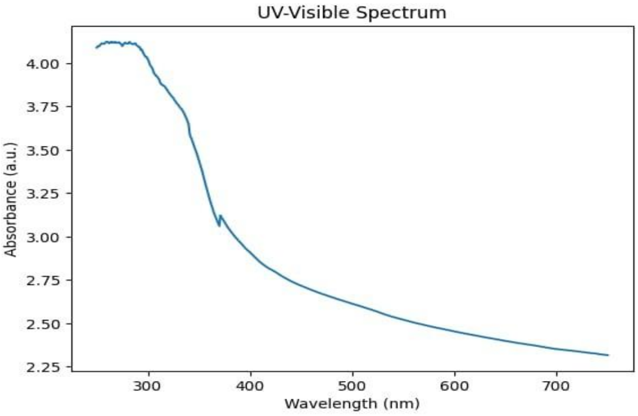

FIGURE 3 UV–Visible Spectroscopy

3.4.Moisture Content Test

The initial weight (W_1) of each bioplastic film sample was recorded using a digital analytical balance. The samples were then dried in a hot air oven at 100°C for 24 hours to remove absorbed moisture. After drying, the films were cooled in a desiccator and reweighed to obtain the final dry weight (W_2). The moisture content (MC) of the film samples was calculated using the following equation: shown as Table 1 Moisture Content Test Result

$$Moisture\ Content\ \% = \frac{W1-W2}{W1} \times 100 \qquad (6)$$

Table 1 Moisture Content Test Result

Parameter	Value
Initial Weight (W1)	0.500 g
Final Weight (W2)	0.470 g
Moisture Content	6 %

3.5.Film Water Solubility

Small samples were dried to constant weight at 100°C for a day in order to determine their initial dry mass (W1). Then the specimens were kept in 50 ml distilled water at room temperature for one day, stirring the solution occasionally. Afterwards, the remainder of the bio-plastic was dried to constant weight at 100°C for another 24 hours following filtration of the water.

$$Swelling\ Index\ \% = \frac{Ws-Wd}{Wd} \times 100 \qquad (7)$$

Table 2 Film Water Solubility Test

Parameter	Value
Dry Weight (W_d)	0.500 g
Swollen Weight (W_s)	0.497 g
Swelling Index	0.52 %

3.6.Biodegradability Test

The biodegradability of the developed hybrid bioplastic was evaluated using the soil burial method. Pre weighed film samples (W_0) were buried in natural soil at a depth of approximately 5 cm under ambient environmental conditions for 30 days. After 30 days, the samples were removed, washed gently to remove soil particles, dried at 60°C until constant weight, and reweighed (W_1). The percentage weight loss was calculated using:

$$Weight\ Loss\ \% = \frac{W0-W1}{W0} \times 100 \qquad (8)$$

Parameter	Weight loss after 10 days	Weight loss after 20 days	Weight loss after 30 days
Initial Weight (W_0)	0.500 g	0.500 g	0.500 g
Final Weight (W_1)	0.41 g	0.29 g	0.155 g
Weight Loss	18 %	42 %	69 %

Table 3 Biodegradability Index

3.7.Swelling Behavior

The film samples were immersed in distilled water for 24 hours at room temperature. The swelling index was calculated using:

$$Swelling\ Index\ \% = \frac{Ws-Wd}{Wd} \times 100 \qquad (9)$$

Parameter	Value
Dry Weight (W_d)	0.500 g
Swollen Weight (W_s)	0.600 g
Swelling Index	20 %

Table 4 Swelling Index

3.8.Film Thickness

The thickness of the prepared bioplastic films was measured using a digital micrometer at five different random positions, and the average value was recorded.

Table 5. Film Thickness

Measurement	Thickness (mm)
1	0.182
2	0.176
3	0.185
4	0.179
5	0.181
Average	**0.181**

3.9.Thermogravimetric Analysis

The Thermogravimetric Analysis (TGA) technique was conducted to analyze the thermal stability and the degradation properties of the synthesized bioplastic material. Initially, a mass loss was recorded below 100°C attributed to the vaporization of water molecules and other volatiles from the sample. Continued heating led to the decomposition of the polymer structure within the range of 100 °C and 220 °C, denoting the degradation of the natural biopolymer structure. The heat flow pattern revealed the thermal phase transformations and changes during the decomposition process. A minimal residual mass was left under high temperatures corresponding to the thermally stable inorganic and carbonized constituents.

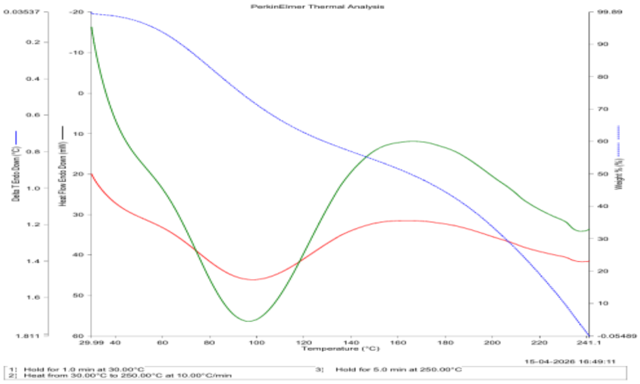

Figure 4 Thermogravimetric Analysis

3.10. Tensile Strength

This tensile strength test was conducted to examine the mechanical behavior of the bioplastic that was produced. As can be seen from the graph, the tensile strength slowly increased as the elongation level increased. This demonstrates that the material had decent stress-carrying capabilities and flexibility. The material attained a maximum tensile strength of about 6 MPa at an elongation level of around 4.2%. The abrupt fall in the graph indicates the failure point of the material. These findings suggest that the fabricated bioplastic had moderate mechanical strength and flexibility, which made it appropriate for use as a sustainable packaging material.

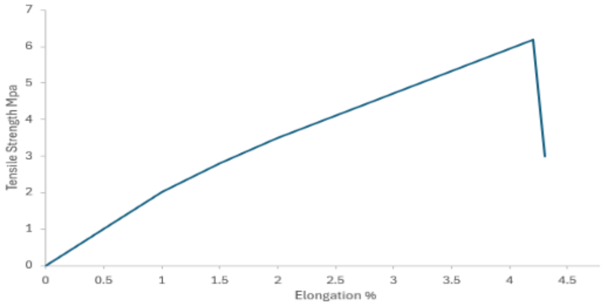

Figure 5 Tensile Strength

Conclusion

In this research, a biodegradable bioplastic material was successfully formulated from a natural polymer source derived from tamarind seed polysaccharide reinforced with calcium carbonate derived from eggshells. The use of agricultural waste materials offers a viable solution in developing a sustainable source for producing a biodegradable polymer composite material. XRD results indicated that the prepared bioplastic material exhibited a semi-amorphous characteristic with a diffraction peak ranging from 20 to 30° (2θ). This characteristic is common in polysaccharide-based materials. The semi-amorphous characteristic allows a certain degree of flexibility in polysaccharide-based materials. The FTIR spectrum results indicated the presence of characteristic absorption bands corresponding to polysaccharide-based materials. The absorption bands observed in this research correspond to the presence of a hydroxyl group (–OH), an aliphatic group (–CH), and a glycosidic linkage (–C–O–C). The UV-Visible spectroscopy analysis showed that the developed bioplastic strongly absorbs ultraviolet radiation with decreasing absorbance as the wavelength increases into the visible spectrum. This is important since the developed bioplastic can block ultraviolet radiation while at the same time showing moderate transparency. Overall, the analysis showed that the developed bioplastic is able to utilize the tamarind seed kernel and eggshell waste while at the same time showing desirable properties. Therefore, the developed bioplastic is a good example of a biodegradable polymer that is environmentally friendly.

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