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Article

STUDY OF MECHANICAL PROPERTIES IN PAPER PACKAGES BASED ON POLYACRYLAMIDE AND NATURAL FIBERS

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Abstract: Natural fibers have been extensively studied as a reinforcement filler in obtaining composites, replacing partially synthetic fibers. The vast majority of these materials originate from agro-industrial waste with a high content of lignin and cellulose making it a very interesting material with low cost and good mechanical properties. The purpose of the study was to obtain a composite based on alkyl ketene dimer resin, for the manufacture of sustainable packaging, made of paper by adding a 10% (w/w) con-tent of green coconut fiber and sugarcane bagasse fiber, and evaluated the impact of the filler on the mechanical behavior of the systems. The studied material was characterized from mechanical tests, such as Ring Crush Test (RCT) and Concora Medium Test (CMT) evaluating the maximum resistance supported by centimeter in the pre-pared composites, through the specimens. It was observed that the composite prepared with the sugarcane bagasse fiber presented the most satisfactory results, thus converging on the material with better mechanical properties, when compared to the compo-site obtained with green coconut fiber. Therefore, the study was funneled with sugar cane fiber, varying the content by 20 and 30 % (w/w), evaluating the impact on the dispersion of this filler in the polymeric matrix and, consequently, the mechanical response of the composite with these compositions.

Keywords: Natural fibers; Sustainable packaging; Mechanical properties.

1. Introduction

Natural fibers have been extensively studied as a reinforcement load in obtaining composites, replacing partially synthetic fibers. Another point is that most of these products originate from agroindustrial residues with a high content of lignin and cellulose making it a very interesting material with low cost and good mechanical properties, such as low specific mass, biodegradability, processing flexibility, renewable sources and non-toxic (Annergren *et al*., 1962; Agrawal *et al*., 2000; Anjos *et al*., 2005). Due to the presence of these chemical structures, these materials have characteristics that can give the systems good mechanical proper-ties in the materials produced (Battistel *et al*., 2001; Callister, 2006). Sugarcane is widely cultivated in tropical and subtropical regions of the country (Campos, 2009), and in its chemical structure it has about 45% lignocellulosic fibers, 2 to 3% insoluble solids and 2 to 3% soluble solids. This composition makes it interesting to use sugarcane bagasse with high reinforcement viability in preparing composites. The consumption of coconut water and coconut pulp generates a quantity of waste. These shells are generally used as boiler fuel or used for the processing of fibers in the production of carpets, mats and other products (Carrijo *et al*., 2002).

For the preparation of composites using natural fibers, it is necessary to deal with the hydrophilic character of this type of filler, which makes them incompatible with the polymer during the process of dispersing the filler in the polymeric matrix (Cuéllar, Muñoz, 2009). Therefore, methods for removing lignin and cellulose through chemical and physical modification (Danielsson, Lindström, 2005; Garcez, 2005; Köhnke, Gatenholm, 2007) are foreseen in the literature, as well as the use of enzymes (Köhnke *et al*., 2008; Luz *et al*., 2008) as a functionalizing agent on the surfaces of the fiber to improve the interaction of the charge with the polymer.

One way of using fibers, through their good mechanical properties, is through the manufacture of paper. The xylan content present in the fiber directly affects the proper-ties of the paper (Battistel *et al*., 2001; Mohanty *et al*., 2002; Molin, Teder, 2002; Rosa *et al*., 2002; Molina *et al*., 2008; Pejic *et al*., Callister, 2006; 2008; Santchurn *et al*., 2012). The mechanical tests of Ring Crush Test and Concora Medium Test were carried out to evaluate the mechanical strength of the prepared paper composites. From these tests it was possible to analyze the load capacity that the material withstands every centimeter of the paper. The composite prepared with sugar cane bagasse fiber presented the best of the mechanical tests, this was attributed to the chemical properties of this fiber, providing greater resistance when compared to green coconut fiber.

2. Materials and Methods

2.1 Materials

The alkyl ketamine dimers (AKD) glue agent purchased from Dynacol - Dynatech was used. The cationic polyacrylamide polymer 9510 HC purchased from Axchem (viscosity = 2.7-4.1 cps). The sodium hydroxide base (NaOH) was acquired from Pooltécnica Química (Mw = 40.1 g / mol; concentration = 5.0%). Green coconut fiber and sugar cane bagasse. Distilled water was used for washing.

2.2 Preparation of composites based on polyacrylamide/natural fibers of coconut and sugar cane

2.2.1 Stripping

In the peeling process, it was carried out manually by removing the outer part of the green coconut fruit using only the fibrous part. In the case of sugar cane, the bark was separated using only the bagasse.

2.2.2 Chopping

The chopping process of both green coconut fiber and sugar cane was done manually, obtaining cuts with sizes between 2 to 5 millimeters, so that there was a reduction in the length of the fibers, and thus optimizing the homogenization in the mixing process.

2.2.3 Cooking

During the cooking process, 0.085 liters of sodium hydroxide solution (NaOH) were mixed with 50g of each dry fiber and added to 1.0 liter of distilled water, to hydrate the fibers and thereby make them more permeable. The sodium hydroxide helped to in-crease the permeability of the fibers through the delignification process.

The cooking process took place at 120 °C for 90 minutes, and then the material was washed with distilled water and sieved. After cooking, the fibers were dried in an oven at 100 °C until the solvents were completely evaporated.

2.2.4 Physical separation of fibers

The process of separating the threads from the fibers was carried out using a mixer with short blades, to only separate and not cut into smaller sizes. Dry recycled paper with 1.0 liter of distilled water and fiber were added to the mixer cup. The proportion of each input was modified for each test in order to find the best ratio for obtaining the composite. The ideal density for this system was 0.699 g/l.

2.2.5 Fiber suspension

To prepare the suspension with the fibers, a Styrofoam container with a capacity of 80 liters was used, adding a proportion of 40 grams of fiber to 57.2 liters of distilled water to this container. The flotation process was achieved by adding from the cationic polymerization of the system, with the addition of 0.104 liters of polyacrylamide polymer solution. At the end, 0.075 liters of the alkyl ketene dimer glue were added to the solution.

2.2.6 Obtaining the packaging paper

To obtain the sheets of paper prepared with the fibers, it consisted of immersing the suspension prepared with a canvas to distribute evenly, achieving a better result in the interlacing of the fibers and in the appearance of the paper obtained. Uniform distribution was essential to achieve better mechanical properties of the material.

2.2.7 Drying

The drying of the leaves occurred in an oven, controlling the temperature at 115 °C, for 25 minutes. Subsequently, the paper sheet was placed on a ceramic plate for 20 minutes, followed by another 5 minutes at 115 °C in the oven, to ensure drying with total evaporation of the solvent.

2.3 Characterization of composites

From the leaves obtained with the addition of fibers, triplicate tests were carried out to evaluate the mechanical properties. For the beginning of the mechanical tests, the specimens were placed in the oven at 115 °C, for a period of 40 seconds, to guarantee the absence of solvent for the execution of the tests.

The Ring Crush Test (RCT) mechanical test was used to determine the maximum amount of compressive strength presented by the material. The RCT crushing disc from Quality Labor and the mechanical crush tester press 9 (MODEL MCT-400), from Tecnomeca, were used to perform the RCT test. The paper was cut into three strips of paper with dimensions of 12.7 mm ± 0.051 mm wide and 152.4 mm ± 0.051 mm long, using a guillotine. After obtaining the specimens, they were inserted vertically into the equipment's crushing discs with the size from 0.37 mm to 0.42 mm and placed in the lower press plate for the Ring Crush Test (RCT). The tests were concluded with the start of the buckling of the specimens, reaching the maximum value obtained. The test was performed based on the NBR ISO 9001: 2015 item 7.1.5 of the standard, with guidance of NBR 14192 - paper and cardboard - determination of the compressive strength - method of crushing the ring.

The mechanical test of Concora Medium Test (CMT) was performed on the soft paper to determine its maximum capacity to resist compression forces, that is, crush resistance. The samples are cut and the waves are formed in a transverse direction [20]. To perform the Concora Medium Test (CMT) the specimens were cut 12.7 mm ± 0.051 mm wide and 152.4 mm ± 0.051 mm long with the aid of guillotine and placed in the Global Enterprises Concora Medium Flutter equipment at a temperature of 178 °C. After removing the specimen, it was placed on the comb and rack, a set of Tecnomeca equipment, to check if the waves made on the paper are in conformity. Then the paper sample placed on the bottom plate of Tecnomeca's mechanical crush-tester 9 (MODEL MCT-400), with the waves facing upwards for the compression test. The test was carried out based on the NBR ISO 9001: 2015 standard, item 7.1.5 of the standard, with guidance to NBR ISO 7263 - determination of crush strength after being corrugated.

3. Results and Discussion

The tests were performed with a paper sample without the addition of fibers and varying the content of these loads by 10, 20 and 30% (w/w) of fibers in the preparation of the systems analyzed to assess the impact of the variation in the load content on the mechanical properties.

3.1. Mechanical RCT tests for 10% (w/w)

The Table 1 shows the values obtained from the mechanical tests of Ring Crush Test (RCT) performed with composites of 10% (w/w) natural fiber compared to paper without the load, evidencing the impact of the load on the mechanical properties. The coconut fiber composite showed a decrease compared to unloaded paper. This can be attributed to an inefficient dispersion, causing weak points in the composite, introducing defects in the matrix (Schönberg *et al*., 2001). But also, it can be attributed to a wear of the fiber by the caustic treatment (Shin, Stromberg, 2006). For the cane bagasse fiber composite, there was a reinforcing effect due to the presence of bagasse xylan, increasing the material's resistance (Sihtola, Blomberg, 1975; Battistel *et al*., 2001; Sjöberg *et al*., 2002; Callister, 2006; Silva, 2006; Molina *et al*., 2008; Santchurn *et al*., 2012).

Table 1. Results of RCT mechanical tests of composites obtained with 10% by weight of natural fiber filler.

Specimen	Composite with unloaded paper	Green coconut fiber composite	Sugarcane fiber composite
	[kgf/cm]	[kgf/cm]	[kgf/cm]
1	8.0	6.5	8.0
2	7.0	6.5	8.0
3	7.0	6.0	8.0

3.2. Mechanical CMT tests for 10% (w/w)

The Table 2 shows the results achieved by the mechanical tests of the Concora Medium Test (CMT), for 10% (w/w) of concentration of the coconut fibers and sugarcane bagasse. For CMT tests, it was observed that the addition of fibers increased the maxi-mum capacity of resistance to the forces of compression, evidence or reinforcement in the systems. The treatment in the cooking stage with the addition of NaOH in the green coconut fiber, decreased its mechanical resistance and consequently the reinforcement in the matrix, reaching lower values in the tests compared to the composite applied with the sugarcane bagasse. This can be attributed to the fact that a coconut fiber contains a lower percentage of thread and in relation to the thread content present in sugar cane cake (Sun *et al*., 2000; Van de Weyenberg *et al*.,2003).

Table2. Results of CMT mechanical tests of composites obtained with 10% by weight of natural fiber filler.

Specimen	Composite with unloaded paper	Green coconut fiber composite	Sugarcane fiber composite
	[kgf/cm]	[kgf/cm]	[kgf/cm]
1	5.4	6.3	8.2
2	4.5	7.7	8.6
3	5.1	7.5	8.2

From the studies carried out through the mechanical tests of RCT and CMT, using specimens of paper composites, whose composition was varied with the presence of 10% coconut fiber or 10% sugar cane bagasse fiber, it was found that systems pre-pared with sugarcane fiber bagasse achieved the best results of mechanical properties. In view of this, further studies were made on the impact of varying the percentage of sugar cane fiber by 20 and 30% in the preparation of composites.

3.3. Mechanical RCT tests for 20% e 30% (w/w) of natural sugar cane bagasse fiber

The Table 3 shows the results comparing the composites obtained with the sugar cane bagasse fiber, varying the concentration by 20 and 30% (w/w). It is possible to observe that with the load increase to 20%, there was an increase in the mechanical property, showing the reinforcement effect with the presence of fiber in the composite, when compared to the system without fiber. On the other hand, with the addition of 30% fiber to the paper, it was possible to observe a decrease in the mechanical property, since the natural fibers have an essentially hydrophilic structure, and there may be the formation of fiber agglomerates (Danielsson, Lindström, 2005), as well as their dispersed size in the polymeric matrix, significantly impacting and reducing mechanical performance (Yang *et al.*, 2008).

Table3. Results of the RCT mechanical tests obtained with a 20 and 30% (w/w) composite of natural fiber from sugarcane bagasse.

Specimen	Composite with unloaded paper	Composite with 20% sugar cane fiber	Composite with 30% sugar cane fiber
	[kgf/cm]	[kgf/cm]	[kgf/cm]
1	8	8.5	7
2	7	8.6	7
3	7	10	8

Figure 1 illustrates the impact of the reinforcement property with the addition of fiber to the paper. With the percentage load increase by 10 and 20% (w/w), there was an in-crease in the compressive strength. However, with a higher content, 30% (w/w) of sugarcane fiber, due to a hydrophilic fiber resource, favored by the formation of agglomerates (Danielsson, Lindström, 2005), using a chemical resistance capacity of this compound.

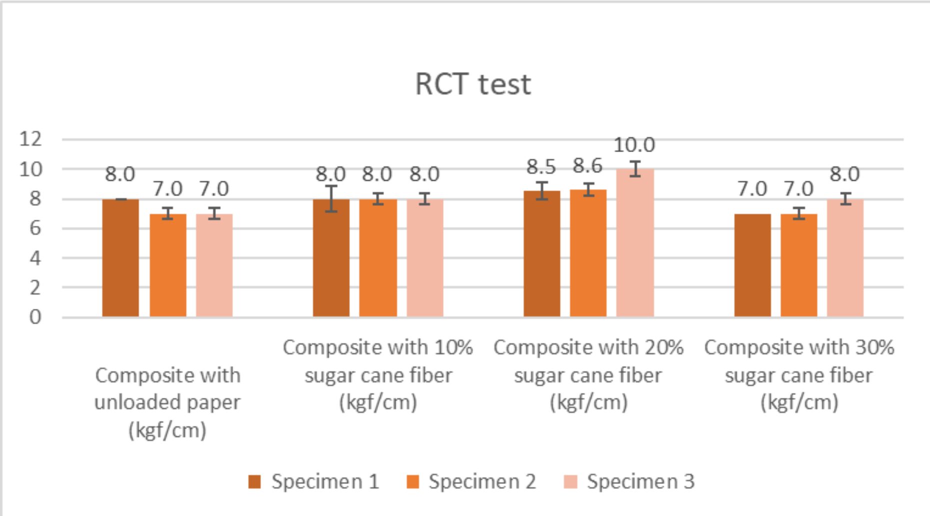

Figure 1. Comparative results of the RCT test between paper without fiber and composites with 10, 20 and 30% (w/w) of natural fiber.

3.4. Mechanical CMT tests for 20% e 30% (w/w) of natural sugar cane bagasse fiber

The Table 4 shows the impact of adding sugar cane bagasse fiber on the property of maxi-mum resistance to compression forces. With the addition of the load, it favored the mechanical property (Sihtola, Blomberg, 1975; Battistel *et al.,* 2001; Sjöberg *et al.,* 2002; Callister, 2006; Silva, 2006; Molina *et al.,* 2008; Santchurn *et al*., 2012). On the other hand, with the high-est content, 30% (w/w) of the sugar cane bagasse fiber, due to the hydrophilic charac-teristic of the fiber, favored the formation of agglomerates [8], reducing the maximum resistance capacity to compressive forces with this content higher in the composite.

Table 4. Results of the CMT mechanical tests obtained with a 20 and 30% (w/w) composite of natural fiber from sugarcane bagasse.

Specimen	Composite with unloaded paper	Composite with 20% sugar cane fiber	Composite with 30% sugar cane fiber
	[kgf/cm]	[kgf/cm]	[kgf/cm]
1	5.4	8.6	5.9
2	4.5	8.6	5.9
3	5.1	8.6	6.3

It is possible to observe in Figure 2 the impact of the addition of load in the mechanical test of maximum capacity of resistance to compressive forces. With the increase in fiber content by 10 and 20% (w/w), there was an optimization of the mechanical property.

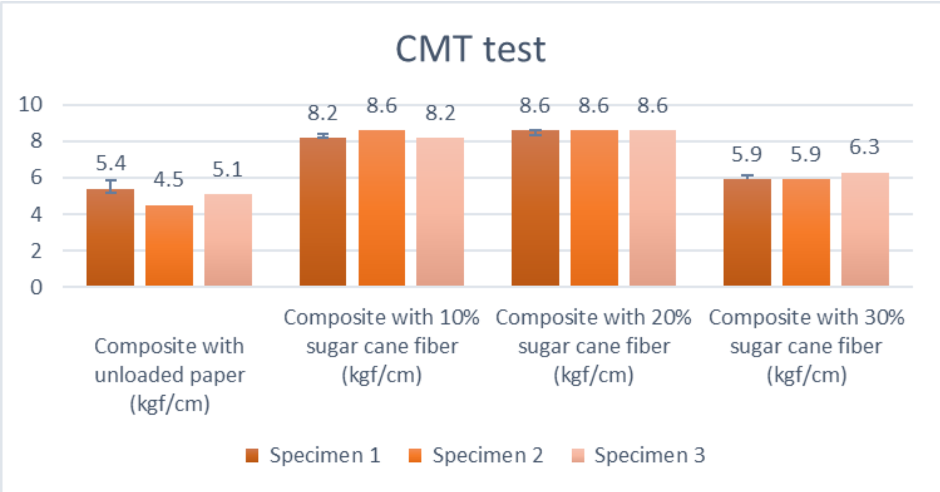

Figure 2. Comparative results of the CMT test between paper without fiber and composites with 10, 20 and 30% (w / w) of natural fiber.

However, with a greater concentration of 30% fiber, due to the hydrophilic characteristic of this load, it favored the formation of agglomerates (Danielsson, Lindström, 2005), reducing the resistance to compression.

4. Conclusions

In general, the addition of fiber to obtain the paper composite was satisfactory when compared to unloaded paper, and this showed that the addition of the fiber promoted a reinforcing effect in the systems, both in the RCT and CMT tests.

In the RCT tests, where 10% (w/w) coconut fiber and sugarcane bagasse were used, it was possible to verify that the composite prepared with sugar cane fiber presented more satisfactory results, attributing its chemical composition with a percentage of components that they add mechanical reinforcement characteristics, but can also be attributed to the dispersion efficiency of the loads in the polymeric matrix.

With the addition of 20% (w/w) of the sugar cane fiber there was an increase in the mechanical strength of the composite, showing optimization in the mechanical proper-ties. In contrast, when the load content increased to 30% (w/w), a decrease in mechanical strength was observed through the RCT and CMT tests. This was attributed to the hydrophilic characteristic of the fiber, favoring the formation of agglomerates, and due to this deficient dispersion, it had a negative impact on the mechanical response of this system.

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Conflicts of Interest: The authors declare no conflict of interest.

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