PROPERTIES OF BIOCOMPOSITES BASED ON CELULOSE ACETATE BUTIRATE REINFORCED WITH LYOCELL REGENERATED CELLULOSE FIBERS

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ABSTRACT

In the present work, a biocomposite is prepared using cellulose acetate butyrate (CAB) as a biodegradable polymer matrix and short lyocell regenerated cellulose fibres as reinforcement. Mechanical tensile testing, water absorption test, fracture surface microstructure scanning electron microscopic analysis and soil burial test were performed to study the properties of the composite for several mass compositions of fibre reinforcement.

1. INTRODUCTION

The development of natural fibres (from agricultural resources and/or wastes) reinforced composites has become an emerging area of interest in several areas of technical applications. Most studies have been focus on the use of cellulose fibres [1-5] (wood, flax, etc.) as a reinforcement of several thermosetting and thermoplastic synthetic matrices. The advantages of using cellulose based reinforcements, in front the conventional synthetic fibres (glass, carbon or aramid) are: low density, environmental considerations, non-abrasive to processing and low cost [2]. However, most synthetic matrices used to prepare these composites are based on non-degradable polymers (polyethylene, polypropylene, etc.) that cause a waste-disposal problem after their life cycle has been completed. For this reason, the use of new or modified existing degradable polymers as a composite matrix, such as cellulose acetates, that leads to cellulose reinforced composites with properties comparable to the existing ones is an emerging area of interest [6].

Among all cellulose fibres, regenerated cellulose fibres can be obtained from manufactured processes by dissolution of cellulose. A new high modulus regenerated cellulose fibre, known by the generic name of lyocell, was manufactured based on the direct dissolution of cellulose, using the monohydrate of N-methylmorpholine-N-oxide (NMMO) organic solvent [7]. Lyocell fibre offers good mechanical properties: high modulus and tenacity.

In the present work, a biocomposite is prepared using short lyocell regenerated cellulose fibre and flax fibre (used as reference) as a reinforcement and cellulose acetate butyrate (CAB) as a biodegradable polymer matrix. Mechanical properties, dimensional stability and biodegradability of the composite have been studied for several mass compositions of fibre reinforcement. Tensile testing, soil burial test and water absorption test were performed to evaluate the biocomposite properties. In addition, fibre-matrix compatibility was studied by SEM analysis.

2. EXPERIMENTS

Cellulose acetate butyrate (CAB 419044), was supplied by Aldrich Chemical Company.

Flax fibres with a density of 1470 kg/m3 and Lyocell fibres, supplied by Lenzing AG (Austria) with a linear density of 1,7 dtex (0,17g/1000m) and an average length and diameter of 38 mm and 13,4 µm, were used as a fibre reinforcement.
Lyocell and flax fibres were ground using one 25 mm stainless-steel sphere for 2,5 g of fibre and proceeded at a frecuency of 29 Hz for 5 minutes. Particle size above 800 µm was separated by sieving for 60 minutes. The short fibres were dried in an air oven at 100ºC for 24 hours previously to the blend with CAB matrix.

Composite samples were obtained by mixing the previously ground and dried lyocell and flax reinforced fibres (10, 20, 30 and 40 % weight of fibre) with CAB matrix and consolidating the blend at at 100 kN and 210ºC for 5 min in a hot plates press forming square plates, measuring 150x150x2 mm (Collin Mod. P200E). The cooling process is carried out for 5 min under pressure using cool water until room temperature. Test samples were properly shaped according to the ASTM 412 specifications to perform tensile test mechanical measurements.
Tensile tests were carried out in a universal machine Instrom 3366. Speed of the test was set at 1 mm/min. Temperature 23 ± 2 ºC and relative humidity of 50 ± 5% was used. From load versus displacement test curves, Young's modulus, toughness, tensile strength and the percentage of breaking deformation were calculated. Four replicate samples were analyzed, and average and standard deviation were calculated for each studied property.
Water absorption of composites was tested to study the dimensional stability of the composites. Rectangular specimens (25,4 x 12,7 mm2) with 2 mm thickness were cut from mechanical testing fracture samples and dried in an air oven at 70 ºC for 24 h, cooled in a desiccator and weighed (wo). Water absorption of composites was determined by immersion of the specimens in water at 25 ºC for 24 h (ASTM D570-99). Then, excess of water on the surface of the specimens was removed before weighing (w). Four specimens were tested and average and standard deviation was reported in the results section. The percentage of water absorption (WA in %) was calculated using Eq.1:

$$WA = \frac{(w - w_o)}{w_o} x100 \qquad (1)$$

where: w_o and w represent the weight of the specimen before and after water immersion, respectively.
The soil burial test was carried out to study biodegradation of composites. The samples were incubated in contact with soil. Composites samples sheets (25,4 x 12,7 x 2 mm^3) were buried at a depth of 4 cm covering all sample sides. The samples were kept in a container at 25 ± 2 ºC with a soil moisture content of 40 ± 4 % related to dry mass for 4 weeks. After the test, samples were washed in distilled water and dried at 105 ºC in an oven for 24 h and then kept in a dessiccator. The weight of each sample was recorded before and after soil burial test. Four specimens were tested and average and standard deviation was reported in the results section. The percentage of loss weight (WS in %) was calculated using Eq.2:

$$WS = \frac{(w_s - w_{so})}{w_{so}} x100 \qquad (2)$$

where: w_{so} and w_s represent the weight of the dry specimen before and after biodegradation, respectively.

Scanning electron microscopy (SEM) was used to qualitatively examine the fracture surface of the broken samples to study the fibre-matrix compatibility. The photographs were taken in a JEOL 5610 scanning electron microscope at the accelerating voltage 10 kV and previously to the observations, the samples were coated with a 15 nm layer of gold-paladium in order to increase their conductivity.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1 Tensile properties

From the results it is observed (Figure 1) that the lyocell and flax reinforced composites have a higher modulus compared to the pure CAB at all fibre contents studied. The modulus increased from approximately 2 GPa for pure CAB to 4 GPa for a composite with a lyocell fibre content of 40%. Similar trend was obtained for referenced flax fibre reinforced CAB biocomposites. However, flax fibre based composites having 30 and 40 % by weight fibre loading, show higher modulus than lyocell based composites, with values of 5 GPa for a flax fibre composition of 40 %.

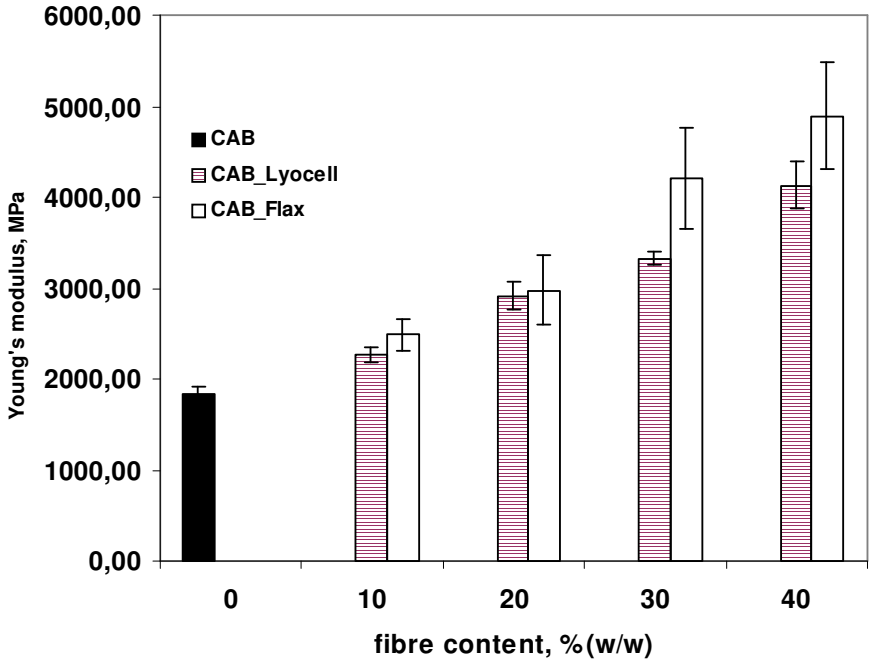

Figure 1: Young's modulus of CAB short fibre composites versus lyocell and flax fibre content.

Figure 2 describes the tensile strength at yield of lyocell reinforced CAB composites. Compared to the 100 % CAB specimen, tensile strength was not improved by the fibre reinforcement. Flax fibre based composites results in tensile strength similar to pure CAB, whereas for the lyocell based composites a decrease of the tensile strength was noticed at lyocell fibre content higher than 20%, indicating low compatibility when lyocell fibres are used as reinforcement.

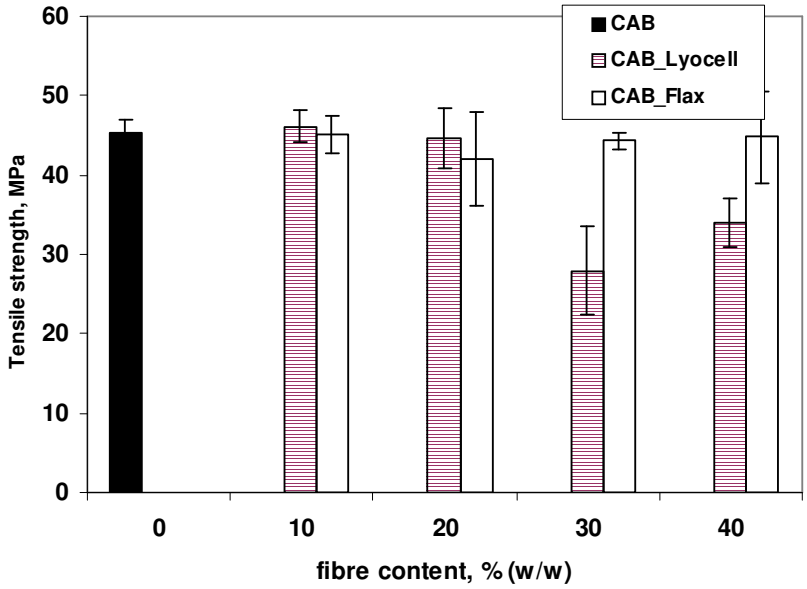

Figure 2: Tensile Strength of CAB short fibre composites versus lyocell and flax fibre content

In addition, compared to pure CAB, elongation at break and toughness decrease for all composites studied (Figure 3 and 4, respectively).

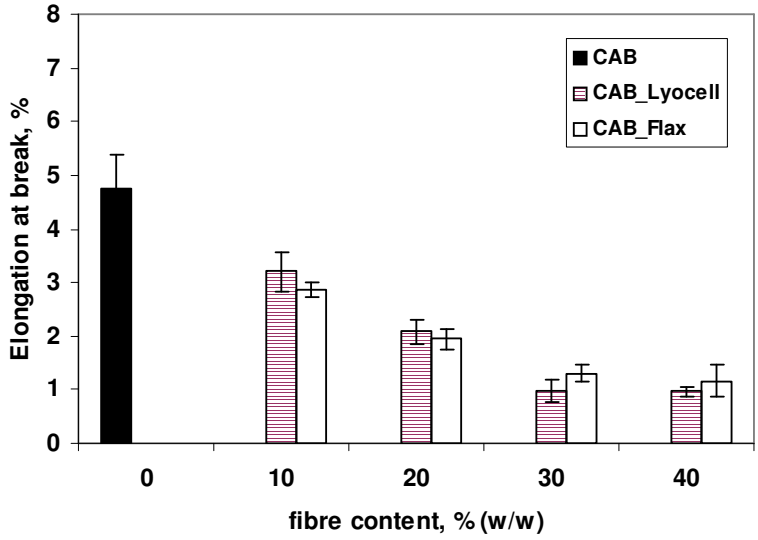

Figure 3: Elongation at break of CAB short fibre composites versus lyocell and flax fibre content.

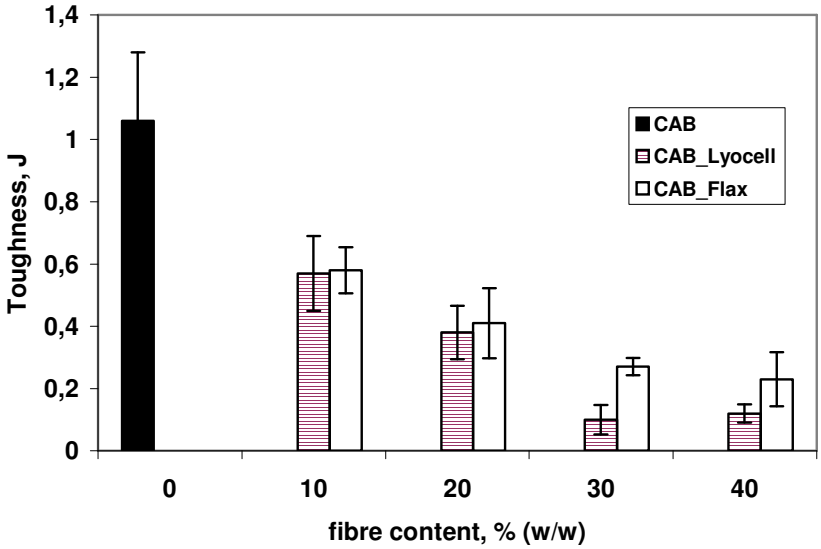

Figure 4: Toughness of CAB short fibre composites versus lyocell and flax fibre content.

4.2 Water absorption of composites

Water absorption of composites relates to composite properties such as dimensional stability. Figure 5 shows the dependence of the water absorption of composites at 25 ºC as a function of fibre content.

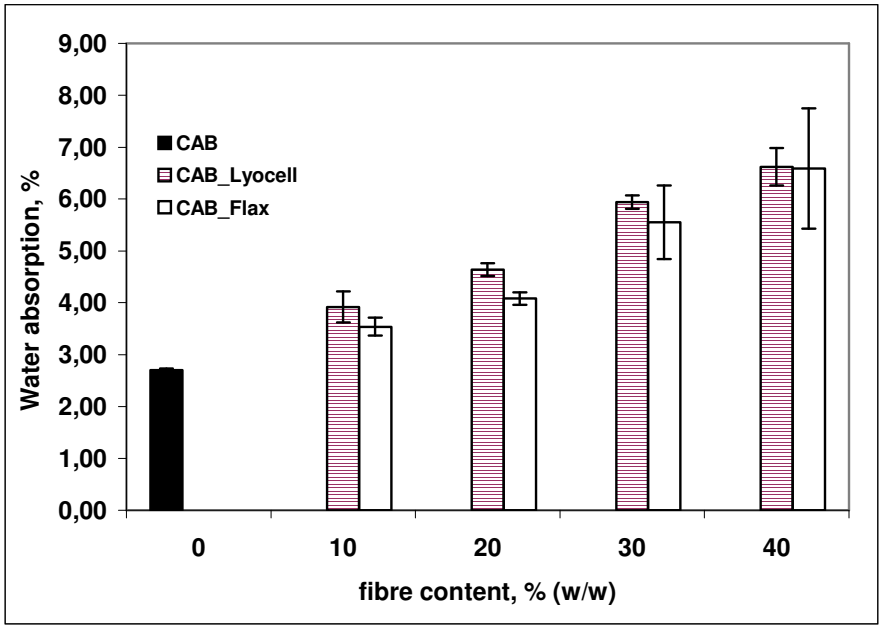

Figure 5: Water absorption of CAB short fibre composites versus lyocell and flax fibre content. Water absorption for lyocell and flax fibres themselves: 52,6 ± 3,7 % and 72,3± 0,22 %, respectively.

Fibre based composites showed significantly higher water absorption than pure HDPE due to the hydrophilic character of lyocell and flax fibres. However, compared to water absorption of lyocell and flax fibres themselves, average of 52,6 ± 3,7 % and 72,3± 0,22 %, respectively (not shown in figure 5), composites exhibit much low water absorption values. These much lower values obtained for composites than for fibres themselves is because cellulose fibres are covered by CAB layers that slow down the diffusion of water. In addition, an increase of the fibre content from 10 to 40 % results in a significant increase of water absorption of all composites. Moreover, the water absorption of lyocell fibre composites show similar behaviour but slightly higher absorption on lyocell than for flax based composites, although the absorption of water of lyocell fibres themselves is lower than that of the flax fibre. This behavior corroborated that lyocell based composites results in a more porous structure due to heterogeneities than that of the flax based composites, indicating major processing problems and the requirement of smaller fibre sizes for an adequate application in composites.

4.3 Biodegradation of composites

Figure 6 shows the dependence of the weight loss of composites after 30 days of soil degradation.

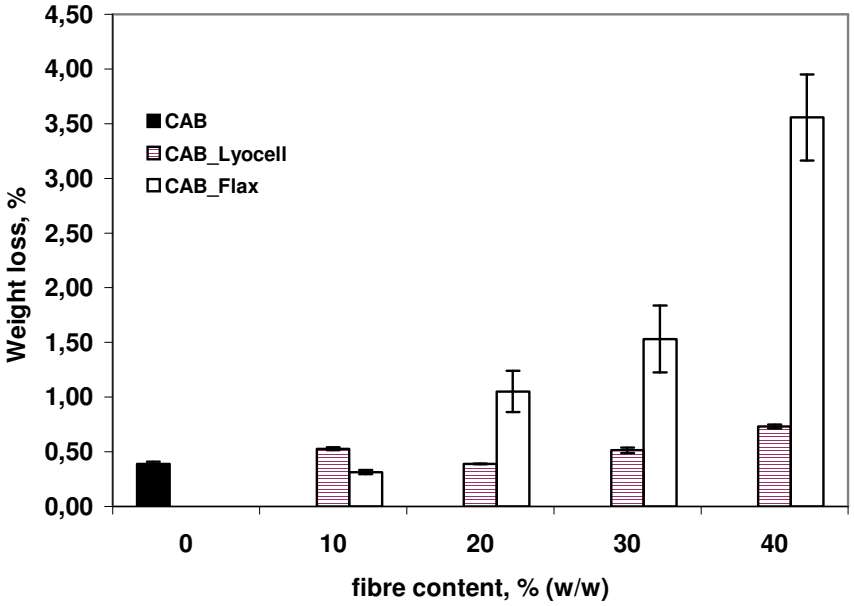

Figure 6. Weight loss of composites after 30 days of soil degradation for different fibre content composites

From biodegradation study, it was found that after 30 days of soil burial about 1% and 3,5 % weight loss was observed for 40 % fibre content of lyocell and flax fibres based CAB composites, respectively. Comparatively to pure CAB specimen, similar weight loss occurs for biodegraded lyocell composites samples. This result indicates that biodegradability at 30 days of composite seems to be controlled by the CAB layer recovering the fibres, which was caused by the hot plates pressing technique used to

prepare the composites. More studies at long degradation times are being performed currently.

4.4 Composite microstructure

Interface study of lyocell-CAB composites are carried out by SEM to evaluate microstructure of composite. Figure 7 shows the fracture surface of 20% lyocell fibre CAB composite indicanting poor fibre-matrix interfacial adhesion that leads to limited stress transfer and failure of the composites. The fibre surfaces are smooth and clean and significant fibre pullout are observed on the composites.

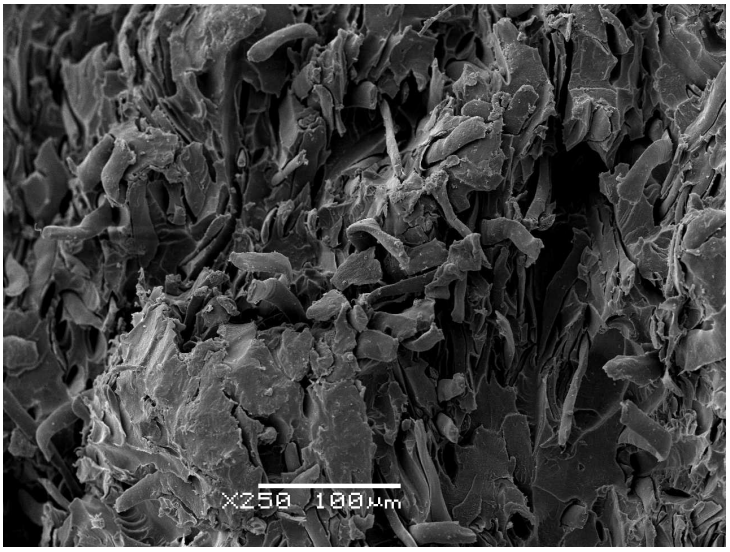

Figure 7. SEM microphotography of fracture surface of 20% lyocell based CAB composite.

5. CONCLUSIONS

From the results it is observed that lyocell/CAB reinforced composites have a higher modulus compared to the pure CAB at all fibre contents studied. Moreover, it is observed that the tensile strength at yield of composites with lyocell fibre content higher than 20% shows lower values than pure CAB. Comparatively to pure CAB, elongation at break and toughness decrease for all composites studied. This behaviour may probably due to an inadequate stress transfer between fibre and matrix which was confirmed by SEM analysis. From the dimensional stability study, it was observed that increasing the fibre content of the composite the water absorption and swelling of the lyocell fibre composites was higher than pure matrix due to the hydrophilic nature of the lyocell fibres. From biodegradation study, it was found that after 30 days of soil burial about 1% and 3,5 % weight loss was observed for 40 % fibre content of lyocell and flax fibres/CAB composites, respectively.

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