Properties of Lignocellulosic Composites Containing Regenerated Cellulose Fibers

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The aim of the paper was to examine the application of regenerated cellulose fibers as a reinforcement material in particleboard production. Single-layer, 10 mm thick panels, with the density of 800 kg/m^3 were produced with addition of regenerated cellulose fibers in the range of 0 do 15% by mass during panels' production. The mechanical and physical parameters of the produced panels were tested, as well as work of fracture. The results showed that addition of regenerated cellulose fibers to the structure of wood-based composite did not improve their modulus of rupture, modulus of elasticity, or internal bond. The physical parameters of the produced panels (water absorptivity, swelling in thickness when soaking) also were reduced. The work of fracture of the tested panels increased with increasing content of regenerated cellulose fibers. A strong linear regression between work of fracture and regenerated cellulose fibers content was observed.

Keywords: Regenerated cellulose fiber; Viscose; Particleboard; Composite; Work of fracture; Mechanics; Modulus of rupture (MOR); Modulus of elasticity (MOE)

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INTRODUCTION

The range of potential applications of cellulose fibers, which can be regenerated, has been growing. This trend is especially intensive in research of materials for packaging and printing purposes. There have been successful trials to produce printable films from kenaf cellulose/polyvinyl alcohol (Kaco *et al.* 2014). Such films can be de-inked and re-printed, and the print quality and number of re-prints is growing with polyvinyl alcohol/cellulose ratio. While it is important that the cellulose fibers source is a non-wood plant, it is highly desirable that these can be produced also from short-rotation crops plantation.

Although there are other types of regenerated cellulose fibers (RCF) that exhibit better mechanical (Gindl *et al.* 2006) and physical (reaction to water) (Kreze *et al.* 2002) parameters, the economic aspects still provide for a higher popularity of viscose fibers. The development of pro-ecologic methods of next generation cellulosic fibers production (Lyocell – 3rd generation) leads to reductions in amounts of toxic substances from the production, as well as to shortening and simplification of the processes. Additionally, the solvents used in production of new generation regenerated cellulose fibers are recovered with an efficiency of 99.5%.

According to Abbott and Bismarck (2010), there are possibilities to produce a self-reinforced cellulosic material from regenerated cellulose microcrystals. The level of reinforcement can be controlled by tailoring the cellulose crystallinity by changing the dissolution of microcrystalline cellulose (MCC) before the regeneration process. The dissolution of microcrystalline cellulose, which can be controlled, results in the different structural, thermal, and physical properties, as well as influences on reaction to water of regenerated cellulose.

Stanzl-Tschegg (2006) showed that the ability of the material to be divided into smaller parts depends on several parameters, *e.g.* fracture toughness, specific fracture energy, mean density, fibers' orientation, *etc*. Different properties of fracture influence the load-carrying capacity of the material when applied in construction, but also have an impact when processed by typical tools for wood and wood-based materials machining. These tools have the wedge-shape edges, so the results of measuring of fracture properties can provide information about further machining.

The improvement of the fracture properties of wood loaded across the grain, by addition of reinforcing fibres, was suggested by Jeronimidis (1980). Due to this, some of the morphological features of wood cell walls, responsible for energy absorption when cracking, can be improved. Since the typical, commercial particleboard is also quite a brittle material, introduction of an additional, fibrous component has the potential to change the fracture energy characteristics. Regenerated cellulose fibers have favorable strength-to-weight ratio due to small fiber thickness. They can be easily deformed and incorporated between wood particles, causing mechanical features improvement. The potential application of the panels with increased energy needed to divide them, especially by the forces working in the plane of the panel, may consist of the web parts of an engineered wood product, such as an I-joist, where the more expensive OSB panels are used nowadays. The properties of such a composite deserve to be studied in detail, since the potential advantages of increased energy absorption may need to be weighed relative to a likely higher usage of energy during cutting.

The aim of this work was to investigate selected mechanical and physical properties, including work of fracture, of the particleboards produced with different content of regenerated cellulose fibers in their structure.

EXPERIMENTAL

Materials

The investigated material in this work were single layer particleboards, produced with the following:

- Industrial particles applied in particleboard plant to surface layer production (over 90% of coniferous wood species, mostly *Pinus sylvestris* L.), with particles' size in the range of >0.5 mm and <4 mm; the small dimensions of the particles were selected to avoid the influence of irregularity of wood particles' properties on the scattering of results of mechanical tests of lignocellulosic composites (Kociszewski *et al.* 2012),
- RCF, 3 denier, acquired as a 50 mm long viscose fiber mat, which was pneumatically fragmented to fibers' mass (kind of dry pulp) with low bulk density (about 5 kg/m^3),
- Commercial, industrial urea-formaldehyde (UF) resin, used in particleboard industry,
- Aqueous solution of NH_4Cl as hardener.

The following production parameters were applied: amount of RCF added – 0, 5, 10, and 15% respect to weight of oven dry wood particles (hereinafter called *panel type*), assumed density – 800 kg/m^3, panel thickness – 10 mm, RCF/wood particles resination – 10%, dry mass of resin/dry mass of hardener ratio – 0.01, press temperature – 180 ºC, pressing cycle (pressure and time): closing 20 s, 2.5 MPa – 70 s, 1.7 MPa – 36 s, 0.8 MPa – 36 s, opening 18 s.

The wood particles were blended with RCF in drum blending machine during glue spraying. Prior to testing the produced panels were stored in 65% R.H./20 ºC conditions to stabilize their weight.

Methods

After conditioning, the panels were cut into the necessary samples. The following tests were conducted:

- Determination of modulus of elasticity in bending (MOE) and of bending strength (modulus of rupture - MOR) according to EN 310:1994,
- Determination of tensile strength perpendicular to the plane of the board (internal bond - IB) according to EN 319:1999,
- Determination of swelling in thickness after immersion in water according to EN 317:1999,
- Measurement of water absorptivity after 2 and 24 h of soaking in room temperature water,
- Density profile on GreCon Da-X (x-ray) analyzer, with 0.02 mm panel thickness sampling step,
- Work of fracture, measured on standard testing machine equipped with load-displacement recorder, with the load methodology presented by Matsumoto and Nairn (2012) (where the load forces are in the plane of the panels), and calculated as area under load-displacement curve (examples in Fig. 4) according to Eq. 1; the shape and dimension of the samples used to work of fracture measurement are displayed on Fig. 1.

Not less than 12 samples were used in mechanical and physical tests. The work of fraction was calculated as shown in Eq. 1,

$$W_f = \sum \frac{(P_i + P_{i+1}) \cdot (a_{i+1} - a_i)}{2} \quad \text{[Nm]} \tag{1}$$

where W_f is the work of fracture [Nm], P_i is the interim load value [N], and a_i is the interim displacement value [m].

The shape and dimensions of the samples used for fracture measurement, displayed in Fig. 1, are based on the sample proposed by Matsumoto and Nairn (2012). However, after initial tests, the height of the sample was changed from 90 to 110 mm.

The preliminary tests showed that the crack propagation did not run in exactly the proper direction, resulting in an irregular surface of breakage, which was not acceptable for making comparisons. The problem of deformation (bending) of the side parts of samples subjected to the wedge-splitting testing method was indicated also by Ehart *et al.* (1996).

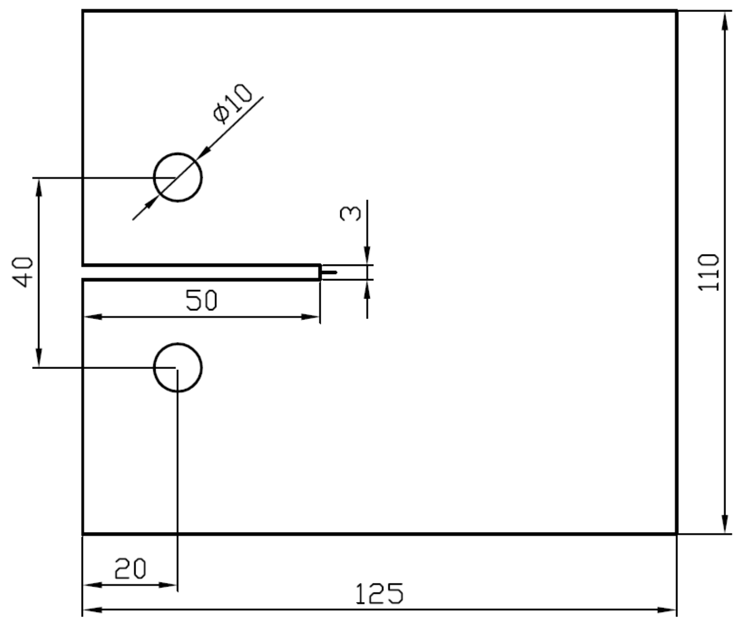

Fig. 1. Shape and dimensions (in millimeters) of the samples used to work of fracture measurement (based on Matsumoto and Nairn 2012)

RESULTS AND DISCUSSION

Density Profile

The results of the density profile measurement show that the density profiles of the panels with different RCF content differed mostly in the near surface regions (to about 1.5 mm in thickness) (Fig. 2). In case of samples without RCF content (0), the maximum density occurred about 1.3 mm under surface. This could be easily recognized, despite the fact that the panels were a single layer.

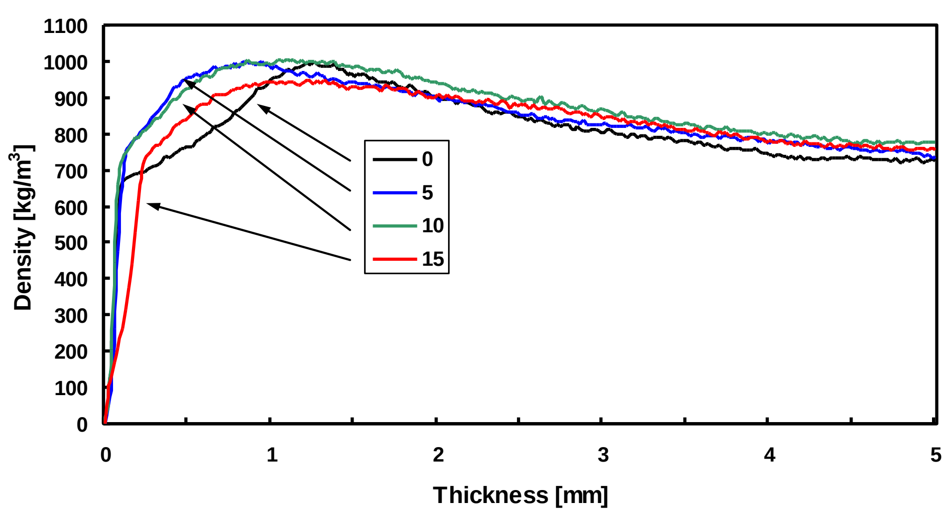

Fig. 2. Density profiles of the tested panels (half of thickness)

With the RCF content increase from 5 to 10%, the density peak became less intensive and more flat, and the maximum density was located closer to the surface (about 1 mm under surface). With increasing RCF content, the entire density profiles became more flat, and the maximum/minimum density differences were smaller. This was clearly visible in the case of panel type 15. The reason for this is due to the thermal insulating properties of RCF. The heat transfer during pressing the panels with RCF content into the core was less intensive. Since the raised amount of heat energy accumulated close to the panels' surface, these layers were more compressed.

Modulus of Rupture

The values of the modulus of rupture of the investigated panels are shown in Fig. 3a. There was a significant decrease of the bending strength, especially when the RCF content was 5%, compared to the panel without RCF content. The reason of this MOR decrease of the panels with RCF content, compared to panel type 0, can be the significant change in the density profile, as presented in Fig. 2. In case of material bending, the bending strength depends mostly on the tension/compression strength of the surface layers. In the investigated samples with non-zero RCF content, the density of the surface layers was flattened and is not clearly marked as it can be seen for panel type 0. The differences between average MOR values were statistically significant between following panel types: 0 and 5, as well as 0 and 10. There was no statistically significant difference between panel type 15 and the remaining panels.

Modulus of Elasticity

The results of modulus of elasticity testing showed that the highest values were for panel type 0 (Fig. 3a). The MOE for the panels with RCF content from 5 to 15% was about 23% smaller compared to panel type 0. This provides information that the addition of regenerated cellulose fibers to particleboard during its production gives rise to less rigid and less brittle properties of the panel. The MOE decrease with RCF addition can be explained by the nature of cellulose fibers, which are incorporated into wood particles structure. Since the fibers, which were significantly longer than the wood particles, are not specially oriented and are randomly mixed with wood particles, the stiffness of such composite is based mostly on wood particles, which present higher elastic ability in the mentioned panel. In the described structure, the RCF do not improve the elastic properties, irrespective of their amount in the panel. However, the average MOE value differences are statistically significant only between panel type 0 compared to the rest of the panels. According to Gindl *et al.* (2006), a better solution to improve the mechanical properties of wood particles – regenerated cellulose fibers composite – could be the application of Lyocell fibers, which have an elastic modulus of about 22,300 N/mm^2 (11,600 N/mm^2 for viscose), and tensile strength of about 750 N/mm^2 (310 N/mm^2 for viscose).

Internal Bond

The internal bond of the panels produced with different regenerated cellulose fibers content decreased with increasing RCF content (Fig. 3b) in the range of 0 to 15% RCF content. Regenerated cellulose fibers, as it was mentioned above, have quite high mechanical properties, compared to wood particles, but since these fibers are distributed randomly in the composite, without any set orientation, these high features did not result in strength improvement of produced panel. According to observations of faces and cross cuts, the cellulose fibers were partly curled up, and they had a tendency to stick to adjacent fibers. Due to the mat forming and pressing system (horizontal) in the resulting structure, the fibers were oriented mainly in bi-dimensional space. Additionally, the wettability of RCF fibers is smaller than wood particles, which has the potential to result in a reduced ability to become covered with water-soluble UF resin. Although, according to the regression line presented, the decrease was asymptotic, and the increase of RCF content could potentially have increased the internal bond of the composites. However, the differences between average IB values were not statistically significantly different.

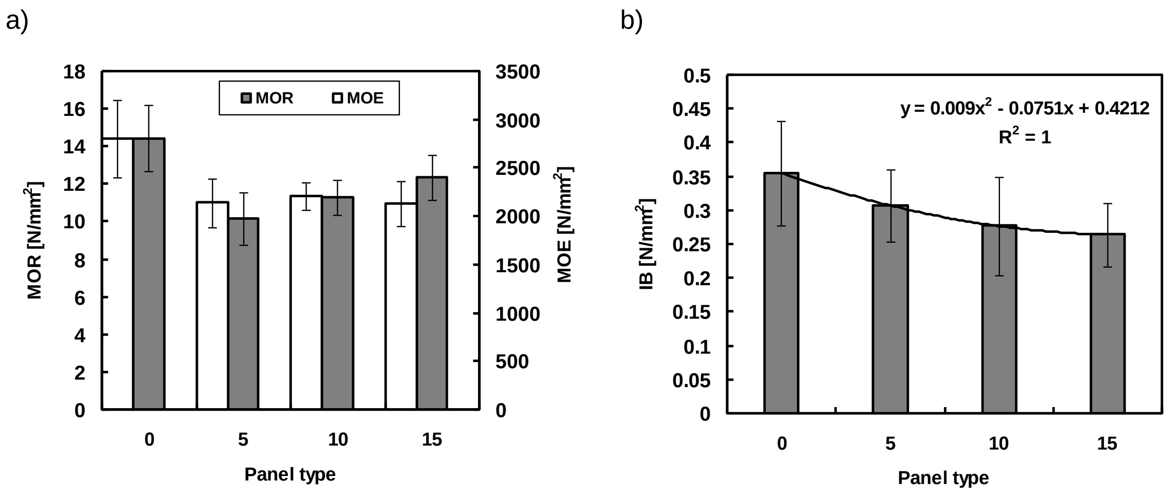

Fig. 3. Modulus of rupture a), modulus of elasticity a) and internal bond b) of tested composites

Water Absorption

When analyzing the results of water absorptivity (Fig. 4a), it can be noted that a significant difference occurred when comparing panel types 0 and 5, when the increase of water absorption was observed, both, after 2 and 24 h of soaking. The average water absorptivity for panel types 5, 10, and 15 after 2 h of soaking was equal and was about 87%, while for panel type 0 it was 73%. After 24 h of soaking, the water absorptivity for 0 panel type was 90%, which corresponds to a percentage point increase of about 25. The average water absorptivity for panel types 0 to 15 after 24 h of soaking was about 97%, which corresponds to less than 12 percentage points of increase. It can be concluded that the presence of the RCF in the composite structure resulted in a greater intensity of the water uptake at the beginning of soaking. This remark can be explained by higher capillarity of panels with RCF content. Such structure, although less porous, can be more easily penetrated by water, which is additionally sucked into the panel by capillarity forces. There was no significant influence of the RCF content (in the investigated range) on the tested panels’ water absorptivity.

Thickness Swelling

The thickness swelling measurement results are displayed in Fig. 4b. The results show that there was a slight difference between thickness swelling of panel types 0, 10, and 15 (28% for panel type 0 after 2 h and 31%, average for panel types 10 and 15). The only significant difference was exhibited by the panel type 5 (38% after 2 h). The thickness swelling of panel types 0, 10, and 15 after 24 h of soaking was almost equal (about 37%), where for panel type 5 it was 47%. According to Kreze *et al.* (2002), the swelling and water absorptivity could be reduced when applying, *e.g.*, Lyocell fibers instead of viscose. The Lyocell fibers have significantly lower swelling in aqueous medium (22%) compared to viscose (over 36%).

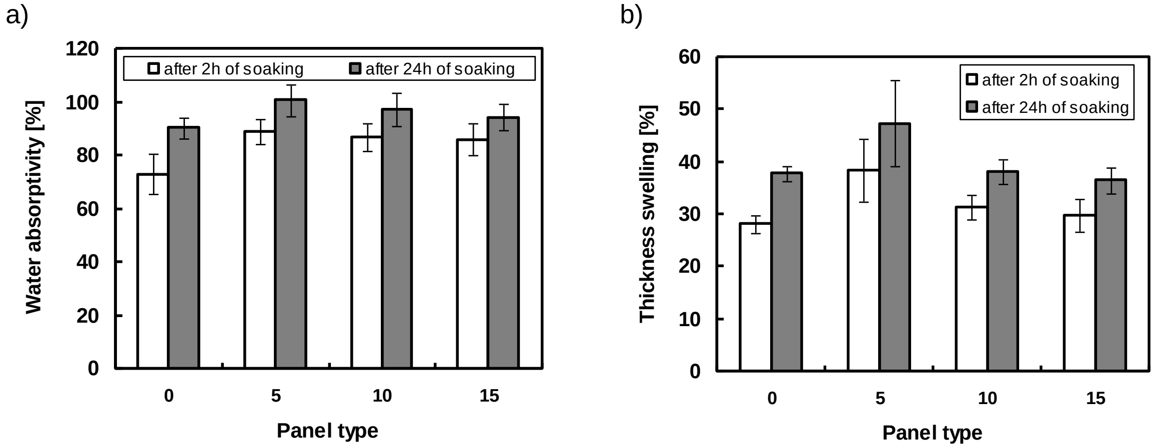

Fig. 4. Physical properties of investigated panels: water absorptivity a) and thickness swelling b)

Load-Displacement Measurement

Examples of the measurement of load-displacement values for the purpose of work of fracture calculations are presented in Fig. 4. Since the work of fracture is defined as the area under the plot, the differences of the composites, which could be explained by the influence of other mechanical parameters, were noticeable. In case of panel type 0, the load-displacement curve was narrow and had a rapid slope after maximum load. This can be caused by the highest MOE, as mentioned above, which implies high brittleness and low plasticity. With the RCF content increasing (panel type 5), where the modulus of elasticity was lowest, also the load value was lowest, but the load-displacement slope was not so intensive; thus, the area under the plot was increased.

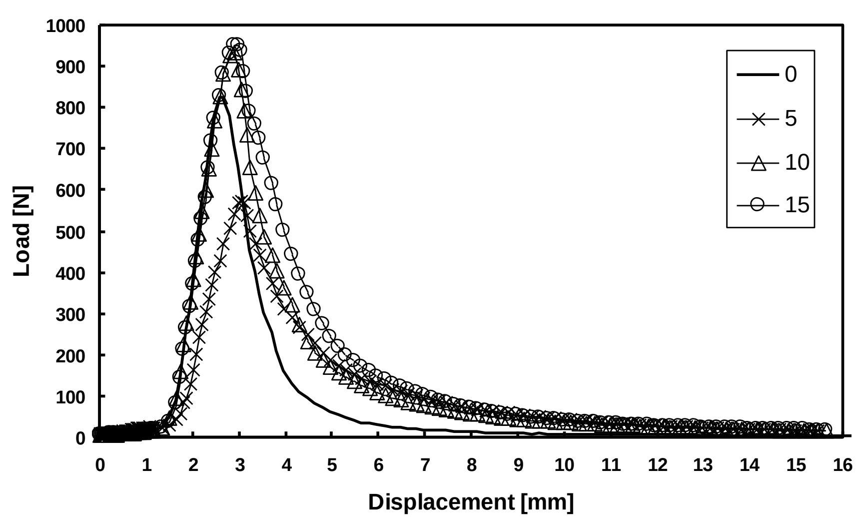

Fig. 4. The influence of regenerated cellulose fibers content in the panels on load-displacement curves

With increasing content of the regenerated cellulose fibers, the maximum load increased. The higher amount of cellulose fibers causes a “bridging effect”, which was also observed by Matsumoto and Nairn (2009), when testing medium density fiberboard (MDF) (Fig. 6Fig.).

Fig. 6. Example of crack of the panel with 10% RCF content (the “bridging effect”)

Work of Fracture

The results of calculation of work of fracture for the investigated panels are displayed in Fig. 7. The lowest average value of work of fracture, 1.36 Nm, was noted for panel type 0, while the highest, 3.00 Nm, was for panel type 15. As it is presented, the dependence between work of fracture and regenerated cellulose fibers content in the panels was almost perfectly linear (R^2=0.992). The increase of RCF content from 0 to 15% caused an increase of over 120% in the work of fracture.

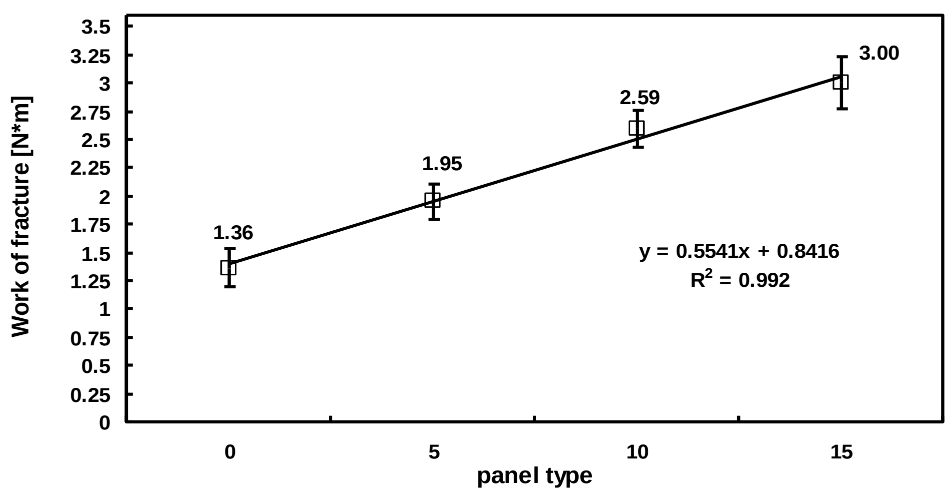

Fig. 7. Dependence of work of fracture of the investigated panels on RCF content

In light of machining of lignocellulosic materials, such as particleboards, the fracture toughness, which can be a base to calculate the energy release rate, can help with quantification of the energy amount needed to separate material during the machining process. According to Sinn *et al.* (2008), there is significant influence of the core structure of particleboard (such as the content of recycled wood) on reduction of fracture energy, whereas the coating type of the panels do not influence this property to a noticeable extent. The further investigation of machining aspects of lignocellulosic composites with RCF would make it possible to describe the interdependence of work of fracture and cutting forces.

CONCLUSIONS

1. The increase of regenerated cellulose fibers content in single layer particleboards in the range of 0 to 15% causes a decrease of mechanical parameters (modulus of rupture, modulus of elasticity, internal bond).
2. The density profile of the panels with increased regenerated cellulose fibers content becomes flatter; the differences between highest and lowest density is reduced. This results in a lowering of modulus of rupture.
3. The presence of regenerated cellulose fibers in particleboard structure intensifies water uptake at the beginning of soaking, whereas the water absorptivity intensity decreases with time.
4. The work of fracture of lignocellulosic composite containing regenerated cellulose fibers increases linearly and significantly with increasing fiber content; the increase of the fiber content from 0 to 15% results in an increase of 120% in the work of fracture.

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