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Sustainable bioplastics based on shrimp chitin: mechanical characterization and biodegradability evaluation

Jorge Braulio Amaya[1]* and **Paola Duque Sarango**[1]

[1]*Ingnieria Ambiental, Laboratorios de Ciencias de La Vida, Universidad Politécnica Salesiana, Cuenca, Azuay, Ecuador*

***jamaya@ups.edu.ec**

Abstract

Plastic pollution caused by synthetic polymers is a global concern demanding environmentally friendly alternatives. This study presents the development of a thermoformable bioplastic composed of shrimp-derived chitin, corn starch, sugarcane bagasse, glycerol, and acetic acid, integrated with polyethylene terephthalate (PET) and maleic anhydride as a compatibilizer. The composite underwent mechanical, thermal, and biodegradability assessments. The formulation containing 5% chitin achieved a tensile strength of 0.833 MPa and a density of 2780 Kg/cm^3, highlighting its mechanical viability. Under controlled composting conditions, degradation was observed in 120 days. Although the primary polymer matrix consists of petroleum-based PET, the term "bioplastic" is justified by the presence of renewable constituents and proven biodegradability, following the definition by European Bioplastics. This structure supports partial replacement of fossil-derived plastics while promoting sustainable waste management. The study underscores the potential of integrating agro-industrial residues into polymeric systems aimed at contributing to circular economy in material science.

Keywords: *bioplastic, biodegradability, chitin, extraction.*

Data Availability: Research data is available upon request from the corresponding author.

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1. Introduction

The environmental challenges posed by plastics derived from petroleum have reached critical levels, positioning them as one of the most significant ecological threats of the 21st century. Recent data indicates that global plastic production hit 400 million tons in 2022, with a mere 9% being recycled[1,2]. This extensive accumulation predominantly impacts marine ecosystems, where plastics constitute 80% of solid waste, thereby exacerbating biodiversity loss and inducing chemical changes in oceanic environments. In light of this situation, bioplastics have emerged as a viable alternative, offering functional properties akin to traditional plastics while also being biodegradable and renewable[3-5]. Notably, biopolymer-based materials such as chitin have shown promise in addressing plastic pollution[1,6]. Chitin, a polysaccharide found in the exoskeletons of crustaceans, represents a renewable resource with significant potential for sustainable applications[7-9]. Its chemical characteristics, particularly the presence of amino groups, render it a versatile candidate for the production of biodegradable bioplastics[9-12]. In Ecuador, the shrimp industry generates around 1.5 million tons of shrimp shells annually, presenting a unique opportunity to merge waste valorization with the creation of sustainable materials[13-16].

Nonetheless, its application in industry encounters notable hurdles, such as elevated production costs, complicated extraction processes, and limitations regarding the mechanical strength of the resulting plastics[17,18]. Nevertheless, the industrial adoption of chitin faces considerable obstacles, including elevated production costs, complex extraction processes, and limitations in the mechanical properties of the resulting plastics [19,20]. Current research endeavors are directed towards optimizing extraction techniques and integrating chitin with other natural polymers, such as starch and sugarcane bagasse, to enhance both the physical properties and overall performance of the materials [9,21,22].

The term *"bioplastics"* is used throughout this study in accordance with the definition by European Bioplastics, which includes materials that are biobased, biodegradable, or both. Although the composite includes petrochemical PET as the primary matrix, the incorporation of renewable components and the demonstrated biodegradability under composting conditions support the use of this terminology.

In this work, we aim to develop a thermoformable bioplastic using chitin extracted from shrimp exoskeletons and evaluate its feasibility. The specific objectives of this study are to optimize chitin extraction methods, formulate a competitive bioplastic, and characterize its mechanical, thermal, and biodegradability properties.

2. Methodology

2.1 Exoskeleton cleaning

In terms of exoskeleton preparation, 6 kg of shrimp exoskeletons from the CRAMAGROMAR company in Guayaquil, Ecuador, were employed for chitin extraction. The initial step involved cleaning the shells to remove any residual meat and contaminants. The cleaned shells were then dried in an oven at 90 °C for 5 hours until they reached a constant weight. Subsequently, the material was processed using a Polymix PX-MFC 90D mill to achieve a particle size of 300 microns, as indicated in the literature[23,24].

2.2 Obtaining chitin

The initial phase of the study concentrated on the extraction of chitin, utilizing shrimp exoskeletons as the primary source material. A 10% hydrochloric acid solution was employed to break down the shrimp exoskeletons, and this solution was maintained at a temperature of 37 °C for a duration of 12 hours.

2.3 Demineralization, deproteinization and discoloration

The exoskeleton, underwent a treatment process involving a 15% (v/v) acetic acid solution at ambient temperature for a duration of three hours, accompanied by agitation at 10 rpm. Following this, the material was subjected to vacuum filtration, rinsed three times with deionized water, and subsequently dried in an oven at 60 °C for six hours. A deproteinization step was then implemented utilizing proteolytic enzymes, specifically papain at an enzyme-to-substrate ratio (E/S) of 0.5:100 and chymotrypsin at an E/S ratio of 0.7:100, with continuous agitation at 140 rpm for three hours at 40 °C, maintaining pH levels of 8.7 and 8, respectively, to effectively decompose the proteins present in the peel. The decolorization process was executed using 96% ethanol at a 1:20 ratio under constant agitation, followed by washing with deionized water to eliminate pigments. The absorbance of the sample was subsequently measured using a UV-Vis spectrophotometer at 470 nm. Upon completion of these procedures, the resultant material was dried once more at 60 °C for six hours, yielding a purified chitin.

2.4 Obtaining bioplastic

Industrial cornstarch (Zea mays) served as a starch source and functioned as the foundational plasticizer for the blend. To enhance the rigidity and durability of the bioplastic, sugarcane cellulose was incorporated. The collected bagasse underwent disinfection, was air-dried, and then ground into a fine powder. Subsequently, it was subjected to hydrolysis using sulfuric acid at a concentration of 60% to extract the cellulose, which was then integrated into the primary bioplastic mixture. Additionally, chitin was incorporated into the bioplastic, along with other ingredients such as corn starch, cellulose from sugarcane bagasse, glycerol, and acetic acid. These components were selected for their environmentally friendly properties and their ability to act as plasticizers[10,25].

The study involved the creation of multiple formulations incorporating chitin alongside corn starch, as presented in Table 1, in addition to sugar cane cellulose, glycerol, and acetic acid. These materials were subjected to a heating process at 80 °C for 30 minutes.

2.5 Mechanical testing (elongation)

In order to assess the mechanical characteristics of the bioplastic, elongation tests were executed following the guidelines set forth by ASTM-D 638. Three bioplastic specimens were created, each conforming to the standard dimensions of 1 mm thickness, 1 cm width, and 12 cm length. These samples were subjected to tensile testing with a high-precision dynamometer capable of measuring forces up to 200 MPa. The breaking points of each specimen were recorded, and the average elongation was computed using the standard elongation formula. The average elongation calculations were derived from Equation 1:

$$eP = \frac{\sum m}{NT} \tag{1}$$

Where: Σm being the summation of the breakpoints and NT number of samples.

Additionally, its density was evaluated using the standard method for liquid density determination. For a sample of 100 g with a volume of 40 ml, the density was calculated using the appropriate formula, and this process was conducted in triplicate to establish an average value.

2.6 Injection

To create an ideal bioplastic, a mixture containing 10% biopolymer was combined with 90% commercial-grade PET, which is frequently utilized in extrusion and injection methods due to its excellent crystallinity and desirable mechanical characteristics. This specific PET has a melt flow rate of 25g per 10 minutes, determined at a temperature of 280 degrees Celsius with a load of 2.16 kilograms.

Table 1. Biopolymers Formulations.

Sample	% chitin	% starch	% cellulose	% glycerol	% acetic acid
M1	10	40	10	15	25
M2	10	40	10	15	25
M3	10	40	10	15	25
M4	9	35	9	15	32
M5	8	35	9	15	33
M6	8	35	8	15	34
M7	7	34	8	15	36
M8	6	33	7	15	38
M9	5	30	7	15	43

Because PET did not interact well with the bioplastic, an addition of 3% maleic anhydride grafted onto polyethylene terephthalate (PET-g-MA) based on total weight was made to enhance the compatibility between the chitin and the polymer matrix. The bioplastic was produced under strictly controlled extrusion settings using a Bossi NB62 plastic injector. The temperature along the barrel was gradually set in three different zones: 180 degrees Celsius in the feeding area, 200 degrees Celsius within the compression section, and 220 degrees Celsius at the dosing stage, to guarantee thorough mixing and to prevent the materials from experiencing thermal degradation. The screw speed was kept within a range of 50 to 100 revolutions per minute, operating under a pressure between 5 and 15 megapascals. The material remained in the extruder for 4 minutes, and it underwent rapid cooling to maintain its shape stability. Additionally, the chitin was pre-dried at 80 degrees celsius for a span of 24 hours to remove any moisture and prevent complications during manufacturing of biopolymer.

2.7 Biodegradability analysis

An biodegradability assessment through composting was performed, employing a method deemed the most straightforward and economical. The methodology involved the burial of 70 grams of granulated bioplastic in a wooden box, which was placed at a depth of 45 centimeters, as depicted in Figure 1. The setup began with a 10 centimeter layer of soil with organic residues. To facilitate the degradation process, 2 cc of Bacillus spp. diluted in 200 ml of water were added prior to covering the entire assembly with additional soil. This evaluation was carried out over 120 days to track the weight reduction of the bioplastic.

The temperature and relative humidity of the soil were assessed using a digital thermohygrometer with an external sensor (model TAYLOR 1523). This device recorded internal temperatures from -10 to +50 °C, external temperatures from -50 to +70 °C, and relative humidity levels between 20% and 99%. The sensor was inserted 2 cm into the soil, and data was collected periodically for analysis. Additionally, in situ observations were conducted on the physical transformation of the bioplastic, highlighting the role of organic residues and microorganisms in its biodegradation.

2.8 Thermogravimetric analysis (TGA)

This examination took place in a lab oven where the sample experienced stepwise increases in temperature, and the weight reduction was noted. A total of eight temperature steps were conducted beginning at 50 °C and concluding at 400 °C, increasing by 50 °C each time.

3. Results and Discussion

3.1 Determination of chitin percentage

During the first stage of the investigation, chitin was successfully isolated from shrimp exoskeletons utilizing an optimized enzymatic technique. The process resulted in the extraction of 55.7 g of pure chitin for every 100 g of sample material, a result consistent with findings from studies[26-30], which suggest that chitin constitutes approximately 40% to 50% of the chemical composition of shrimp shells, varying by species. The subsequent Figure 2(a) presents the dried shrimp shell, and Figure 2(b) illustrates the chitin with a granule size of 300 microns.

The measurement of depigmentation was conducted within the visible spectrum at a wavelength of 470 nm, yielding an absorbance value of 0.2783λ for papain and 0.3612λ for chymotrypsin (refer to Figure 3b). These absorbance values are indicative of the enzymatic efficiency of papain and chymotrypsin. The lower absorbance associated with papain implies a greater capacity for pigment removal in comparison to chymotrypsin. Consequently, this suggests that papain may be more effective in facilitating the depigmentation of shrimp shells under the specified experimental conditions.

The Figure 3 illustrates the sequential steps in the production of a chitin biopolymer combined with PET. In Figure (a), one can observe the deproteinized sample, which contains solid orange fragments. Figure (b) reveals yellowish and orange solutions, which signify the depigmentation process carried out by enzymes like papain and chymotrypsin. Figure (c) displays the bioplastic that has been produced, characterized by a notable presence of white or semi-translucent material. Finally, Figure (d) depicts the bioplastic as small granular fragments.

Figure 1. Box with compost used to place biopolymers.

Figure 2. (a) Dried shrimp shell, (b) ground and granulated chitin.

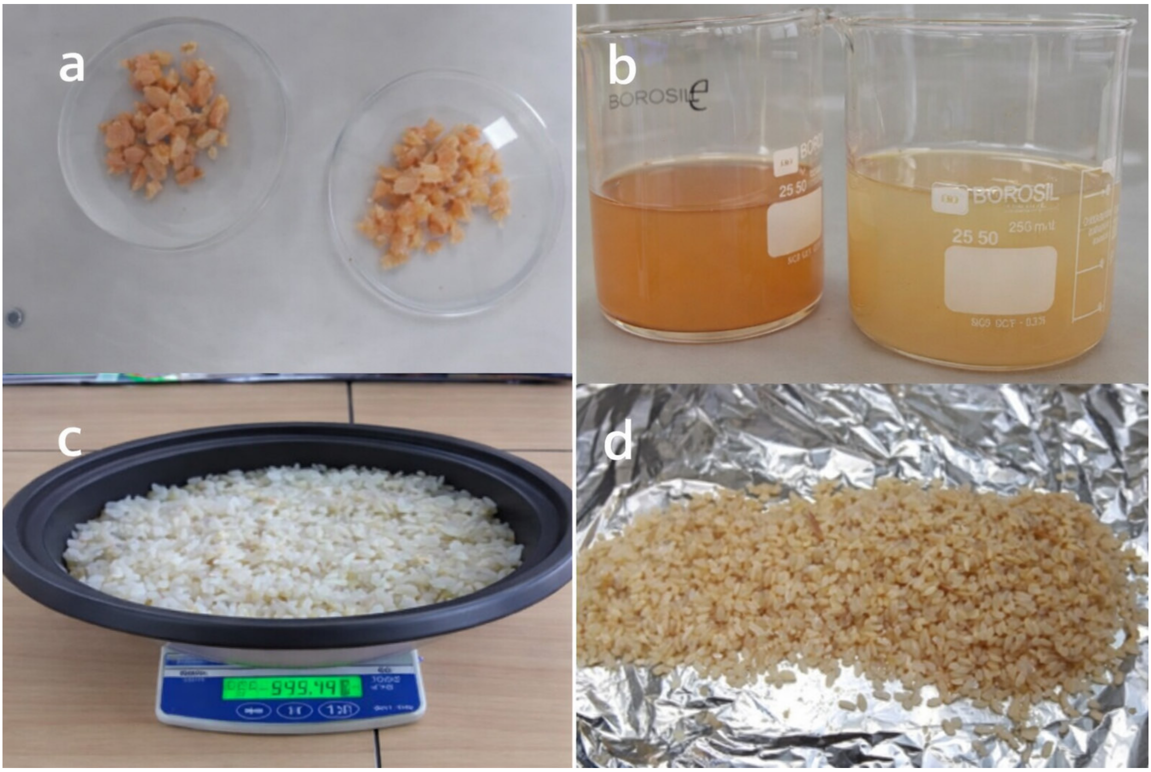

Figure 3. (a) deproteinized sample; (b) depigmented sample; (c) bioplastic obtained (d) granulometrated bioplastic.

Although enzymatic deproteinization represents a less aggressive alternative compared to traditional alkaline methods, the chitin extraction process used in this study still entails significant use of hydrochloric acid for demineralization, acetic acid for decolorization, and high-temperature drying steps. These factors introduce important environmental burdens that moderate claims of sustainability.

The life cycle assessment conducted by[31] on chitin nanocrystal production included in PLA/PET packaging materials reveals that even green extraction routes carry high impacts—particularly in terms of greenhouse gas emissions—when acid hydrolysis and drying energy are not optimized. The study shows that chitin nanoparticle production can contribute 3.21 kg CO_2eq for PET and 4.26 kg CO_2eq for ChNC/PLA composite, with potential reductions of 30–40% if HCl usage and energy consumption during drying are minimized.

Based on these insights, the process presented herein should be regarded as a partially improved alternative—not yet fully sustainable. For future developments, implementing acid recovery, reducing reagent concentrations, optimizing reaction conditions, and adopting solar-assisted or low-temperature drying technologies will be essential to improve the overall environmental footprint of chitin-based bioplastics.

3.2 Mechanical testing

In Table 2, the show the results that indicate that the first sample had a breaking point of 0.9 MPa, the second sample reached a breaking point of 0.6 MPa, and the third sample resulted in a breaking point of 1.0 MPa. As a result, the average elongation was determined to be 0.833 MPa, reflecting adequate resistance. Additionally, samples with lower chitin content were found to have inferior mechanical resistance, particularly sample 2.

In the research presented by[32-38] which similarly examined chitin, values of 0.6 and 0.9 MPa were reported, showing a strong similarity to the findings of the present study. However, the discrepancies may stem from differences in the methodologies or formulations utilized.

Table 2. Elongation test results.

Test No.	Breaking point
1	0.9 MPa
2	0.6 MPa
3	1.0 MPa
Average	0.833 MPa

3.3 Physical properties

The analysis of the samples revealed notable differences in biopolymer flexibility. Samples 7 to 9 demonstrated superior compactness and flexibility, with sample 9 exhibiting increased hardness and thermal resistance after complete drying. In contrast, sample 3 had lower flexibility and reduced thermal resistance, while sample 6 was fragile to the touch. Additionally, samples with more than 6% chitin showed structural inconsistencies, whereas those with 5% chitin maintained a manually flexible structure. These findings emphasize the impact of composition and treatment on the biopolymer's final mechanical properties.

3.4 Density test

Sample densities were determined through the application of the mass-volume relationship, with the results outlined in Table 3.

The average density measured was 2780 Kg/m^3, which bears a resemblance to the findings reported in studies[39,40] this outcome suggests that the product exhibits thermoformable characteristics, attributed to the inclusion of polyethylene, a material commonly utilized in applications where weight is a crucial factor, alongside enhanced mechanical strength and stiffness.

3.5 FTIR spectroscopic analysis

The infrared spectroscopy analysis of the chitin, PET, and maleic anhydride mixture reveals notable interactions among the components, suggesting an enhancement in polymer compatibility (refer to Figure 4). The blue spectrum,

Table 3. Sample densities.

No. Test	Mass (g)	Volume (ml)	Density (g/ml)	Density (Kg/m^3)
1	100	40	2,5	2500
2	120	40	3	3000
3	114	40	2,85	2850
Average	-	-	2,78	2780

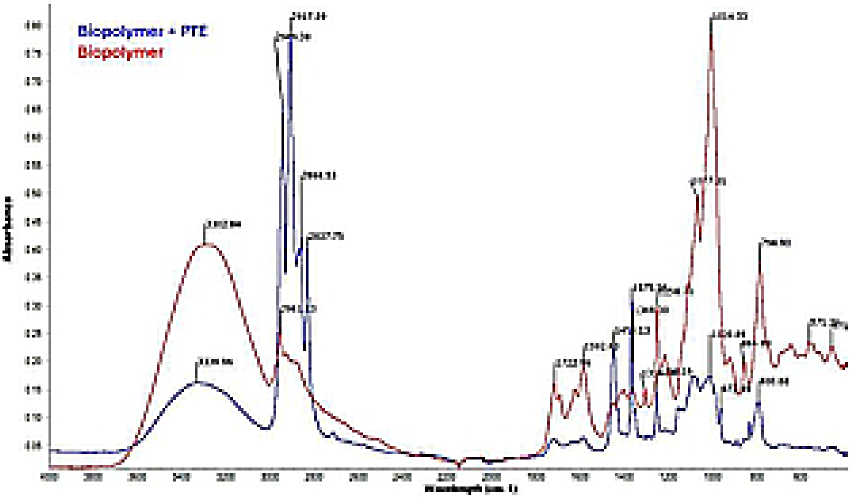

Figure 4. IR spectrum of the mixture of chitin, PET and maleic anhydride.

representing the biopolymer and PET blend, displays a peak at 3339.96 cm-1, which is associated with the -OH and -NH groups of biopolymer. This observation implies the potential formation of hydrogen bonds or reactions involving maleic anhydride. Furthermore, the peak at 1722.96 cm-1, indicative of the carbonyl group (-C=O) in PET, may reflect the influence of maleic anhydride, which exhibits bands in the 1780-1850 cm-1 range, suggesting the establishment of hydrogen bridge-type bonds with either chitin or PET, particularly as chitin is in a more hydrophilic state. Additional peaks at 1020.84 cm-1 and 1077.81 cm-1, characteristic of C-O-C stretching in esters, further imply interactions with maleic anhydride. A comparison with the red spectrum, which corresponds to pure biopolymer and shows a prominent band at 3302.84 cm-1, indicates that the introduction of the compatibilizer alters the polymer interactions[41]. Despite the inherent hydrophobic nature of PET and the hydrophilic characteristics of chitin, maleic anhydride serves as a coupling agent, facilitating reactions with the functional groups of chitin and potentially with the PET esters. This interaction enhances the adhesion between the phases and promotes a more uniform dispersion. The observation of shifts or broadening in significant bands would confirm the compatibilization of the mixture, thereby improving the integration of chitin within the PET matrix and augmenting its mechanical and structural properties for composite biopolymer applications.

3.6 Thermogravimetric analysis (TGA)

The results of the TGA analysis are presented in Table 4, which indicates an initial temperature range of 50-100 °C. This range corresponds to the initial moisture loss associated with cellulose, starch, and chitin, attributable to their hydrophilic characteristics. Subsequently, a temperature range of 150-250 °C is noted, which relates to the volatilization of glycerol and acetic acid, as these plasticizers exhibit low thermal stability and decompose swiftly within this temperature interval. Finally, the analysis reveals a range of 250-400 °C, during which thermal degradation of natural polymers occurs, leading to the decomposition of both cellulose and chitin, resulting in carbonaceous residues. It is also observed that there is an initial mass loss of 5% due to the components of cellulose, starch, and chitin. Furthermore, at temperatures exceeding 400 °C, a significant mass loss of 85% is recorded, with the remaining mass comprising non-volatile carbonaceous residues. These residues include amorphous carbon resulting from the incomplete thermal decomposition of organic materials such as cellulose, starch, chitin, and glycerol, as well as mineral ash derived from inorganic compounds present in the raw materials or produced through the degradation of salts or impurities, including metal oxides[42,43].

The graph under (Figure 5) consideration illustrates three fundamental stages of the decomposition process. The first

Table 4. Results of the thermogravimetric analysis.

Temperature (°C)	Mass Loss (%)	Behavioral Description
50	5	Loss of adsorbed moisture (cellulose, starch and chitin).
100	12	Water desorption and initial volatilization of acetic acid.
150	23	Partial decomposition of starch and glycerol.
200	31	Continuous decomposition of glycerol and degradation of acetic acid.
250	43	Initiation of thermal decomposition of chitin.
300	55	Advanced starch and cellulose degradation.
350	72	Total degradation of chitin and cellulose.
400	85	Carbonaceous residues present (non-volatile compounds)

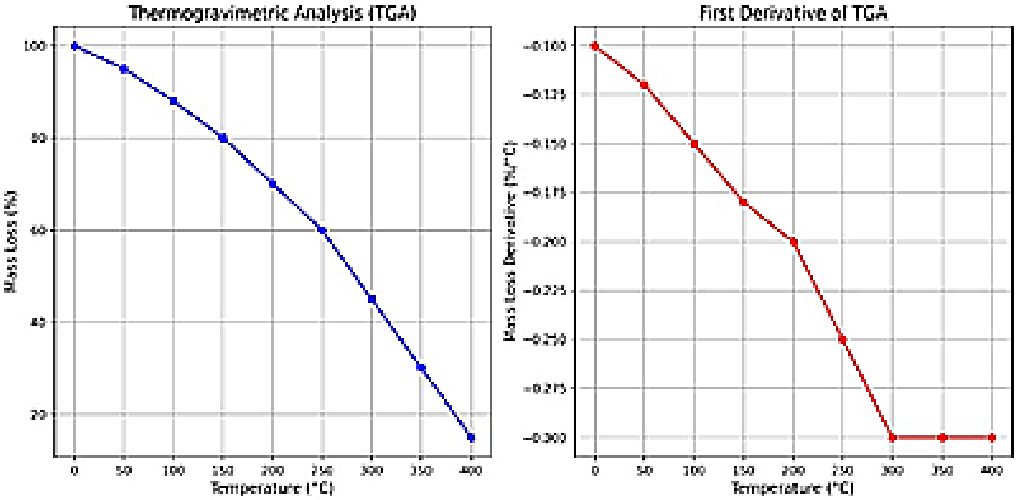

Figure 5. Thermal analysis graph.

stage, occurring within the temperature range of 50-200°C, shows an initial mass loss of approximately 5-30%, likely due to the removal of adsorbed water and volatile materials. The second stage, which spans from 200-400°C, exhibits a considerable mass loss of around 30-85%, associated with the thermal degradation of cellulose and chitin. The final stage, beginning at 400°C, indicates a residual mass loss of approximately 85-100%, which is related to the degradation of PET. Furthermore, the first derivative of the thermogravimetric analysis (TGA) reveals distinct peaks that indicate the temperatures at which the rate of decomposition is maximized: the first peak (100-200°C) corresponds to the initial decomposition of cellulose and the release of water, the second peak (300-400°C) relates to the primary decomposition of cellulose and chitin, and the third peak (400°C and above) is associated with the degradation of PET[30,44-46].

3.7 Biodegradability analysis

The composting tests conducted provided an assessment of the degradability of the bioplastic produced under controlled conditions, as detailed in Table 5. The results indicate that the degradation of the samples is predominantly influenced by the duration of exposure, with temperature exerting a moderate effect, while relative humidity appears to have a minimal role in the evaluated conditions. In the initial assessment, no degradation was recorded after one day, even at a temperature of 20.2 °C and a humidity level of 30%. In the subsequent evaluation, an increase in temperature to 24.6 °C and an exposure period of 30 days resulted in a degradation rate of 25%, despite a decrease in relative humidity to 20%. Ultimately, the third evaluation, which extended over 120 days at a temperature of 23.7 °C and a consistent humidity of 20%, resulted in complete degradation (100%). These findings suggest that the duration of exposure is the most critical factor influencing the degradation processes observed.

In Table 6 presents the findings indicating that the bioplastic underwent nearly complete degradation over a period of 120 days. This is further evidenced by a notable degradation rate of 25% observed at the 30-day mark in the second evaluation, which may also be attributed to the carbon dioxide produced during the degradation process. The study referenced in[47,48] supports this observation, noting that a similar dosage resulted in complete degradation within 100 days, thereby reinforcing the correlation between temperature and the rate of decomposition. Additionally, a decrease in humidity was noted, likely influenced by temperate climatic conditions. In the third evaluation, conducted at a temperature of 23.7°C and 20% humidity (as illustrated in Figure 6b), nearly total degradation was again achieved within 120 days. This outcome suggests that, despite the lower temperature compared to the second evaluation, the prolonged duration facilitated maximum degradation, particularly under stable temperature and humidity conditions. The graphical representation in Figure 6 further illustrates these findings.

Table 5. Results of the conditions of each evaluation.

Evaluation	Temperature (C°)	Relative Humidity (%)	Degradation (%)	Time (days)
First	20.2	30	0	1
Second	24.6	20	25	30
Third	23.7	20	100	120

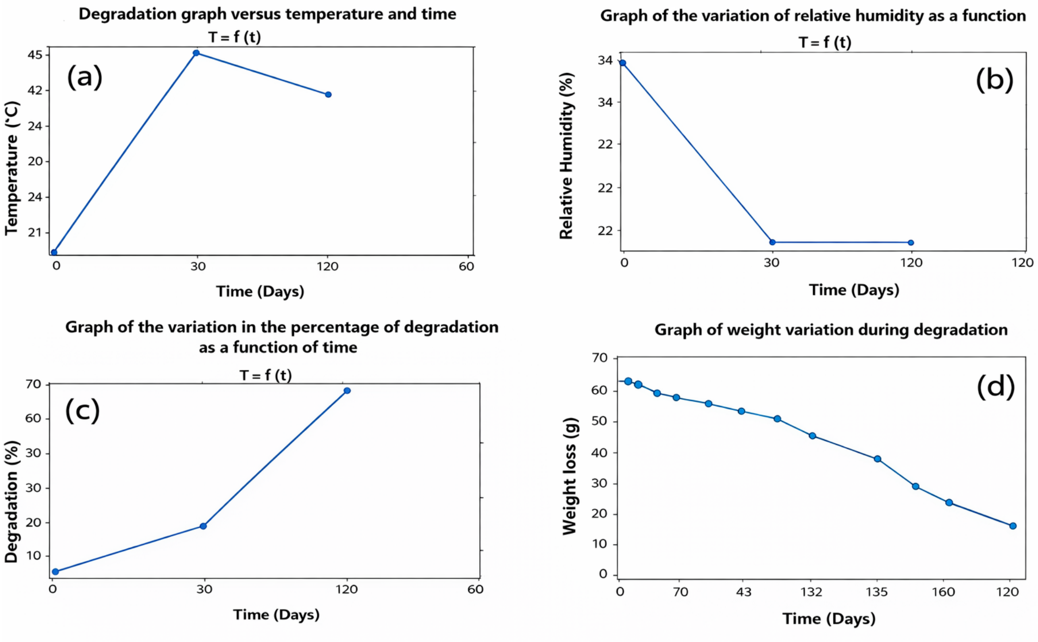

Figure 6. (a) Temperature as a function of degradation time; (b) humidity as a function of degradation time; (c) degradation in each evaluation, (d) weight loss in degradability.

Table 6. Results of weight loss in the degradability analysis.

Day	Weight (g)	Day	Weight (g)
1	70	30	61
2	70	40	58.2
3	70	50	55.6
4	69.5	60	49.2
5	69.1	80	40.3
10	65	90	30.5
15	63.2	100	24.6
20	61.5	120	15.6

The results depicted in Table 6 and Figure 6d indicate that the weight loss of the bioplastic remained relatively stable during the initial days, followed by an acceleration in degradation over time Figure 6c, culminating in almost total weight loss by the end of the 120-day period Figure 6a.

It is important to note that the biodegradability results presented in this study are based on visual disintegration and mass loss measurements under composting conditions. While these findings indicate structural breakdown of the material, they do not confirm complete molecular degradation. Future work should include advanced analyses such as gas evolution (CO_2 monitoring), spectroscopic techniques (e.g., FTIR, NMR), and microbial assays to fully characterize the biodegradation pathways and assess environmental safety of the degradation products.

4. Conclusions

The current research illustrates the potential for developing a thermoformable bioplastic from chitin sourced from shrimp exoskeletons as the principal raw material. The results underscore that this bioplastic exhibits mechanical properties that are conducive to industrial applications, with an average tensile strength recorded at 0,833 MPa and a density of 2780 Kg/m^3. Additionally, the bioplastic formulation containing 5% chitin was found to possess optimal characteristics in terms of stiffness and structural flexibility, making it the preferred candidate for prospective applications. In terms of biodegradability, the bioplastic achieved complete degradation within a span of 120 days under regulated composting conditions, thereby supporting its compatibility with circular economy frameworks and environmental sustainability initiatives. Thermogravimetric analyses indicated that the thermal degradation process of the material is influenced by the polymeric composition and the thermal stability of the constituent components, particularly chitin, starch, and glycerol.

5. Author's Contribution

- **Conceptualization** – Jorge Braulio Amaya.
- **Data curation** – Jorge Braulio Amaya.
- **Formal analysis** – Jorge Braulio Amaya.
- **Funding acquisition** – NA.
- **Investigation** – Jorge Braulio Amaya; Paola Duque Sarango.
- **Methodology** – Jorge Braulio Amaya.
- **Project administration** – Jorge Braulio Amaya.
- **Resources** – Jorge Braulio Amaya.
- **Software** – NA.
- **Supervision** – Jorge Braulio Amaya.
- **Validation – Verification**, Paola Duque Sarango.
- **Visualization** – Jorge Braulio Amaya; Paola Duque Sarango.
- **Writing – original draft** – Jorge Braulio Amaya; Paola Duque Sarango.
- **Writing – review & editing** – Jorge Braulio Amaya; Paola Duque Sarango.

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