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ORIGINAL ARTICLE

Nanocellulose reinforced starch biocomposite films via tape-casting technique

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Abstract

The objective of this study was to characterize the physicochemical-mechanical properties of corn and cassava starch films reinforced with CNF via Tape-Casting. There were differences in size and shape of the starch granules. Corn starch nanocomposites (NCO) showed a significant increase in tensile strength (5.14 to 25.58 MPa) and significant decrease in strain (24.81 to 2.76%) as the CNF concentration increased. Among the cassava starch nanocomposites (NCA), only the cassava starch sample with 1% CNF (NCA-1) showed significant difference both in the maximum stress (4.94 MPa) and strain (15.17%). The corn starch sample with 2% of CNF (NCO-2) presented a lower roughness and NCA-1 a smooth surface. There was no difference in chemical composition between the samples. The CNF-free starch films showed more transparency than other films. The NCA showed more transparency than NCO. Tape-casting technique unveils enhanced mechanical properties of cellulose nanofiber-reinforced starch films. Starch nanocomposites exhibit improved tensile strength and surface characteristics.

Keywords: *nanocomposites, biopolymers, starch films, nanocellulose, tape-casting.*

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1. Introduction

Petroleum-based polymers are commonly used in packaging products due to their characteristics, such as malleability, low cost, and chemical and mechanical properties. However, due to the difficulty of separating and reusing its resins used in their composition, these materials have limited recycling and degradation between 100-450 years in the environment, contributing to environmental pollution[1-4].

The search for more sustainable packaging has increased, aiming the environmental preservation and conscious consumption. A range of biopolymers derived from biomass such as polysaccharides, proteins and lipids have been used as polymeric matrices for obtaining biodegradable packaging, as well as the development of films from renewable sources. However, the major challenge for the industry is to make these films capable of replacing conventional packaging, thus having adequate and specific stability, mechanical and barrier properties for each application[5-13].

The starch has shown itself as one of the raw materials more suitable to thermoplastic films production, because of its high biodegradability potential[3,14-16]. Structurally, starch is composed of two different fractions, amylose, and amylopectin[17]. Native corn starch has a proportion of 25-28% amylose, while cassava has approximately 17% in its variations. Although starch films have properties of transparency, non-toxicity, and low cost, they still have some mechanical limitations, such as low elasticity and high permeability[17-20].

Several investigations have been carried out to improve the mechanical properties through the incorporation of other components such as natural fibers, nanofibers (CNF) and cellulose nanocrystals, oils, proteins, nanoparticles, among others[17,21]. CNF enhances bio-composites with high rigidity, low density, biodegradability, hydrophilicity, and affinity with natural polymers. The properties of biofilms depend on interactions and preparation techniques[14,21,22].

The Tape-Casting technique used in the field of flat and thin ceramics, mainly in the electronics industry in the production of blades, membranes, load cells for power generation, heat exchangers, among others[23-26]. This technique is not widespread for the production of biofilms yet, but it is an alternative for obtaining films with lower thicknesses compared to the conventional methods adopted, such as the traditional casting technique, extrusion and immersion coating[14,27]. The objective of this study was to produce and characterize films with different sources of starch reinforced with CNF through the Tape-Casting method.

2. Materials and Methods

2.1 Materials

The Cassava starch used to prepare the films was the sour powder by *Qualitá* brand and the Corn starch used was by *Yoki* brand (both are brazilian brands). The CNF suspension was produced by mechanical shear using a Grinder Masuko Supermasscolloider the concentration of 4g of nanofibers for 96g of demineralized water and glycerol (Sigma-Aldrich). The tool used in the production of the films was *Tape-Casting* with 2.5 mm of the blade opening (Figure 1).

2.2 Preparation of films and nanocomposites

The films were prepared using the Tape-Casting method. To prepare the filmogenic solution, corn starch (10%) and cassava starch (10%) were dissolved in 200 ml of water. This solution was initially weighed and heated using a heating plate and kept under constant stirring until it reached gelatinization (80ºC). Then, the solution was removed from the heating plate, 2% glycerol was added and the solution was completed with water until it reached its initial weight. For each type of starch, three samples were prepared containing the CNF suspension of 0 (standard), 1, and 2% concerning the amount of water in the solution and then deposited the filmogenic solution under the plastic substrate. The films were dried at room temperature and stored in desiccators containing silica gel for approximately three weeks, and then characterized. The formulations used were chosen through preliminary tests and are described in Table 1.

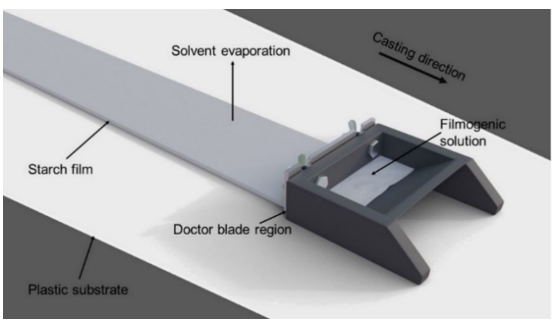

Figure 1. Tape-Casting used for the deposition of filmogenic solutions under the plastic substrate.

2.3 Subjective analysis

The subjective analysis evaluates the appearance of the film through visual and tactile observations. The samples evaluated as homogeneous and continuous appearance were considered for the other analyses. In the other hand, the defective samples, that is, the samples which presented fissures and a lot of bubbles, were not considered for the other analyses.

2.4 Thickness and density

For density determination, film samples with dimensions of 2x2 cm were obtained. These samples were kept in a desiccator with silica gel for approximately 5 weeks and then weighed on the analytical balance[28]. The thickness measurements using the Fowler Pro-max digital micrometer with 0.01mm resolution.

2.5 Mechanical properties

The mechanical tests were performed according to the ASTM-D882–09[29] standard, using the EMIC traction machine, DL line, 200KN load cell. The specimens were initially cut to the dimensions of 24x150 mm and thickness measurements were obtained in 5 random positions using a Fowler Pro-Max digital micrometer with 0.01mm resolution, with a useful length during the test of 100 mm, with an advance of 9 mm/min.

2.6 Scanning Electron Microscopy (SEM)

This analysis was used to observe the fracture surface of the films after the mechanical tests and also for the characterization of both types of starch. The equipment used was the SEM 3000™ Hitachi model with an acceleration of 15kV and an increase of 600x, 1000x, 3000x, and 7000x. To perform the analysis, the samples were cut into small dimensions of approximately 2 x 4 mm.

2.7 Fourier Transform Infrared Spectroscopy by Attenuated Total Reflection (FTIR-ATR)

It was performed with the Parkin Elmer Spectrum 400 FT-IR spectrometer Model Spectrum 400FT Mid-IR with scanning from 4000 to 600cm^{-1} and 32 scans. The samples were cut into small strips approximately 0.5 x 2 cm and performed in triplicates.

2.8 Ultraviolet-visible absorption spectroscopy (UV-Vis) for determination of of transparency

The Shimadzu UV-VIS-NIR 3600 Plus spectrophotometer was used to measure the degree of transparency of the films.

Table 1. Formulations used for the production of CNF-reinforced starch films.

Samples	Water (g)	Corn Starch (g)	Cassava Starch (g)	Nanofibers Suspension (g)*	Cellulose Nanofibers (g)	Glycerol (g)
f-CO	200	20	-	-	-	4
NCO-1	130	20	-	50	2.083	4
NCO-2	90	20	-	100	4.16	4
f-CA	200	-	20	-	-	4
NCA-1	130	-	20	50	2.083	4
NCA-2	90	-	20	100	4.16	4

*The suspension concentration is 4g CNF to 96g water.

As only the degree of transparency was the purpose of the analysis, the samples were analyzed in transmittance modes in the visible region of the spectrum, that is, from 400 to 700 nm with a baseline using air as a standard.

2.9 X-ray diffraction

The crystallinity indexes of the corn and cassava starch powder samples submitted to CuKα radiation, 30mA, 40kV, were evaluated at a speed of 2Θ = 1°/min in the range of 3-40°. The crystallinity index (Xc) was calculated by the ratio between the area of the absorption peaks and the total diffractogram area and expressed in percentage (%) using the Origin software (version 9.0, Microcal Inc., Northampton, MA, USA).

2.10 Statistical analysis

A Shapiro-Wilk normality test was performed on the database, followed by the analysis of variance (ANOVA) and Tukey test of multiple comparisons with a 5% significance level. The software used for data processing and statistical analysis was PAST software 3.26.

3. Results and Discussion

3.1 Structure analysis of starch granules

The morphology of the starch granules can be seen in Figure 2. It is possible to observe that either corn and cassava starch have a smooth surface in their granules.

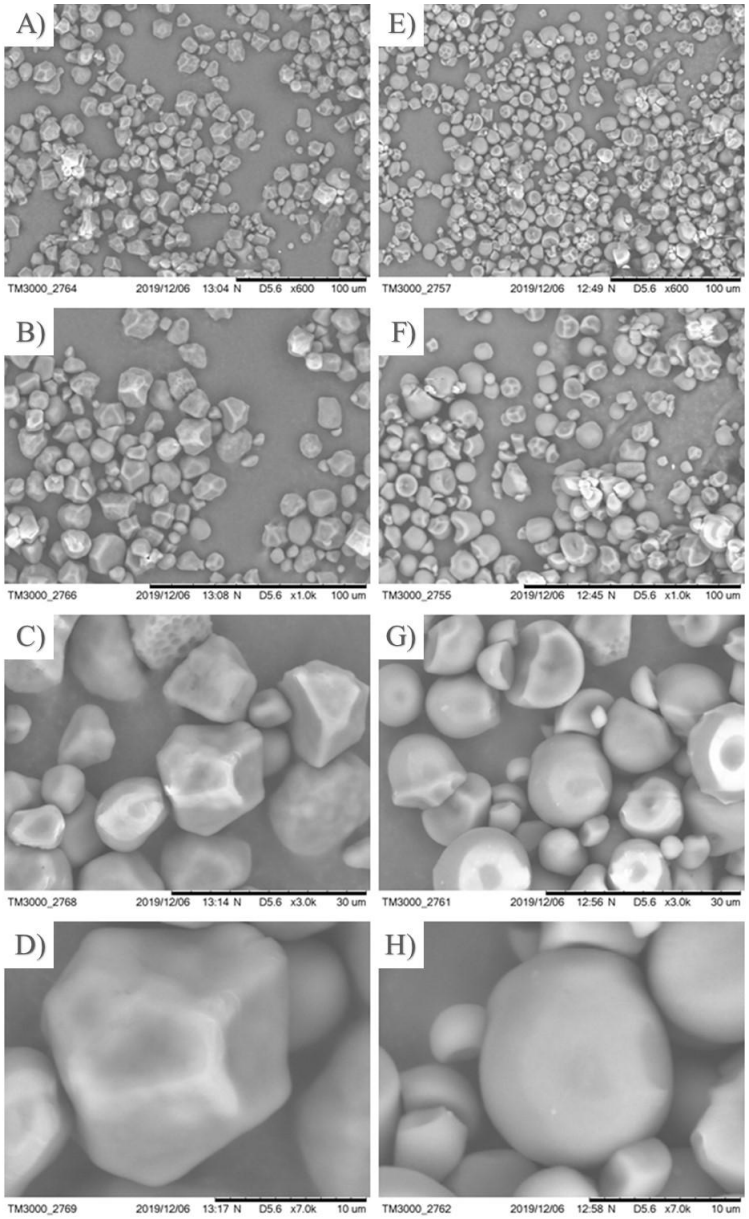

Figure 2. Corn starch granules: A) 600x; B) 1000x; C) 3000x and D) 7000x; And Cassava starch granules: E) 600 x; F) 1000x; G) 3000x and H) 7000x.

Regarding the granules' morphology, it is observed that corn starch showed a high proportion of angular and some rounded granules. On the other hand, cassava starch has a higher proportion of granules in shape of spheres and some angular shapes.

In Figure 3, the distribution of the sizes of corn starch and cassava granules can be observed and measured by SEM. Most of corn starch granules have a size of approximately 10 to 20 µm (Figure 3), values similar to those found in the literature. According to Penfield and Campbell[30], corn starch granules have a diameter of approximately 5-25 µm. The diameter values of cassava starch granules observed showed that most granules have a diameter ranging from 10 to 15 µm approximately (Figure 3). The size and shape of cassava starch granules vary according to species, plant development stage, harvest time, tuber shape, among other factors[31]. Furthermore, the literature reports that cassava starch granules have diameters ranging from 3 to 32 µm [32]; average diameter 15 to 20 µm and 15 to 23 µm[33].

3.2 Analysis of starch films

Some observations regarding the filmogenic solutions prepared with the different starches were: the cassava starch filmogenic solutions were more viscous when compared to those of corn starch. This can be explained by the higher proportion of amylopectin contained in cassava starch, and due to its greater molar mass compared to the molar mass of amylose, as viscosity increases with the rise of molar mass. Such viscosity hampered the deposition process of cassava starch films and nanocomposites on plastic substrate, leading to the appearance of many bubbles during the process.

Another explanation for the formation of air bubbles in the cassava biofilms sample is that the cassava starch used was the sour powder, that is, modified cassava starch, which has the capacity to expand due to its production process, which makes the *casting* process more difficult.

Another observation is that when rolling the films already dried in the plastic substrate, for storage, some particularities were noticed in the malleability and rigidity of the samples. Corn starch films appeared to be more malleable, and as CNF adds, nanocomposites appeared to become more rigid. Meanwhile, the cassava starch films showed a greater rigidity, and as the CNF was added, they appeared to become more malleable. These characteristics were later tested in mechanical tests.

Corn starch films appeared to be very homogeneous and opaquer, and for the most part, free from defects such as blisters and cracks. Thus, only the samples free of defects were selected to further analysis.

On the other hand, the cassava starch films appeared to be more transparent than the corn starch films, however, because the filmogenic solution was more viscous and the difficulty in preparation, it caused the appearance of air bubbles (Figure 4).

Therefore, only the bubble-free regions or regions with the least possible defect were selected to perform the tests.

3.3 Thickness and density

The thickness and density values of the films are shown in Table 2. Analyzing the thickness values, it can be observed that the Tape-Casting technique and the equipment were satisfactory in controlling their thickness. Although the differences between the values are not significant, it can be observed that the films without the nano-reinforcement presented higher density values, when compared to the nanocomposites. This behavior can be explained due to the low-density of the CNF. This behavior was also observed by Almeida et al.[34]. The thickness values obtained were lower than those obtained in other studies, such as the films incorporated with propolis extract by Araújo[35] and lower than those reinforced with pupunha palm nanocellulose obtained by Martins[36] and also lower than the starch films reinforced with CNF obtained by Marques et al.[27] and Fazeli et al.[22].

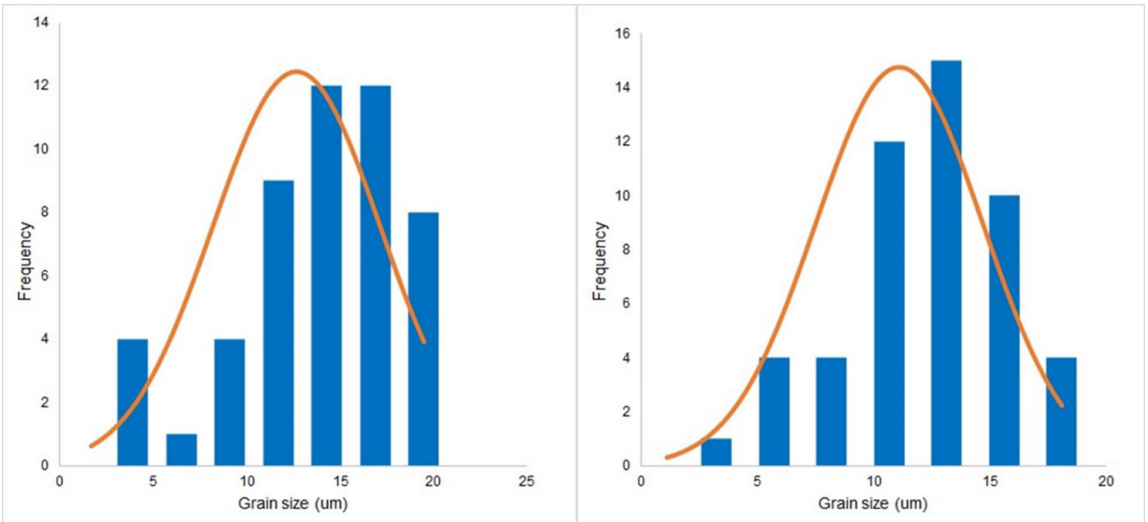

Figure 3. Size distribution of corn starch granules (left) and cassava starch granules (right).

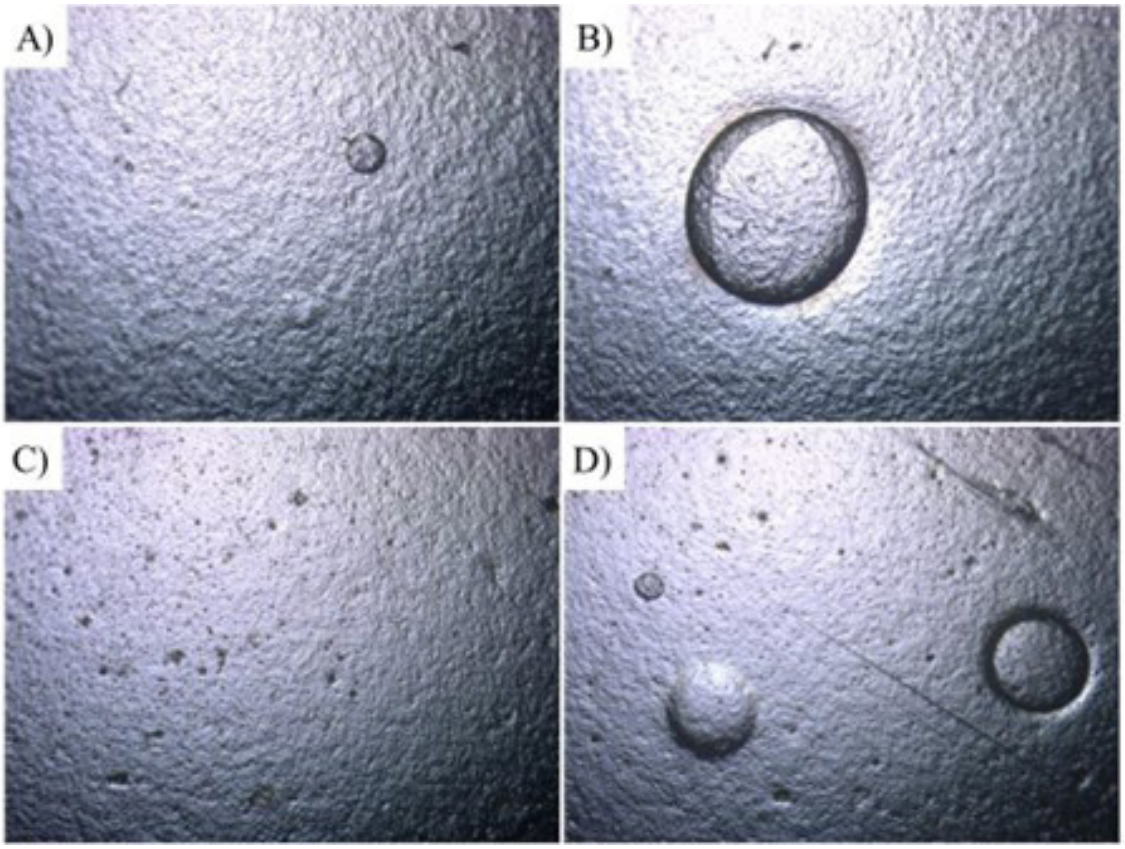

Figure 4. Samples observed in the Binocular Biological Microscope. Corn starch films: A) 5x increase; B) 10x increase; Cassava starch films: C) 5x increase; D) 10x increase.

Table 2. Thickness and density and mechanical properties (mean ± standard deviation) of the samples produced.

Formulation	Thickness (μm)	Density (g/mm³)	Tensile test		
			T (MPa)	E (%)	Y (MPa)
f-CO	85.5 ± 4.1 [a]	1.52 ± 0.03 [a]	5.14 ± 1.73 [a]	24.81 ± 4.54 [a]	307.9 ± 187.8 [a]
NCO-1	81.1 ± 6.0 [a]	1.12 ± 0.09 [a]	13.68 ± 1.73 [b]	5.97 ± 2.29 [b]	860.4 ± 73.92 [b]
NCO-2	92.2 ± 7.4 [a]	1.25 ± 0.06 [a]	25.58 ± 3.05 [c]	2.76 ± 0.21 [b]	1728 ± 181.9 [c]
f-CA	71.1 ± 4.2 [a]	1.58 ± 0.14 [a]	16.19 ± 1.61 [b]	2.63 ± 0.81 [b]	1065 ± 155.8 [b]
NCA-1	86.6 ± 6.4 [a]	1.43 ± 0.19 [a]	4.94 ± 0.35 [a]	15.17 ± 3.27 [c]	309.3 ± 74.95 [a]
NCA-2	94.4 ± 6.4 [a]	1.33 ± 0.10 [a]	10.64 ± 0.6 [b]	2.40 ± 0.31 [b]	807.6 ± 22.59 [b]

T = Maximum tensile strength or Stress; E = Elongation at rupture or Strain; Y = Young's modulus. a,b,c = Means in the same column with different letters differ significantly (p 0.05) by Tukey's test.

All these studies used the Casting technique for the production of biofilms, which reinforces the efficiency of the method for obtaining ultrafine biofilms.

A study by Moraes et al.[37] used the Tape-Casting technique to produce starch films, the author noticed that the suspension viscosity as well as the opening of the blade used in the process has a great influence on the final thickness of the films. The results showed that in the formulation of 3g/100g of suspension with and without the addition of nanofibers and with an opening of 3 and 4mm of the blade they presented a thickness between 0.07mm and 0.106mm (70μm to 106μm), which shows that the thickness of the nanocomposites obtained in this study was similar for different formulations.

3.4 Mechanical properties

The mechanical tests results (Table 2 and Figure 5) showed there were significant differences in the maximum tensile strength between f-CO (5.14 ± 1.73 MPa) and NCA-1 (13.68 ± 1.73 MPa) and NCO-2 (25.58 ± 3.05). It was observed that the tensile strength of the corn starch films increases proportionally to the concentration of CNF.

The rupture elongation values (E%) of corn starch nanocomposites (f-CO, NCO-1, NCO-2) showed the opposite behavior, as the CNF concentration increased, the resistance increases, and its deformity decreases, making nanocomposites more rigid with the addition of the nano-reinforcement.

The same behavior is expected for cassava starch-based nanocomposites. However, the samples did not show this behavior. Thus, there were no significant differences (P≥05) in the maximum tensile strength between the f-CA (16.19 ± 1.61 MPa) and NCA-2 (10.64 ± 0.6 MPa), and consequently, the elongation values of rupture showed a significant opposite behavior (P≥0.05) between f-CA (2.63 ± 0.81) and NCA-2 (2.40 ± 0.31), which implies that the resistance and flexibility of both materials have an inverse correlation. The same behavior was observed by Cerqueira et al.[1].

The results obtained from the cassava starch nanocomposites proved to be different from the results obtained by other authors such as Cerqueira et al.[1], Silva et al.[23], Martins[36] and Marques et al.[27] in studies on cassava starch films reinforced with different sources of nanofibers with similar formulations, but with lower nano-reinforcement concentrations than those tested in this study.

Marques et al.[27] used the formulation of 4.5g of cassava starch, 1.5g of glycerol, and evaluated the following concentrations of nanofibers: 0.10g; 0.30g; and 0.50g. The author observed with such concentrations the maximum tensile strength increased with the increase of the CNF concentration and consequently, the elongation had the opposite behavior. Thus, the concentrations adopted for evaluation of the present study may have saturated the nanocomposites, leading to stabilization and decrease in their maximum tensile strength.

The decrease in tensile strength observed in cassava starch films reinforced with nanofibers is a complex phenomenon that can be attributed to several factors. Undesirable interactions between nanofibers and starch, along with the influence of film morphology, are two plausible hypotheses to explain this occurrence. Inherent challenges in the preparation process, such as increased viscosity and bubble formation, may have contributed to the reduction in mechanical properties of cassava starch films, particularly with the addition of cellulose nanofibers, resulting in a less cohesive structure and lower mechanical resistance.

The NCA-1 nanocomposite showed a significant difference (P≤0.05) when compared with the other samples of cassava starch films (f-CA and NCA-2). Such behavior may have been caused by the presence of small air bubbles that cause tension points on the material's surface, reducing the tensile strength (16.19 ± 1.61) and increasing its rupture elongation (15.17 ± 3, 27) (Figure 5).

Comparing all samples, it was observed that f-CA was as resistant as NCO-1 and NCA-2, which shows the potentiality of cassava starch even without the addition of nano-reinforcement. Thus, lower concentrations than those tested in this study should be tested in cassava starch-based nanocomposites, since the adopted concentrations appeared to have saturated the material.

Among all samples, NCO-2 presented a higher tensile strength, which demonstrates the potentiality of corn starch to be used in the production of this type of nanocomposite. Accordingly, the mechanical test demonstrated that corn starch performed better in relation to its mechanical properties when compared to cassava starch.

3.5 Surface analysis of nanocomposite fractures

SEM images were performed to evaluate the fracture surface of the cross-section of the samples (Figure 6). It can be seen that there are no starch granules in the films, as seen in Figure 2.A and 2.E, which presents corn and cassava starch granules with the same magnification (600x) as the SEM image of the films (Figure 6). This means that all the starch was completely gelatinized during the film formation process, and that fractures are characteristic of brittle materials with little deformation due to the fibrous zone and the presence of voids[38].

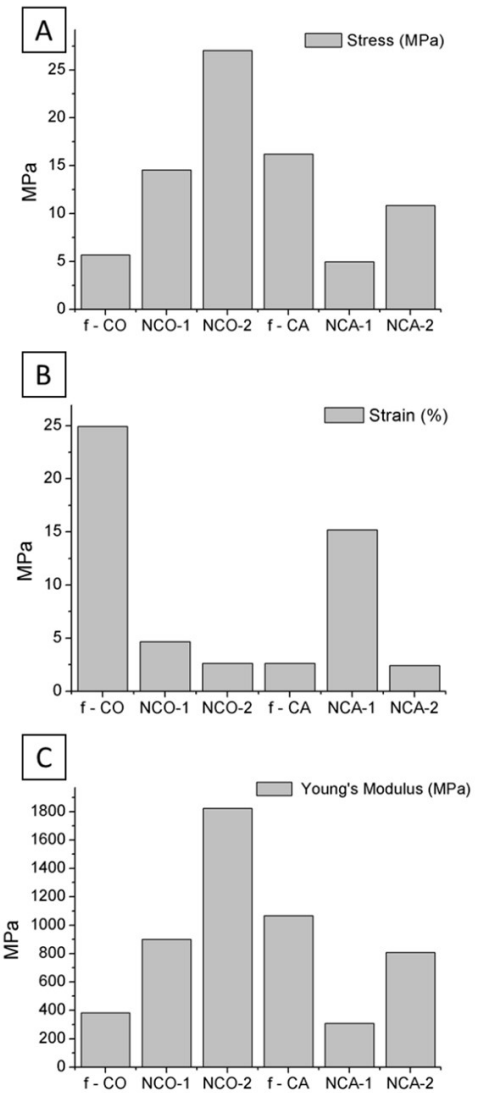

Figure 5. Mechanical properties of films with different CNF concentrations. A) Maximum Tensile Strength or Stress (MPa); B) Elongation at Rupture or Strain (%); C) Young's Modulus (MPa).

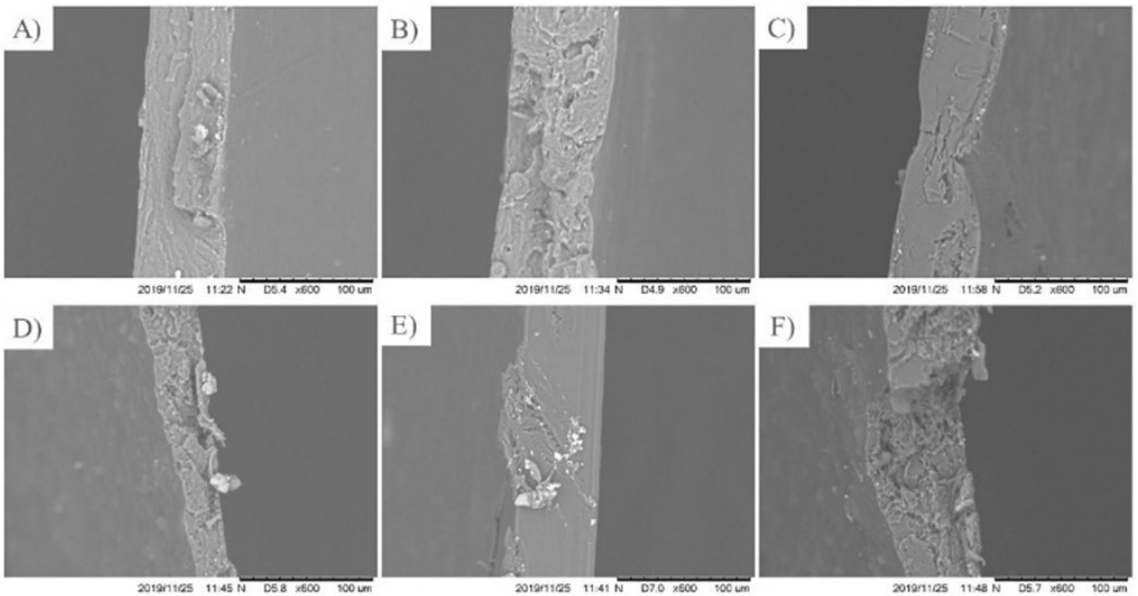

Figure 6. Edges of films after the tensile test (600x increase). A) f-CO; B) NCO-1; C) NCO-2; D) f-CA; E) NCA-1 e F) NCA-2.

Regarding to the corn starch samples, the (f-CO) surface is smoother than its nanocomposites (Figure 6A) . This is because there are fewer obstacles in f-CO films, which facilitates the propagation of fissures. With the addition of CNF, the surface of nanocomposites has become rougher and rugged (Figure 6B e 6C). The presence of nanofibers prevented the spread of fissures, resulting in cracks in the weaker parts of the matrix. However, the NCO-2 (Figure 6C) showed lower roughness due to the lower strain rate and the higher maximum stress among the others.

On the other hand, among the cassava starch samples, only NCA-1 (Figure 6E) exhibited a relatively flat and smooth fracture surface, indicating a typical brittle fracture. The brittle fracture surface suggests a low-resistance crack formed within the matrix during the stress. Among all the samples, this was the one with the lowest tensile strength and Young's modulus results (Table 2).

3.6 Fourier Transform Infrared Spectroscopy (FTIR)

Figure 7 shows the FTIR spectra obtained from samples of corn and cassava starch and their respective nanocomposites. A wide peak at 3289 cm^{-1} attributed to the stretching of the OH bond. The peak at 1646 cm^{-1} corresponds to the formation of hydrolysis bonds with water. The 1150 cm^{-1} peak is attributed to the stretching of the C-O glycosidic bond, characteristic stretching of starch. The intense peak at 998 cm^{-1} corresponds to C-O-H vibration[39].

Even with the addition of CNF, there was no change in the characteristic starch bands. CNF is composed of cellulose molecules, also formed by glucose units such as starch. The structural difference between starch and cellulose is in the type of bonds of the D-glucose units: in starch, there are α-1,4 and α-1,6 bonds, while in cellulose these bonds are of the β-1 type, 4 and do not affect the respective infrared spectra[40], so the functional groups of both are the same.

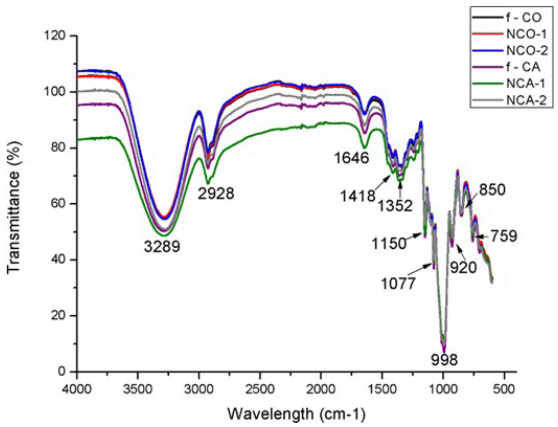

Figure 7. FTIR spectrum of corn and cassava starch-based nanocomposites samples.

3.7 X-Ray diffraction

Figure 8 shows the diffractograms corresponding to the corn and cassava starch used in the preparation of the films.Starch granules have a semi-crystalline structure with crystallinity between 20% and 45%. Two types of crystalline structures can be found in the starch structure: monoclinic or type A (short chains of amylopectin and dense branching), found in cereals, and hexagonal or type B (long and less dense chains of amylopectin), which is found in tubers. There is also a third, type C, which is believed to be a mixture of the first two (He & Wei, 2017). Cassava starch showed peaks at 2θ = 15, 17, 18, and 23°, characteristic of type C polymorphism. The crystallinity indexes observed were corn starch 46.1% and cassava starch 45.9%[41,42].

3.8 Determination of transparency

The spectrum was used in the transmittance mode to assess the degree of transparency of the starch films in the visible region of the spectrum by means of light transmittance (%), as shown in Figure 9.

It was observed that starch films without CNF showed a higher degree of transparency than the others. On the other hand, nanocomposites showed a lower degree of transparency when compared to films without CNF. For this reason, the addition of CNF decreases the transparency of the nanocomposites. The presence of nano reinforcements acts as an obstacle for the passage of light, consequently, the increase of the concentration of CNF in the starch films, decreases the degree of transparency of them. Another point that explains such behavior is that CNFs present crystalline regions, causing an increase in the crystallinity of nanocomposites, and consequently decreasing transparency. Another factor is the semicrystalline granular structure of the starches and the proportion of amylose and amylopectin because the more disorganized the chains are (amorphous), the emptier spaces the material will present, and it is in these empty spaces that the light will pass through, increasing the transparency of the material. Amylopectin has crystalline behavior, while amylose has amorphous behavior, however amylose after gelatinization and transformation into thermoplastic starch is crystalline in character.

When it is heated together with a plasticizer, its semicrystalline structure is destroyed, consequently obtaining an amorphous material, which after cooling, both amylopectin, which presents crystallinity, and amylose, which does not present crystallinity in the form of granules, tend to crystallize, but amylose tends to crystallize earlier than amylopectin. Storage conditions also influence the formation process of crystalline regions of thermoplastic starch[43,44]. Therefore, the greater degree of transparency observed in the cassava starch is due to the higher content of amylopectin, which despite the crystalline character in its granule form, after the gelatinization and cooling process will have its re-crystallization process more time consuming, consequently becoming more amorphous when compared to corn starch film. On the other hand, corn starch has a higher amylose content, after being transformed into thermoplastic starch, it will crystallize more quickly, becoming an opaque material.

After the addition of nano-reinforcement in the films, the situation is reversed and the corn starch-based nanocomposites become more transparent than the cassava starch-based nanocomposites. This may be related to the saturation of nanofibers in cassava starch-based films, making them opaquer and also affecting their tensile strength, as already observed in mechanical tests (Table 2). Another factor relates to the surface roughness of NCA films. The NCA films exhibited a surface rougher than the NCO films, a critical factor in determining film transparency. Surface roughness contributed to an increased optical path length required for light to pass through the film, resulting in higher light absorption. Additionally, surface roughness scattered light, rendering the films more opaque.

Table 3 shows that the greatest variation in transparency along the visible spectrum was observed in the 5.84%

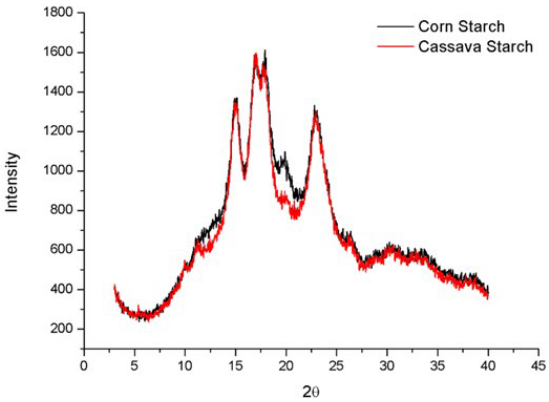

Figure 8. Corn and cassava starch diffractogram.

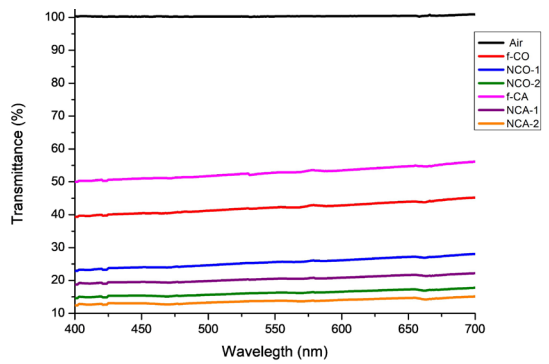

Figure 9. Transmittance of samples in the visible spectrum region. f-CO: Corn Starch film CNF-free; NCO-1: Corn Starch film CNF 1%; NCO-2: Corn Starch film CNF 2%; f-CA: Cassava Starch film CNF-free; NCA-1: Cassava Starch film CNF 1%; NCA-2: Cassava Starch film CNF 2%.

Table 3. Variation of the nanocomposite's transparency across the visible spectrum.

		Degree of transparency of films (%)					
Wavelength (nm)	**Color absorbed**	**f-CO**	**NCO-1**	**NCO-2**	**f-CA**	**NCA-1**	**NCA-2**
400 to 450		39.35 to 40.43	22.91 to 24.02	14.66 to 15.38	50.01 to 51.02	18.79 to 19.56	12.46 to 13.08
450 to 480		40.43 to 40.80	24.02 to 24.20	15.38 to 15.31	51.02 to 51.36	19.56 to 19.49	13.08 to 12.89
480 to 495		40.80 to 41.07	24.20 to 24.53	15.31 to 15.58	51.36 to 51.60	19.49 to 19.76	12.89 to 13.21
495 to 570		41.07 to 42.29	24.53 to 25.64	15.58 to 16.20	51.60 to 53.04	19.76 to 20.50	13.21 to 13.66
570 to 590		42.29 to 42.66	25.64 to 25.98	16.20 to 16.43	53.04 to 53.28	20.50 to 20.71	13.66 to 13.90
590 to 620		42.66 to 43.30	25.98 to 26.56	16.43 to 16.87	53.28 to 54.01	20.71 to 21.18	13.90 to 14.33
620 to 700		43.30 to 45.19	26.56 to 28.05	16.87 to 17.76	54.01 to 56.10	21.18 to 22.23	14.33 to 15.13
Total variation in transparency		5.84	5.14	3.1	6.09	3.44	2.67

variation of cassava starch (f-CO) films, followed by the nanocomposite with 1% nanofiber (NCO-1) with 5.14%, corn starch film (f-CA) with 6.09%, NCA-1 with 3.44%, NCO-2 with 3.1% and NCA-2 with 2.67% variation. Even with variations, the samples obtained the same optical behavior, where transmittance started at 400 nm increasing in percentage to 700nm.

4. Conclusions

In conclusion, this study successfully characterized the physicochemical-mechanical properties of corn and cassava starch films reinforced with cellulose nanofibers (CNF) using the Tape-Casting technique. Despite encountering challenges in the preparation process, such as increased viscosity and bubble formation impacting the mechanical properties of cassava starch films with CNF, the research offered valuable insights into the intricate interplay of factors influencing film characteristics. The findings underscore the importance of considering starch type and CNF concentration in tailoring film properties, providing essential knowledge for potential applications of these nanocomposites. The results uncovered distinct behaviors in mechanical properties, with corn starch films demonstrating an increase in tensile strength with CNF concentration. In contrast, cassava starch films exhibited a more complex response, and despite their higher transparency without CNF, experienced a significant decrease in transparency with the addition of nanofibers. These nuanced outcomes underscore the need for a tailored approach in utilizing starch-based nanocomposites in various applications. The study not only expands our understanding of the interactions between starch and CNF but also provides crucial knowledge for optimizing the potential applications of these nanocomposites.

5. Author's Contribution

- **Conceptualization** – Giovana Ladislau Garuti; Roberta Ranielle Matos de Freitas; Vitor Hugo de Lima; Karina Palmizani do Carmo; Franciane Andrade de Pádua; Vagner Roberto Botaro.
- **Data curation** – Giovana Ladislau Garuti.
- **Formal analysis** – Giovana Ladislau Garuti.
- **Funding acquisition** – Franciane Andrade de Pádua; Vagner Roberto Botaro.
- **Investigation** – Giovana Ladislau Garuti; Roberta Ranielle Matos de Freitas; Vitor Hugo de Lima; Karina Palmizani do Carmo.
- **Methodology** – Giovana Ladislau Garuti; Roberta Ranielle Matos de Freitas; Vitor Hugo de Lima; Vagner Roberto Botaro.
- **Project administration** – Roberta Ranielle Matos de Freitas; Vagner Roberto Botaro.
- **Resources** – Franciane Andrade de Pádua; Vagner Roberto Botaro.
- **Software** – NA.
- **Supervision** – Vagner Roberto Botaro.
- **Validation** – Vagner Roberto Botaro.
- **Visualization** – Giovana Ladislau Garuti; Roberta Ranielle Matos de Freitas; Vitor Hugo de Lima; Karina Palmizani do Carmo; Franciane Andrade de Pádua; Vagner Roberto Botaro.
- **Writing – original draft** – Giovana Ladislau Garuti; Roberta Ranielle Matos de Freitas; Vitor Hugo de Lima; Karina Palmizani do Carmo; Franciane Andrade de Pádua; Vagner Roberto Botaro.
- **Writing – review & editing** – Giovana Ladislau Garuti; Roberta Ranielle Matos de Freitas; Vitor Hugo de Lima; Karina Palmizani do Carmo; Franciane Andrade de Pádua; Vagner Roberto Botaro.

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