Starch-based edible protective coating obtained from sweet potato waste.

Ana Real[1], Raquel Galante[1(*)], Rogério Medeiros[1], Vânia Pais[2], João Bessa[2], Sandra Cardoso[3], Fernando Cunha[1,2], Raul Fangueiro[2,4].

1 - Centro de inovação em Materiais e Produtos Avançados
2 - Associação Fibrenamics - Instituto de Inovação Em Materiais Fibrosos e Compósitos
3 - O.T.M. Verde - Produção e Comércio Agrícola, Unipessoal, Lda
4 - Departamento de Engenharia Mecânica da Universidade do Minho

There is a need for sustainable solutions to increase shelf time and ensure food safety. The use of edible protective coatings in the food industry is and simple and innovative alternative, even more, when the coating is combined with active agents that confer additional functional attributes. Natural polymers such as polysaccharides are very interesting given their edibility, biodegradability, and easy access, among other aspects. Starch is one of the most widely used polysaccharides mainly due to its low cost and simplicity of processing technologies. In this work, sweet potato wastes were used to extract starch. This biopolymer was combined to develop an edible film, by gelation process. The edible coating was characterized in terms of surface morphology (SEM), chemical composition (FTIR) and wettability. For the protective effect, different fruits (bananas, apples, passion fruit, tomato and guava) were coated with 1 or 2 layers of this film. The results showed, in general, that the coated samples revealed better results regarding, texture, color, odor and delayed maturation when compared to the uncoated controls. This is indicative of the potential for increased shelf-time of fruits without significant alterations to their intrinsic properties. Moreover, the extraction process, based on food waste recovery and circularity combined with the protective effect shows the importance of this work and its alignment with several sustainable development goal.

Keywords: Starch Based, Protective Coating, Potato Waste

1. Introduction

Food safety is an essential part of food security. Foodborne disease (FBD) can lead to the spread of infectious illnesses in many places around the globe: World Health Organization recently reported that 70 % of 600 million food poisoning cases resulted in death worldwide (Pires & Devleesschauwer, 2021).

Also, the incidence of that type of disease is very high in countries like the USA and Australia: 76 million and 5.4 million cases of food poison per year, respectively (Mullan & Wong, 2009).

FBD economic burden can also be a problem: human illness caused by food poisoning cost the United States around 2.9 and 6.7 billion dollars (Buzby, Roberts, Lin, & MacDonald, 1996).

These diseases may result from poor food safety procedures followed by farmers and producers, who may use prohibited pesticides to cultivate goods. This is undesirable since it could create not only food poisoning but also environmental damage (Nayak & Jespersen, 2022).

Wrong food safety procedures are not the only cause of environmental damage: food packaging has a major role on this cause. In general, goods are usually packed using plastic since it protects the product and improves the shelf time. (Zhang, Dhir, & Kaur, 2022). Stronger arguments are made for the urgent need to reduce or reconsider the use of plastic in the world (Azzi, Battini, Persona, & Sgarbossa, 2012; Hu, Shen, Zhang, Li, & Zeng, 2019; Shen et al., 2019; Thompson et al., 2004). This is essential to reduce pollution, enhance sustainability, and protect biodiversity (Shen et al., 2020).

Efforts have been made in order to tackle this challenge: Franz and Welle (2022) developed a new food package based on plastic waste; Molina-Besch and Keszleri (2022) explored de possibility of using biobased plastics to reduce the impacts of this material. Others investigated the potential for developing an edible packing for fruits and vegetables (Kumar et al., 2022), based on biopolymers. Some of them use starches, cellulose derivatives, chitosan, animal or plant-based proteins, and lipids. These biocompatible polymers can help extend the shelf life

of fruits preventing also food poisoning (Trajkovska Petkoska, Daniloski, D'Cunha, Naumovski, & Broach, 2021).

Among those different biopolymers, starch (which is found in seeds, roots, and tubers (Bello Perez & Agama-Acevedo, 2017)) has received considerable attention since it is low cost, abundant, edible, and biodegradable (Abral et al., 2019; Davoodi, Kavoosi, & Shakeri, 2017).

This prompt researchers to investigate food waste as a source of starch for the development of innovative food packaging.

Sweet potato (Ipomoea batatas L (Lam)) starch has excellent gas permeability, good film-forming and mechanical tolerance (Oyom et al., 2022) making it adequate to use in food packaging. Also, millions of sweet potato (> 90 million) (Guo et al., 2022) are produced worldwide, generating a considerable amount of waste that does not comply with the food industry requirements Thus, it an be re-used to develop new products such as edible packaging.

In this study, an edible food packaging solution is developed, considering circular economy concept (Zhu, Liu, Ye, & Batista, 2022).

A starch-based edible coating, produced from sweet potato waste, was developed and its protective effect on different fruits was tested. This approach intends to reduce waste through the valorization of food excedents not suitable for human consumption, while developing sustainable alternatives for food safety.

2. Materials and Methods
2.1. Materials

The purple sweet potato was provided by the OTM Verde, an agriculture company from Azores, Portugal; ethanol (96%) analytical grade supplied by Labkit (Azores, Portugal); citric acid from Sigma (Spain); neutral gelatin powder supplied by Panifor (Azores, Portugal).

2.2. Methods
2.3. Extraction of starch from purple sweet potato waste

The purple sweet potato tubers were washed and cleaned carefully with tap water to remove the soil spots. The cleaned tubers were peeled and cut into small pieces. Next, the pieces were transferred to a food processer containing distilled water (1:2 v/v). The mixture was grinded until a pulp was obtained. Thereafter, the pulp was filtered using a cotton cloth and the solid matter was discarded. The filtrated was let to settle allowing for the sedimentation of starch. The wet starch was then kept at -30 °C for 4 h and freeze-dried for 21h (Scanvac Cool Safe). Finally,

the dried starch was grinded into a fine powder, and stored in sealed containers for later use.

2.4. Characterization of extracted starch

The extracted starch was analyzed in reference to its nutritional components, namely: moisture, protein, sugar, and ash content. For that, the starch was kept at 100 °C for 24 hours. The protein content was quantified using the Kjelhahl method, while the amount of carbohydrates was determined using the Luff-Schoorl method. (Senanayake, Ranaweera, Gunaratne, & Bamunuarachchi, 2013).

2.5. Development of starch films

The starch was dissolved in distilled water to form a uniform solution. The solution was subject to heating at 75ºC for 5 min, under constant agitation, and cooled in ambient temperature. After that, neutral gelatin powder was added (0.42/0.30/99.30(m/m/m) (starch/ gelatin/ water).

The mixture was stirred until complete dissolution of the gelatin powder.

2.6. Characterization of starch films
2.6.1. Scanning electron microscopy (SEM)

The surface morphology of coated surfaces were analyzed using NOVA 200 Nano SEM equipment from FEI Company. Before observation, the surfaces were vacuum coated with a thin layer of gold to make them electrically conductive.

2.6.2. Fourier transform infrared spectroscopy (FTIR)

A Shimadzu IRAffinity-1S equipment. was used to record the scanning spectra in the wavenumber range of 4000 and 400 cm-1, operating at attenuated total reflection mode.

2.6.3. Wettability analysis

The wettability was measured through the contact angle, using a Goinometer (DataPhysics Instruments, model OCA 15plus; Germany). The experiments were performed according to the standard ASTM D7334-08 (Standard practice for surface wettability of coatings, substrates, and pigments by advancing contact angle measurement). Briefly: the sessile drop method was applied. The procedure consisted in dispensing a 3 µL drop of water at different zones of each sample. At least ten measurements were carried out ($n = 10$).

2.6. 4. Protective Effect

To assess the protective effect of the liquid film, several fruits were coated with it.

Initially, the fruits were sanitized with a cotton cloth. Secondly, the coating was applied by dipping the fruits into the coating solution for 30 s. The excess solution was left to drip off. When appropriated, a second layer was applied after a 10 minute interval, following the same procedure. Then the samples were air-dried for 12h at room temperature to remove any superficial moisture.

The protective effect was tested in five different fruits: guava, banana, passion fruit, apple and tomato. Each experiment was done in triplicate (n=3). The protective effect was evaluated visually and by analyzing the odor.

3. Results and Discussion
3.1. Starch extraction yield
The nutritional results, for the extracted starch, are presented in **Error! Reference source not found.**

TABLE 1. NUTRITIONAL ASSAY OF EXTRACTED STARCH.

Assay / Method Physical-Chemical Parameters	Units	Freshly harvested
Moisture content	g/100g	4,0
Protein Kjeldahl Method	g/100g	0,9
Carbohydrates Measurement	g/100g	92,3
Sugars (mono- and disaccharides) Luff-Schoorl method	g/100g	<0,1
Fibers *	g/100g	2,1
Lipids *	g/100g	0,3
Total Ash*	g/100g	0,4

*Parameter determined by gravimetric method

The starch content was 92.3%, an acceptable value, especially when considering that there
was no chemical purification process involved.

3.1.1. Effect of potato aging on the yield of the extraction process
In order to determine the effect of aging on the obtained percentage of starch, the same extraction process was applied to freshly harvest potatoes and compared to matured potatoes (1 month after harvest).

The percentage of starch extracted from freshly harvest potatoes, $47 \pm 2\%$, was calculated using Equation 1:

Equation 1: % of starch =

Where is the mass of dry starch (g) and is the mass of dry sweet potato pulp (g).

The same was done for the starch extracted from the matured harvests samples, $27.4 \pm 3\%$.

According to Suraji A. Senanayake, et al., the percentage of starch present in sweet potato can vary between 33% and 65% of dry content (Senanayake et al., 2013) . Considering the middle point as the expected theoretical starch percentage of sweet potato, the yield, η, can
be determined by, equation 2:

Equation 2: η=

Where the amount of starch obtained corresponds to the percentage of dry starch and the Theoretical amounts of starch, correspond to the middle value between the interval referred in the literature.

Thus, the starch yield for freshly harvest extraction was ~96% whereas, the mature harvest only led to ~56%.

The results indicate that there is a decrease in the yield of the starch extraction process as the tuber ages. This decrease is related to the fact that the potato, being deprived of photosynthesis via its branches, resorts to the consumption of the starch stored in its granules in order to survive and be able to germinate again, which is in agreement with other published work (Yang et al., 2022) .

3.2. SEM
The surface of the coatings with both one and two layers had a homogeneous appearance, without pores or cracks, Figure 1 A and B. Figure 1 C and D show the cross-sections of the liquid coatings with one and two layers, respectively. Compact structures can be seen, which are indicative of their structural integrity. In the case of the two-layer coating, granules of insoluble particles can be observed.

Regarding the thickness of the coatings, Figure 1 C and D, with 1 layer there was an average thickness of 2.62 ± 0.5 µm, and with 2 layers an average thickness of 3.76 ± 0.6 µm.

3.3. FTIR
Figure 2 shows the absorption spectra in the infrared region for: the control coverslip (glass), the 1-layer sample and the 2-layer sample. As expected, there is a band related to the presence of water on the film ($3700\text{-}3000\,cm^{-1}$), which is not observed on the control glass coverslip (red). It can also be observed that this band is more pronounced for the two layer coating (green), which is as expected.

Additionally, and as expected, no significant differences were observed between the main peaks of the spectra of the different layered coatings. Nevertheless, the obtain results are in accordance with other starch-based composition found in literature().

3.4. Wettability analysis
The contact angle between the coating (one and two layers) and the water droplet is shown in Table 1.

TABLE 1: CONTACT ANGLE RESULTS

	Control	1 layer	2 layers
Angle (°)	22±5	80±2	82±5

The results show that the glass coverslips coated with the starch film, either with one or two layers, have a higher contact angle than the control coverslip (without coating). It is possible to state that the film coating based on sweet potato starch is hydrophilic (< 90º). However, the value of the contact angle is very close to the hydrophobic/hydrophilic boundary, which may justify the difficulty in adhesion of the coating to some specific surfaces.

3.5. Protective Effects
3.5. 1. Banana
Figure 3, shows the protective effect study on bananas. It can be seen that the starch coating, in general, delayed ripening process. The control samples revealed aging signs after 7 days. For the coated samples the process was delayed until 8-12th days.

3.5. 2. Guava
Regarding the guava, Figure 4 shows the evolution of the test. After 3 days of testing, a change in coloration was observed in the control samples, while in the coated sample, significant alterations were only visible after the 5th day.

The control samples showed at day 5 a considerably less firm texture than the coated samples. Regarding the inside of the fruit, the control sample showed a more yellowish color when compared to the coated. Furthermore, the odor of the control samples revealed a slightly fermentation scent, while the conted samples maintained their characteristic odor.

3.5.3. Passion Fruit
Figure 5 shows the evolution of the test for the passion fruit.

After 5 days, the one layred coated samples presented a wrinkled texture, and the same was not observed for both the control and the two layers samples. On the 10th day of the test, all samples seemed the same.

Regarding the interior of the fruit, it was observed that coated samples, either with one or two layers, showed a more orange color compared to the control samples. In addition, all samples presented the characteristic smell of ripe fruit.

3.5. 4. Apple
Figure 6 presents the test results for apples. It was observed that the coated samples remain the same until the 22 day, when a slight change in coloration was observed. The uncoated samples change appearance by day 16. After 22 days of testing the control samples showed a wrinkled surface when compared to the coated ones. This is indicative of the protective effect regarding the loss of water. The inside of the control apples, showed higher oxidation when compared to the coated samples.

3.5. 5. Tomato
Figure 7 shows the evolution of the test for tomatoes. By the 4th day of the test there was a change in the color of the tomatoes in general. However, this color change was more evident in the control samples, indicating a protective effect of the coatings. Regarding texture, all samples showed firm texture until the 8th day of the test. Nevertheless, by the 10th day, the samples coated with two layers developed mold.

4. Conclusion
In this work, starch was extracted from sweet potato waste. A protective starch-based coating was developed using the extracted biopolymer. The protective effect on several fruits was evaluated. The obtained results allowed the conclusion of pertinent points, such as:

→ In bananas, guavas and passion fruits, the samples coated with either one or two layers of coating at the end of the trial showed better texture and characteristic odor of the fruit, while the control samples showed some considerable fermentation;

→ In apples, samples coated with either one or two layers of the liquid coating showed better results both in color and texture and in the oxidative process inside the fruit;

→ In tomatoes, the coated samples showed better results compared to the control, in terms of delayed maturation. However, the presence of the starch-based coating led to the formation of mold.

In is important to note that there is no formulation that meets all conditions or that can be universally applied to all fruits. Each fruit presents its own physiology, such as respiration rate, maturation and senescence, characterizing it as unique from the point of view of the elaboration of a suitable coating. In addition, since starch is a natural polymer there is a greater predisposition, when used without a preservative, to the development of molds.

5. Acknowledgements

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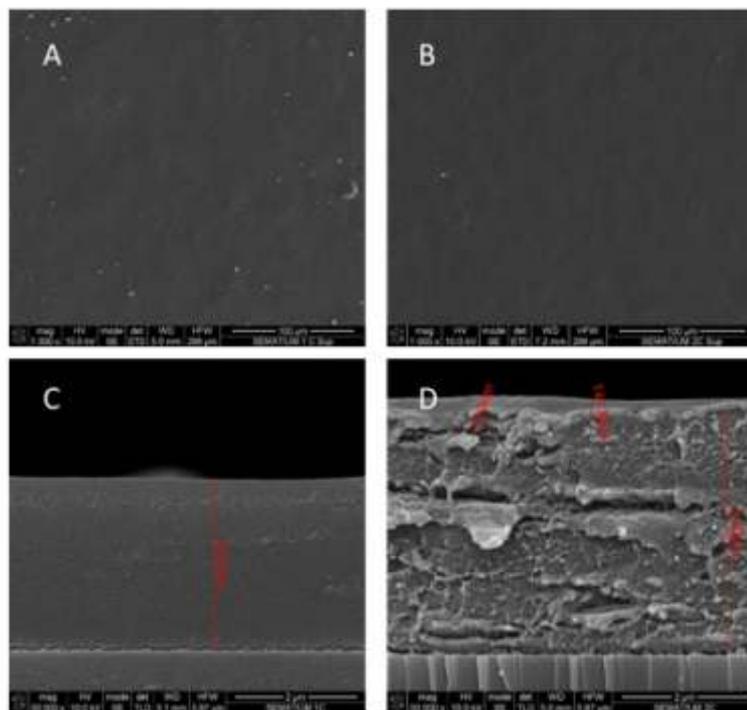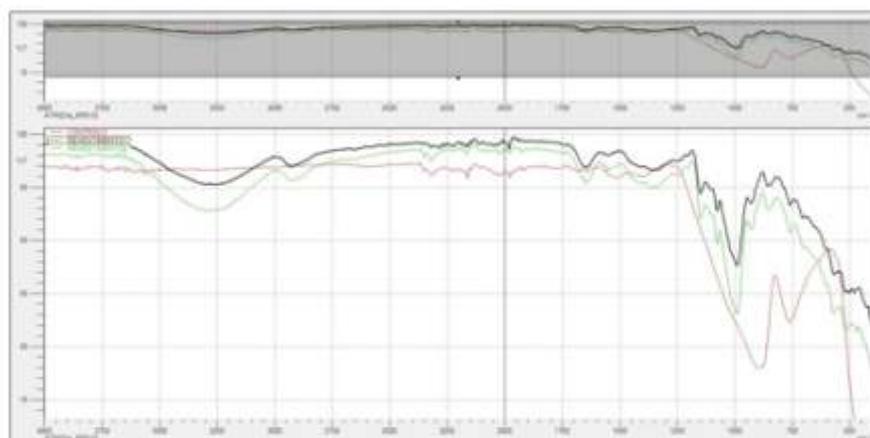
Figure 1: Sem Micrographs of The Starch Film: **A** - Surface of Liquid Coatings One Layer; **B** - Surface of Liquid Coatings Two Layers; **C**- Cross Section of Liquid Coatings One Layer; **D** - Cross Section of Liquid Coatings Two Layers.
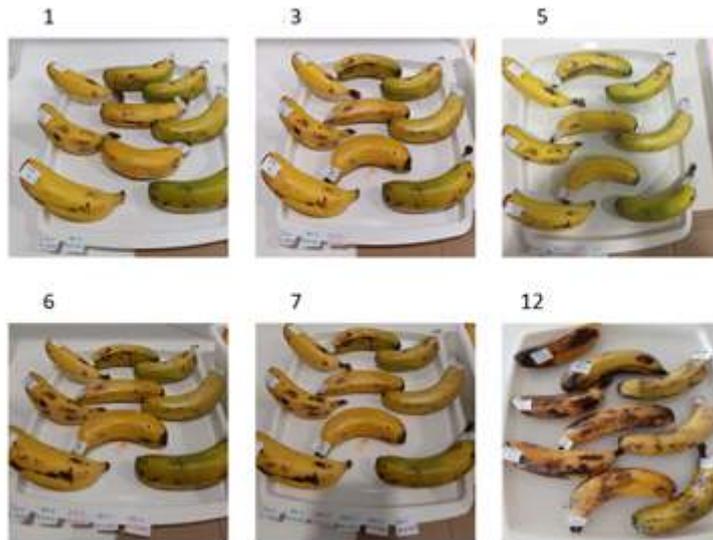
Figure 3: Study of The Protective Effect (starch Coating) on Bananas During 8 Days of Testing. for each Image, Consider: Column 1 - control; Column 2-1 Layer Coating; Column 3-2 Layer Coating.
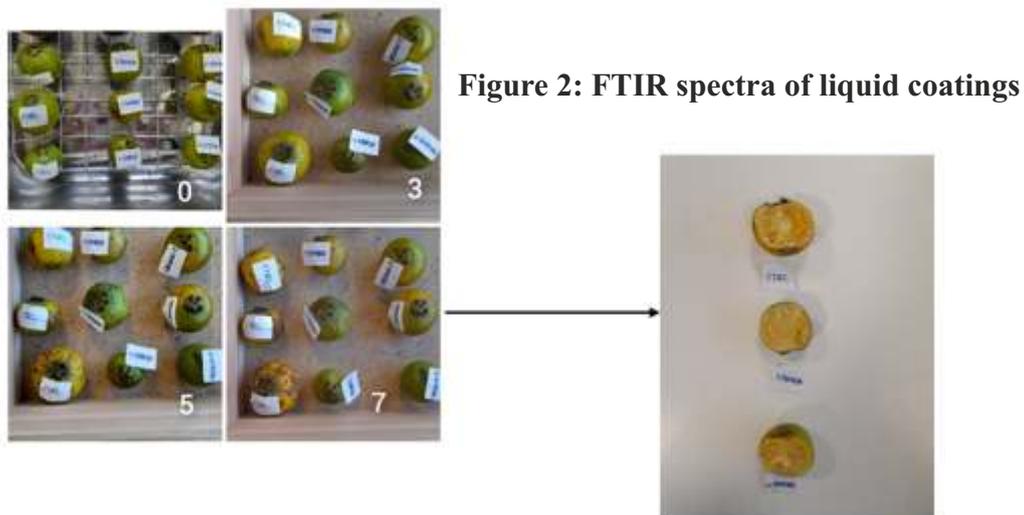

Figure 2: FTIR spectra of liquid coatings

Figure 4: Study of The Protective Effect (starch Coating) on Guavas After 3, 5 and 7 Days of Testing, On the **Left**. for each Image, Consider: Column 1 - control; Column 2- 1 Layer Coating; Column 3- 2 Layer Coating. On The **Right** Side It can be seen The Interior of Bananas after 7 Days of Testing (control, 1 and 2 Layers Starch Coating).

Figure 5: Study of The Protective Effect (liquid Starch Coating) on passion fruit after 5, 10 And 12 Days of Test on The **Left**. for each Image, Consider: Column 1 - control; Column 2- 1 Layer Coating; Column 3- 2 Layer Coating. On The **Right** Side it can be seen The Interior of Passion Fruit After 13 Days of Test (control, 1 and 2 Layers of Coating).

Figure 6: Study of The Protective Effect (starch Coating) on Apples After 5, 8, 16 And 22 Days of Testing on The **Left**. on The **Right** Side it can be seen The Interior of Apples after 23 Days of Testing (control, 1 and 2 Layers). for each image, Consider: Column 1 - control; Column 2- 1 Layer Coating; Column 3- 2 Layer Coating.

Figure 7: Study of The Protective Effect (starch Coating) on Tomatoes (optimal Picking Maturity) After 1, 4, 6, 8, 9, 10, 11, 14 Days of Testing on The **Left**. for each Image, Consider: Column 1-control; Column 2- 1 Layer Coating; Column 3- 2 Layer Coating. Molds After 14 Days of Testing on The **Right**.