

Development and Characterization of Edible Films from Resistant Polymers of Native Potato Starch as a Food Coating

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The production of biodegradable plastic materials using natural resources is very important due to environmental concerns. The purpose of this research was to develop and characterize a film based on native potato starch employing glycerol as a plasticizer without additives, for use as an edible coating in food preservation. An experimental method was applied with three treatments (concentrations of 2, 3 and 4 % of native starch), glycerol (2 %) and three repetitions. The results showed that the biofilms had good mechanical properties, and the higher of starch concentration the better was the structural cohesion of the films and better tensile strength. It is concluded that using pigmented native potato starch at 4 %, glycerol at 2 %, without additives, a good quality of the film was obtained with rapid degradation, resistant, economical, and non-toxic, suitable for the use of food coatings. The degradation time was at 14 d with constant degradation, and 100 % degradation was attained after 19 d. The effectiveness of described method in obtaining the films showed good results for food coatings.

1. Introduction

Starch is cheap and abundant because it is an energy storage medium found in plants. Starch-based biofilms are transparent, odorless, and tasteless, characteristics similar to those of synthetic polymers used in food packaging (Daza et al., 2018), making starch an appropriate substance for use as edible biofilms. This starch is mainly obtained from corn, wheat, potatoes, and rice (Basiak et al., 2017). The production of biodegradable plastic materials using natural resources has attracted increased attention due to environmental concerns (Kamal, 2019). Recently, the use of natural polymers such as starch to produce packaging films has received much attention as a substitute for conventional petroleum-based packaging films, as starch is biodegradable and sustainable (Zahiruddin et al., 2019).

The production of biodegradable plastic materials using natural resources has aroused increased attention due to environmental concerns, biodegradable films being an alternative to non-renewable films (Moraes et al., 2017). Several studies have reported on the use of starch as the main source in the preparation of edible food films and coatings (Gómez et al., 2023). This is due to its profitability, high biodegradation rate and abundance in nature, in addition, starch-based films are flexible and resistant (Ahsan et al., 2023). Native Andean potato starches have high starch, apparent amylose and phosphorus contents, high paste clarity and high molecular order (Martínez et al., 2019).

Plasticizers are an important class of low molecular weight non-volatile compounds that are widely used in polymer industries as additives (Ledniowska et al., 2022). In the production of biopolymer-based films and

coatings, plasticizers are also essential additives, as they can improve the flexibility and handling of the films, maintain integrity, and prevent pores and cracks in the polymer matrix (Abdullah et al., 2022). Plasticizers improve the stability and strength of polymers and are used to form effective edible coatings and films (Rai et al., 2021). Most starch-compatible plasticizers have been specified as polyols, such as mannitol and glycerol (Chavan et al., 2021).

Recent years have seen great developments in bio-based polymer packaging films for the serious environmental problems caused by non-biodegradable petroleum-based packaging materials. An edible film is a thin wrapping or packaging material used in the food industry that can be consumed as part of food (Tien et al., 2021). Increased consumer demand for higher quality food in combination with the environmental need to reduce disposable packaging waste have led to increased interest in research into edible films and coatings (Zhou et al., 2021) since they come from biodegradable, non-toxic polymers that help increase the quality of food during its conservation (Kim et al., 2023).

This study presents an innovative approach to the production of edible films using native potato starch and glycerol as plasticizers. Although the use of glycerol has been explored in other contexts, the specific combination with native potato starch and the biodegradability assessment has not been extensively reported in the literature. This work not only contributes to reducing the use of synthetic polymers but also offers a potentially scalable solution for applications in the food industry.

2. Method

2.1 Plant material

6 kg of pigmented native potato tubers *Solanum tuberosum* subsp. *andigena* were collected in high Andean potato-producing places in the Apurimac-Peru region and La Libertad-Perú region, in March 2023. Good conditions such as physical appearance and weight were considered, and spoiled, bruised, or rotting tubers were avoided. The sweet potatoes were washed, peeled, and cut into pieces; the slices were homogeneously blended in a laboratory blender. Subsequently, the suspension was sieved through a 100-mesh screen and rinsed with distilled water. An excess of 0.25 % (w/v) NaOH solution was added to the precipitate and maintained for 24 h at 4 °C. Afterwards, the top layer was removed, and the precipitate was centrifuged, then washed three times with distilled water until a neutral supernatant was obtained. Finally, the wet starch was dried in a tray dryer at 50 °C (Ghoshal and Kaur, 2023).

2.2 Obtaining the starch films

To obtain the films, an adaptation of the method described by Acquavia et al. (2021) was used. Several tests were evaluated to define the appropriate formulation at 2, 3 and 4 % of extracted potato starch designated as treatment 1, 2 and 3, respectively, mixing with distilled water (50 %) and 2 % glycerol, with pH 6. The preparation of each treatment was boiled at 80 ± 2 °C for 10 min. A quantity of 20 mL of the preparation was poured into Petri dishes, previously conditioned with Teflon tape to facilitate the removal of the film, it was left to dry at 45 °C for 12 h in an oven, and then in a desiccator to prevent it from absorbing humidity. The film was subjected to tensile strength measurements to determine flexibility and strength parameters.

2.3 Film characteristics

pH was determined by potentiometry (Consort, C5010). The thickness of the film was measured by cutting it in a rectangular shape (3 x 2 cm). The measurement was performed using a Micrometer (Digimatic Micrometer - Mitutoyo) under conditions of 21 °C and 65 % relative humidity (RH). The ability of the film to resist stretching efforts – was measured by applying a force on the film until ruptured, according to ASTM D882 method using the following Eq (1) (Song et al., 2023).

$$\text{Tensile strength (MPa)} = \frac{\text{Force necessary to break the sample (N)}}{\text{Cross sectional area (mm}^2\text{)}}$$

Starch moisture was estimated gravimetrically by weight difference after drying the sample at 110 °C for 2 h in a Memmert UN30 oven, and ashes by using a muffle at 550 °C.

Previously dry moisture-free films were used. To determine this parameter, a square of edible film (2 x 2 cm) was placed in a 250 mL Erlenmeyer flask with 80 mL of distilled water for 10 min on a stirring plate at 25°C. Subsequently, the film was recovered and placed in an oven at 60°C for 24 h, until constant weight was obtained. Ten small squares (2 x 2 cm) were cut measuring the length, width and thickness of each one with a micrometer, to determine their volume; then the mass of each frame was obtained with an analytical balance, to calculate the density of the film (Han et al., 2020).

2.4 Degradation process

To evaluate the film degradability under environmental conditions, the gravimetric method based on weight loss was used (Arrieta et al., 2013). Film samples were placed in stainless steel mesh containers, which allowed direct contact with the soil and facilitated the recovery of the material at different stages of biodegradation. This procedure was designed to ensure accurate degradation monitoring in a controlled environment. Biopolymer films were prepared at three different concentrations (2%, 3%, and 4%) with three replicates per concentration. Each replicate was placed in a steel mesh container. The steel meshes, together with the films, were buried at a depth of 8 cm in the soil, simulating natural biodegradation conditions. The meshes were removed and the samples were weighed every two days using an analytical balance with a resolution of 0.001 g. The samples were subjected to controlled composting conditions, following the guidelines of ASTM D5338.

2.5 Statistical analyses

The results were compared by ANOVA and Tukey test at 0.05 probability using the analytical software Statistix 10 (Tallahassee, FL, USA).

3. Results and discussion

In relation to the thickness of the film, the observed values were slightly above 0.180 mm without significant differences between them, as illustrated in Figure 1A. These values are compared with the 0.098 ± 0.002 mm reported by Khairunnisa et al. (2018), who used alginate and glycerol (1.1 %) based films in their study. The results were also found to be higher than the 0.035-0.085 mm range of Rusli et al. (2017) who developed biofilm based on glycerol (15 %) and carrageenan (3 %), and smaller than the 0.073-0.095 mm determined by Tanjung et al. (2020), where used films composed of starch and sorbitol, indicating that the films share a similar material base. Film thickness is an important parameter as it directly affects the mechanical and barrier properties. According to Longares et al. (2004), the addition of a higher concentration of starch not only increases the tensile strength but can also cause a slight increase in thickness, which translates into a stronger interaction between the polymers and, consequently, greater resistance to the physical actions to which films are subjected. This behaviour has been corroborated by Sartika et al. (2022), who highlighted the impact of starch concentration on film properties. On the other hand, the resistance to traction is shown in Figure 2B. It is observed that the 4 % concentration of potato starch has 4.19 MPa of resistance, unlike the concentrations of 2% and 3% of potato starch, with a significant p-value of 0.027. There are significant differences between the treatment with 2 % starch concentration treatment and the one with 4 % concentration. The tensile strength found in this investigation is lower than the 4.73 ± 0.43 MPa found by Khairunnisa et al. (2018), and the range of 8.36-15.66 MPa obtained by Hidayati et al. (2021). The result is attributed to the concentrations of potato starch with glycerol (4 % starch with 2 % glycerol).

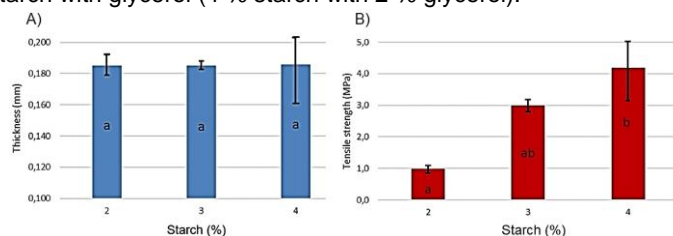


Figure 1: Thickness (A) and Tensile strength (B) of the edible films at different concentrations (%) of starch. Columns with distinct letters are different each other according to Tukey test ($p \leq 0.05$).

There were significant differences between all treatments on the solubility index of potato starch films (Figure 2A). The higher solubility in water indicates that the film shows good characteristics of permeability, and it is possible to obtain films with favourable properties to be used in the preservation of food in the food industry. Statistically, the composition of the film exhibited significant differences in terms of humidity content. The humidity content is closely associated with the concentration of glycerol, which serves as the plasticizer, and has a substantial impact on the film's mechanical and barrier properties. In Treatment 3, the highest humidity content was observed, correlating with the highest concentrations of starch (Figure 2B).

Treatment 3 shows the highest film density with an average of 0.41 g/cm^3 (Figure 2C), and exhibited other appropriate characteristics regarding moisture, solubility and thickness. The treatments in the different concentrations of starch have been repeated 3 times in each case, making a total of 9 repetitions, the highest concentration of starch (4 % in treatment 3) has shown better results for the density of the films even with smaller volumes with higher weight of the films than in treatments 1-2. The density displayed a relationship with the thickness of the film, the greater the concentration of starch, the greater the thickness of the film.

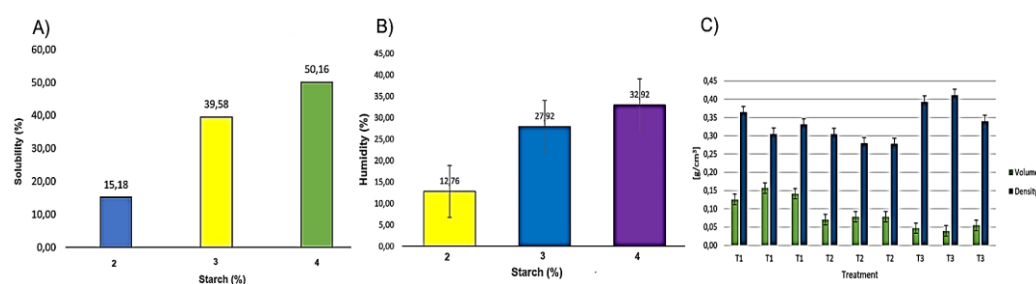


Figure 2: Determination of the Solubility (A), average Humidity (B) and Density (C) of films.

Statistically at least one of the treatments used is different from the others in terms of the density of the films of pigmented native potato starch (Table 1). The results show a proportional relationship between density and thickness in most treatments, which can be explained because of the molecular interaction of the components in the treatment (glycerol and starch). Also, the degree of cohesion seems to have an impact on the density of the film. According to Van et al. (2023) it is necessary to have an appropriate density to ensure the stability of the film, because it helps to maintain the structural integrity of the product.

Table 1: Anova test on potato starch films characteristics.

	Sum of squares	df	Square mean	F	Significance
Between groups	0.018	2	0.009	6.235	0.034
Within groups	0.009	6	0.001		
Total	0.027	8			

Table 2 shows that treatment 3 with three repetitions of 4% starch concentrations, shows better conditions of solubility, moisture, density, tensile strength, therefore demonstrating that the film obtained without additives, with 2 % glycerol, can be used in food preservation.

Treatment 3 has up to 4.20 Mpa of tensile strength which makes a film with good characteristics of be used as food coating; also, the other physicochemical characteristics studied show better barrier properties as natural sources without additives. The humidity and the ash content were within the parameters established in the Peruvian Technical Standard (Jamanca-Gonzales et al., 2022). These characteristics allow films to have an adequate structure and distribution to be more homogeneous.

The composition of the formulation of the films (T1, T2 and T3) influence their properties, the most optimal formulation being T3, demonstrating greater solubility compared to the percentage of starch and achieving the formation of uniform films, without cracks, good resistance and good physicochemical characteristics such as solubility in water, humidity, density and thickness. The result shows a direct relationship with glycerine and starch concentration, achieving good quality films.

Table 2: Physical and Mechanical Properties of film samples.

	Thickness (mm)	Density (g/cm ³)	Humidity (%)	Solubility (%)	Endurance (Mpa)
T1	0.12 ± 0.09	0.33 ± 0.03	12.76 ± 0.18	15.18 ± 0.34	0.65 ± 0.41
T2	0.10 ± 0.01	0.29 ± 0.01	27.92 ± 0.27	39.58 ± 0.53	3.00 ± 0.32
T3	0.06 ± 0.01	0.40 ± 0.06	30.47 ± 4.66	50.16 ± 1.25	4.20 ± 1.80

The degradability of the films was determined during the 19 days for total degradation (100%). After exposure time of 14 d the samples were considered only traces (Figure 3). A constant weight loss till 14 d was observed when the mean weights of the films were around 0.47 g. Following that day, the weight decreased very slowly with the degradation process well advanced. The statistical comparison between the three starch concentrations resulted in a p-value of 0.540, that is, no significant differences between the treatments.

Starch is widely used in the food industry due to its properties such as low gelatinization temperature and low tendency to retrograde (Borges et al., 2015). However, starch must be mixed with other types of substances to obtain materials with desirable characteristics. This is accomplished by adding glycerol which has shown to be an excellent plasticizer. Fatma et al. (2015) evaluated different levels of glycerol to make edible films and found that increases of glycerol decreased the tensile strength with no effect on thickness of the film. The combination of glycerol with native potato starch in the concentrations used in this research showed to be a good combination, achieving films resistant to higher concentrations of starch and lower concentrations of glycerol with adequate film moisture.

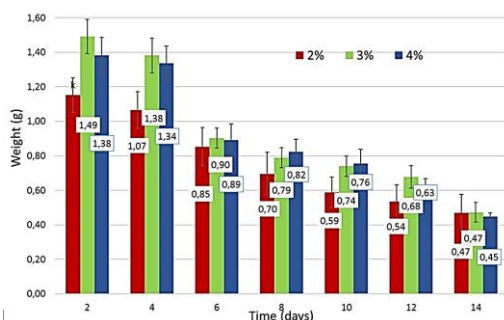


Figure 3: Film weight and degradation with time in the Petri dishes as a function of different percentages of starch.

4. Conclusions

The biofilm from potato starch showed adequate properties. The film thickness was not affected by the three different starch concentrations evaluated, while the tensile strength increased with the increase of concentrations. The use of glycerol at 2 % is considered a suitable combination as a plasticizer with pigmented native potato starch at 4 %. A homogeneous film without cracks was obtained, being a continuous material and with greater compaction, with high resistance and ease of processing considered as an alternative to the use of synthetic polymers and suitable for coatings in food preservation. The degradation of the films was fast, with a constant rate, with presence of traces at 14 d and 100 % degradation in 21 d, showing possibilities of sustainable alternatives over time compared to the accelerated use of synthetic polymers. The results represent a promising alternative for degradable edible coatings without additives in the food industry, obtaining a good combination with the glycerol plasticizer. Films made without additives should be further investigated as alternatives for food coatings.

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