# EVALUATION OF PHYTOTOXICITY AND BIODEGRADATION OF CELLULOSE REINFORCED STARCH BIOCOMPOSITES

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With increasing environmental pollution, sustainable alternatives for packaging materials are important. This study aimed to evaluate the phytotoxicity and biodegradation of corn starch films reinforced with cellulose extracted from pineapple crowns. The films were produced by casting mixtures of corn starch, glycerol and cellulose at different concentrations (5%, 10% and 15%), along with a pure starch control film. The phytotoxicity assay showed that the addition of films to the soil does not negatively influence the germination and growth of lettuce seeds (*Lactuca sativa*). In the biodegradation assay, the films were completely degraded after 60 days, especially the film containing 15% cellulose, which experienced rapid biodegradation, with a high rate of initial mass loss. Analysis of the Fourier transform infrared (FTIR) spectra after degradation of the films showed no structural changes. These materials have no harmful effects on the environment and have great potential for application as rapid-use packaging.

*Keywords*: eco-friendly, fibers, films, packaging

## INTRODUCTION

Today, we are part of a rapidly growing and over-consuming population, where there is no balance between accelerated progress and the preservation of the resources needed to live. Therefore, the conscious use of resources, reasonable consumption of materials and the prioritization of sustainable solutions must serve as guiding lines in all development areas.[1]

Synthetic polymers are among the main environmental contaminants. When improperly discarded, they significantly pollute nature, as these products are designed to have excellent durability and longevity, thus, they take centuries to decompose. Also, there is an urgent need to reduce the dependence on landfills for waste management and the amount of plastic discarded, which ends up in the streets and oceans. It is estimated today that approximately 380 million tons of plastics are produced annually worldwide, with approximately 8 million tons of plastics going into the oceans, and by 2050, the quantity of discarded plastics will exceed that of marine fish.[2]

In the present-day scenario, it is no longer enough to remediate environmental problems: prevention of this type of pollution is needed.[3] Therefore, biodegradable polymers are an interesting alternative to the use of materials from non-renewable resources. This is especially true in the development of food packaging, since traditional packaging is prone to contamination with organic particles when in contact with food, making it difficult or impracticable to recycle these materials.[4]

For a polymer to be considered biodegradable, it must meet several basic criteria. First, the material must disintegrate and be metabolized by microorganisms in a natural environment within a given period of time. It is also necessary that the material does not release toxic components into the environment at the end of its decomposition. The process of biodegradation of the material is also known for some modifications in the

characteristics of the polymer, such as a decrease in molecular weight, mechanical properties and changes in the visual aspect of its surface.[5] The simulated soil test is one of the mechanisms to assess the biodegradability of a given polymer. In this process, the material is exposed to an organic compound, under specific temperature and humidity conditions that favor the growth of microorganisms.[6]

Given this, it is remarkable that biodegradable materials can be applied in single-use packaging, such as food packaging, garbage bags, among others. In recent years, several research works have been carried out to improve the general properties of biofilms, resulting in composite bioplastics. Among the biopolymers most frequently used for biocomposites are starch and cellulose, which have interesting properties for this type of application, such as rapid decomposition in the soil and the ability to be combined with other materials to achieve the desired properties. These plant-based biopolymers have shown high potential for developing biodegradable products, with no biological hazard risks and with low cost, being a sustainable alternative to petrochemical plastics.[7]

Analyzing the mechanical properties and characteristics of these biopolymers is essential to evaluate the potential of these new materials to present a valid alternative to petrochemical plastics. The present manuscript is part of a larger research that developed biodegradable films based on corn starch, incorporating cellulose obtained from pineapple crowns, and evaluated their barrier, mechanical and morphological properties.[8] In a previously reported study, we demonstrated that the film with higher fiber concentration had a lower water vapor permeability rate, and the biocomposites were less soluble in water, compared to the pure starch film. In terms of mechanical properties, it was observed that the Young's modulus of the biocomposites

increased in relation to that of the starch film, without affecting the tensile strength.[8]

In this part of the research, we evaluated the phytotoxicity and biodegradability of the corn starch films reinforced with pineapple crown cellulose as an alternative form of single-use packaging to reduce the impact caused by improper disposal of packaging in the environment. The importance of this study is given by the low number of published articles illustrating the visual aspects of the degradation steps of starch films, in addition to assessing the time taken for total degradation of the films. Moreover, the findings point out the sustainability and environmental friendliness of the proposed packaging materials, verifying that their disposal will not cause damage to plants and the environment.

## EXPERIMENTAL
### Materials
Maize starch was obtained from Bom Gosto, Brazil. Glycerol was purchased from Dinâmica, a commercial soil substrate was obtained from Carolina Soil II Classe V CE 0.7, for phytotoxicity tests, and cellulose was previously obtained from pineapple crowns.

### Methods
#### Preparation of composite films
The films were made using the casting method, where a solution of 5 g of starch and 1 g of glycerol dispersed in distilled water was prepared and stirred at 75 °C to gelatinize the starch.[9] Different concentrations of cellulose, previously obtained from pineapple crowns, were added to make different films. In this manner, films reinforced with 5% cellulose (CF5%), 10% cellulose (CF10%) and 15% cellulose (CF15%), as well as a pure starch film (SF), were prepared, as summarized in Table 1.

To form the films, 18 g of the solution was placed on kiln-dried acrylic plates at 35 °C for 24 h. After drying, the films were removed from the mold and packed.[9]

Table 1
Formulation of films

| Samples | Cellulose (g) | Glycerol (g) | Starch (g) | Water (g) |
|---------|---------------|--------------|------------|-----------|
| SF | 0 | 1 | 5 | 94 |
| CF5% | 0.25 | 1 | 5 | 93.75 |
| CF10% | 0.50 | 1 | 5 | 93.50 |
| CF15% | 0.75 | 1 | 5 | 93.25 |

#### Simulated soil preparation
The biodegradation test was conducted in soil according to ASTM G 160-03. The soil was prepared

from a mixture of equal parts of beach sand, horse manure and fertile soil, and left to mature for three months. The test setup was formed of containers, into

which the prepared soil and 5 cm × 5 cm samples of the films were added. All the films had their initial weight determined (day 0). The films were then packed in the containers and removed after 15, 30, 45 and 60 days. Each container held five samples of each formulation. The setup were placed in a laboratory oven kept at 30 °C, and each film was cleaned and weighed when it was removed.[8]

## Characterization
### Phytotoxicity

The phytotoxicity of the films was evaluated to investigate the production of toxic compounds during degradation, which might affect the growth of plants. This experiment was conducted using lettuce seeds (*Lactuca sativa*) as a biological model, widely used in studies with this purpose.[7] The experiment was carried out in styrofoam trays, where the films (1 g) were crushed and mixed into the commercial soil substrate. Three replicates of three lettuce seeds were planted for each film formulation, in addition to a control cell without added material.[10]

Plant growth was monitored for 20 days, and then the plant with the largest size of each cell was chosen for the analysis of fresh and dry mass to investigate the influence of the addition of the film on the substrate of the plants.[10]

### Biodegradability test - mass loss

The evaluation of the biodegradability of the films was conducted in simulated soil, according to ASTM G 160-03. The biodegradability was measured by the mass loss of the films, determined after 0, 15, 30 and 45 days.[11] The mass loss was calculated using Equation (1), where $W_i$ corresponds to the initial weight, and $W_f$ refers to the final weight:

$$Mass\ Loss\ (\%) = \frac{(W_i - W_f)}{(W_i)} \cdot 100 \tag{1}$$

### Visual inspection and optical microscopy (OM)

The macroscopic effect of biodegradation was determined by visual inspection of the films through photographs taken at the same intervals mentioned above. The microscopic effect was analysed through optical microscopy analysis with 4x magnification (Laborana model 06-KTESD5000).[12]

### Fourier transform infrared spectroscopy (FTIR)

The structural characterization of the functional groups of the films before and after biodegradation was performed by Fourier transform infrared (FTIR) spectroscopy using a Shimadzu Prestige-21 spectrometer. A number of 16 scans were recorded between 400–4000 cm$^{-1}$.

### Statistical analysis

To assess whether there was a difference in seedling development through dry and fresh mass of the plants among the soils with different concentrations of biofilms, the analysis of variance (ANOVA) was performed. If necessary, the results were subsequently evaluated by Tukey's test at 5% of significance by the program PAST v. 3.20.

## RESULTS AND DISCUSSION
### Phytotoxicity

There are several unanswered questions about biodegradable polymers and their effects on the environment. Despite this, there are few studies on the toxicity of materials when discarded in the natural environment. Figure 1 shows lettuce seedlings planted in soil containing samples of the films after 20 days from sowing the seeds. All the seeds germinated; therefore, germination and survival was 100%, in other words, the addition of the films to the soil did not influence germination.

The growth of the lettuce seedlings was also evaluated, in the presence and in the absence of films, by determining the average fresh and dry weights of the seedlings after 20 days. After statistical analysis, there was no significant difference in seedling growth evaluated through the fresh and dry mass of the seedlings (fresh mass, p = 0.736; dry mass, p = 0.655) grown in the presence of different films buried in the soil. The data are presented in Table 2.

Thus, the addition of films to the soil does not hinder seed germination or the development of lettuce seedlings. Similar results were found when testing the ecotoxicity of PLA/starch and PLA/PBAT/starch blends, respectively, indicating that these materials did not affect the ecosystem adversely.[10,13] In the research on xylan/starch biofilms, ecotoxicity data resulted in 100% seed germination. Also, the degradation of the bioplastics had a good influence on root and hypocotyl growth, which thus proved to be non-ecotoxic biodegradable materials.[14]

The starch present in the biofilms can contribute to the growth of lettuce seedlings, being an important source of nutrients, since it is a fundamental polysaccharide in the development of plants. Thus, at the end of its degradation, it results in a rich content of carbon, returning to the soil with beneficial effects. From this point of view, the evaluation period of 20 days chosen in the present study may have been too short, as the films did not reach the end of the decomposition process and the formation of the biomass.[15,16]

Considering the findings, these materials are of interest as an alternative not only for food packaging, but also for other applications, such as

in bags for seedlings, because they can be planted in the soil without the need to remove the packaging, offering convenience in large plantations, greater security for plant roots and favoring their development through the availability of carbohydrates for plants.[17]


Figure 1: Lettuce seedlings 20 days following planting

Table 2
Fresh and dry weights of lettuce seedlings

| Samples | Fresh weight (g) | Dry weight (g) |
|---|---|---|
| Control | $0.11^a$ | $0.017^a$ |
| SF | $0.12^a$ | $0.017^a$ |
| CF5% | $0.13^a$ | $0.020^a$ |
| CF10% | $0.11^a$ | $0.017^a$ |
| CF15% | $0.16^a$ | $0.030^a$ |

*Values followed by the same letter indicate non-significant differences at 95% confidence level (p < 0.05)

**Biodegradability test - mass loss**

Mass loss is a very common method used to determine the changes caused by microbial attacks in biodegradable polymers. For this, an average of five samples for each formulation was weighed on days 15, 30, 45 and 60 of soil burial, and the percentage of mass loss was calculated after each assay (Table 3).

In the first 15 days of contact of the samples with the soil, there were significant mass losses in all the sample groups – of around 50% of the mass of the specimens. The losses were higher in the composite films than in the SF, and the higher the percentage of cellulose, the greater the mass loss.

On days 30 and 45 after soil burial, the degradation percentage becomes more uniform with film type; however, the composite with the highest percentage of cellulose still degraded more quickly. This relationship can be explained by the detachment of the fibers, which grant the entry of water and microorganisms into the matrix, causing a higher degradation rate. After 60 days, no more residues were found, indicating that all the films were completely degraded. Other researchers observed similar behavior when they performed soil testing of starch films with similar fibers, with total degradation in this period of time.[18,19]

Mass loss occurs because of degradation, and this process happens continuously at different decomposition stages: biodeterioration, biofragmentation, assimilation and mineralization. Biodeterioration occurs on the material's surface, determining the modification of its chemical and physical characteristics, with the production of extracellular substances that help break down the polymer. The second step is biofragmentation, breaking the macromolecule into smaller portions through enzymatic cleavage. Assimilation is about the consumption of food made in the cytoplasm through the formation of biomass, energy and proper growth. The last stage is mineralization, in which biodegradable materials are transformed into gases, water and minerals. Compounds such as carbon dioxide ($CO_2$), methane ($CH_4$) and water ($H_2O$) are generated. Mineralization occurs when only full-film

components are manufactured or all transformed.[20]

In a study using loads of 15 or 20% of samaúma fibers, the mass loss was 7 to 10 times greater than that of pure PHBV films after 45 days of biodegradation in the soil.[19] In another research, a similar behaviour was observed for the developed corn starch films with corn husk fiber reinforcement. After eight days of burial in the soil, the control film had a loss of 47.1% in weight, while the composite with the highest reinforcement content (8% fiber) had a greater weight loss, corresponding to 73.2%, suggesting that this occurs because, in the presence of humidity and temperature, the microorganisms show a preference for attacking the fibers.[18]

Another study has shown that the biodegradability of bioplastics is linked to the nature of the starch used, for example, films based on cassava starch degrade faster than those based on corn starch. Thus, the crystalline structure of corn starch is a monoclinic system with eight water molecules per cell. Therefore, the moisture content of corn starch is lower than that of cassava starch, hence, its lower rates of degradation. Furthermore, the presence of moisture promotes the biodegradability of bioplastics, especially composite bioplastics, as demonstrated in a study that used a pectic polymer extracted from *Cola cordifolia*, which has the ability to intercalate into the free volume in the starch grains and is more susceptible to the presence of moisture. Therefore, it can be predicted that the addition of reinforcements accelerates and favors the biodegradation of films based on biopolymers.[21]

Table 3
Percentage mass loss of the films during degradation in the soil

| Samples | Day 15 | Day 30 | Day 45 | Day 60 |
|---------|--------|--------|--------|--------|
| SF      | 33.9%  | 84.1%  | 93.9%  | 100%   |
| CF5%    | 49.4%  | 84.9%  | 94.4%  | 100%   |
| CF10%   | 52.1%  | 84.6%  | 94.8%  | 100%   |
| CF15%   | 66.7%  | 87.7%  | 96.1%  | 100%   |

**Visual aspect and optical microscopy**

The changes in the pure and composite films subjected to the biodegradation tests were observed on days 0, 15, 30 and 45, as shown in Figure 2.

Visual changes on the surface of the films, such as holes, cracks, disintegration and color, are assumed to be the first signs of fungal and bacterial activity on films.[22] In the first 15 days, the differences are more visible because the SF had slightly clearer regions with a lower biodegradation rate. For the films with added cellulose, it was no longer possible to observe regions free of microorganisms at this time. With a longer burial duration, the films became more fragile, and they ruptured, in addition to experiencing noticeable changes of color. Corroborating the mass loss analysis, there were greater indications that the degradation was more accelerated in the cellulose reinforced films.[23]

The film containing 15% cellulose presents greater crack extensions, agreeing with the higher percentage of initial weight loss, compared to the other films discussed previously. Thus, the addition of cellulose as reinforcement can accelerate the biodegradability of starch films due to the high hydrophilicity of the cellulose fibers, allowing greater aggregation of soil particles and probably greater microbial activity. In addition, a study with PVA and vegetal fibers indicated that the highest percentage of degradation was observed in the composite films, with the increase in the amount of fibers; it is also possible that the matrix cannot envelop all the reinforcement, which makes the film more susceptible to the detachment of the fibers and therefore gaps in the structure, favoring the attack of microorganisms.[22,23]

An experiment on the biodegradation of various starches has presented results similar to those described above for corn starch. The films showed major changes in their color and integrity, indicating the beginning of biodegradation after 20 days of burial, and almost complete degradation after 56 days.[11]

Figure 3 shows the optical microscopy of the films after 45 days of biodegradation in the soil. The images show fractures on the surface and the decreasing size of the films, caused by the weakening of the material after contact with the soil, microorganisms, and moisture.[24] The color change of the films is also an indication of

degradation: the darker and yellowish tones indicate the presence of bacteria and fungi. These microbes are responsible for the undesirable colors in the polymer.[25] The presence of hyphae is also noted, evidencing the fungal activity resulting from the humidity and greater roughness of the film after the beginning of the test.[26]
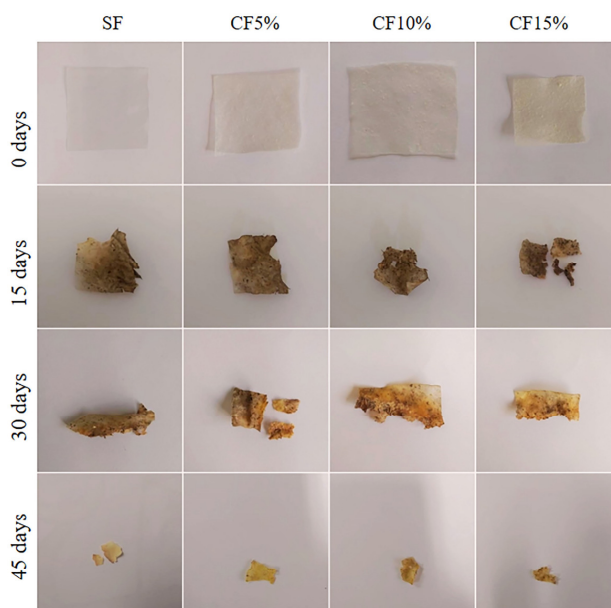


Figure 2: Visual appearance of samples during the biodegradation test
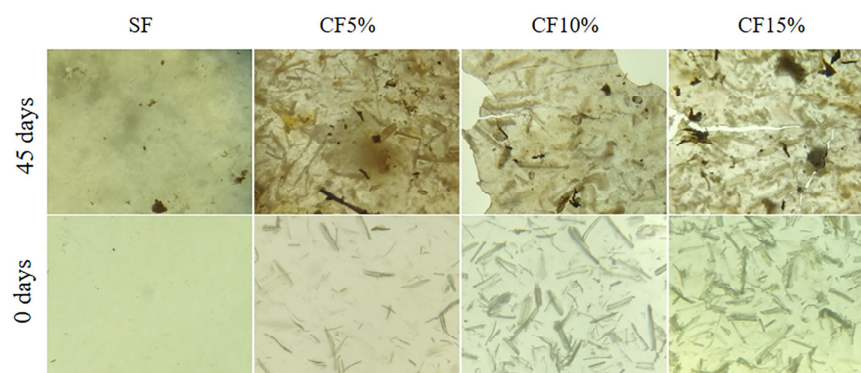


Figure 3: Microscopic images of films before (0 days) and after degradation (45 days)

Changes in surface morphology were also observed in a study on PLA/TPS films by analyzing SEM images. The authors observed, over the weeks, greater surface roughness of the film, growth of microorganisms and, after 30 days, a sudden reduction in thickness, the film showing rupture and disintegration.[26]

**Fourier transform infrared (FTIR) spectroscopy**

Figure 4 shows the spectra of all the samples before (0 days) and after the degradation test (15, 30 and 45 days). All the spectra are similar, and the bands often found for starch films have been identified.

The band analysis was performed in four regions: the first between 990 and 800 cm$^{-1}$, the glucose C–O bands, which are related to the pyranose reaction, and the bands found around 1078 cm$^{-1}$ and 1001 cm$^{-1}$, which are associated with the deformation of the C–OH groups. The second region of interest is between 1500 cm$^{-1}$ and 2800 cm$^{-1}$, where the peak at 1665 cm$^{-1}$ is in accordance with the presence of water in the starch, as it is assigned to the bending mode of the -OH group. The third region analyzed was from

2800 cm$^{-1}$ to 3000 cm$^{-1}$, which included the vibrational stretching of C–H. The last region is above 3000 cm$^{-1}$, where a high intensity peak is present due to the stretching vibrations of the O-H group, indicating that the films are hydrophilic.[18] The characteristic peak comprised at 2900-3000 cm$^{-1}$ (=C-H elongation) is due to the presence of starch. The presence of the peak between 3600 and 3200 cm$^{-1}$ can be attributed to the fact that glycerol contains polyols consisting of many hydroxyl groups.[27] The findings of the FTIR analysis are in agreement with those reported in earlier studies on starch films reinforced cellulose-based fillers, specifically, the bands corresponding to O-H functional groups found around 3000 cm$^{-1}$ and those of C-O glycosidic bonds at 1080 cm$^{-1}$.[27]
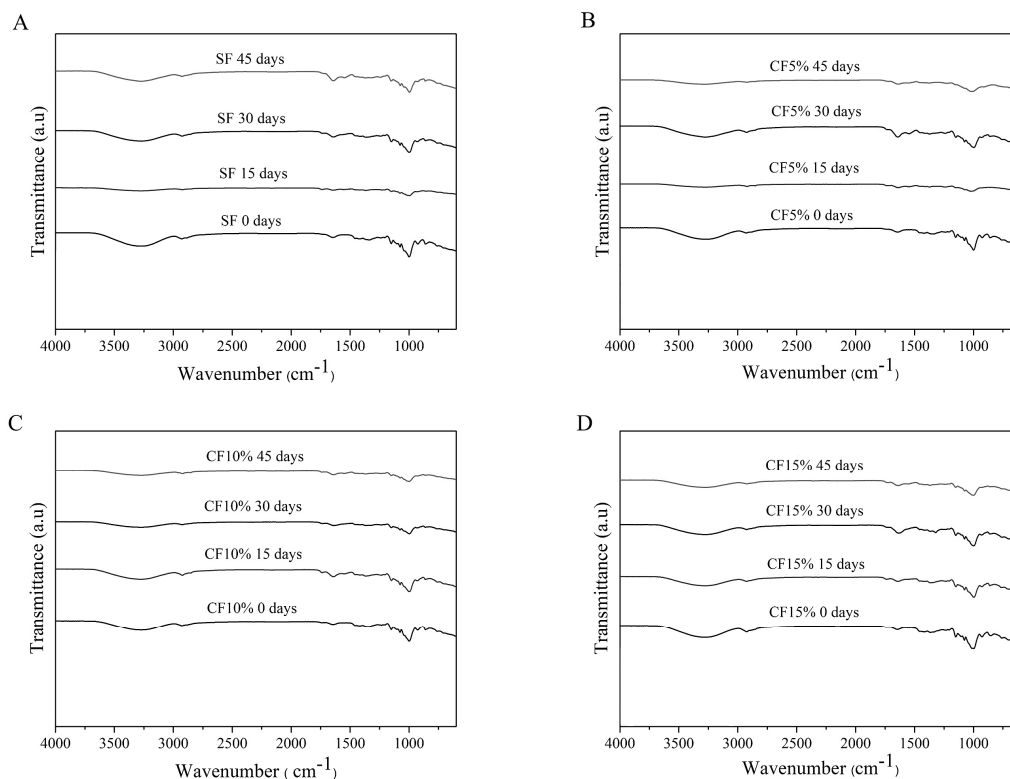


Figure 4: FTIR spectra before and after degradation of a) pure starch, B) biocomposite with 5% cellulose, C) biocomposite with 10% cellulose, D) biocomposite with 15% cellulose

There were no changes in the spectra caused by the degradation since the spectral peaks exist independently of the biodegradation of these materials. Other authors also pointed out that no new bands emerged in the FTIR spectra of starch in similar works, only some changes in the intensity of the bands.[28,29]

**CONCLUSION**

The present study demonstrated through the phytotoxicity assay that the addition of films to the soil does not negatively influence seed germination or the development of lettuce seedlings. It may even be beneficial for plant growth, as the films represent sources of starch and carbon that are made available to plants. The biodegradability study revealed that the initial biodegradation rate is higher in biocomposites, and at the end of the test (60 days), no residues were found, indicating that all the films were completely degraded. FTIR analysis performed during the degradation of the materials indicated the characteristic bands of starch films and showed no changes in the spectra as the materials degraded.

Films containing fibers derived from vegetal waste have great potential for application as food-covering films, enabling the growth of new markets and eco-friendly products, through the valorization of wastes and without harmful effects on humans and the environment.

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