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Papers as functional green materials

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Abstract.
Paper is constituted of natural fibers and represents a perfect example of structural multifunctional materials. Indeed, its fibrous structure is engineered to fit the different end use properties: both optical and mechanical properties are usually required. These requirements may lead to contradictory needs in terms of structure. The influence of the structure on the physical properties is classically tackled based on standard methods such as the estimation of the porosity. However, this macroscopic property is not sufficient in terms of optimization of the fibrous network. For example, fluid transport has to be controlled either in the bulk of the material or only at its surface in the case of health or printing applications. Consequently, the characterization at the macro-level of the structure has to be complemented with an experimental measurement at the fiber level. The X-ray synchrotron micro-tomography, an imaging technique, is based on X-ray transmission. It allows the structure to be analyzed in 3D. It was carried in a large instrument (ESRF, France). The characterization of samples containing different recycled fibers was carried out. In particular, the influence of the number of cycles of drying-pulping is studied. Both qualitative and quantitative characterizations are obtained.
The use of recycled fibers may also be included in the elaboration of materials, taking into account the modification of the fibers in terms of morphology and mechanical properties, essentially flexibility. Mechanical properties (tensile and deformation) constitute the main examples of the analysis showing the effect of the recycling of natural fibers: the decrease in mechanical resistance of the fibrous network is explained in terms of the increase of the global porosity, essentially in the bulk of the materials. The profile of porosity in the thickness direction is found to be essential to understand the evolution of physical properties.

Introduction

Materials constituted of natural ligno-cellulosic fibers are developing nowadays due to the increasing stress on environmental aspects. The design of multi-scale materials is based on the structural properties at the fiber level. The objective is to reach multifunctional optical and mechanical macroscopic properties. A trend is to elaborate the fibrous structures using not only virgin fibers but also recycled fibers. Howard and Bichard in their work described the basic effects of recycling on macroscopic pulp properties [1]. Hubbe et al. reviewed the effect of papermaking and recycling on fiber properties [2]. However, the lack of characterization of the fibrous structure at the microscopic level impeded optimization. It is therefore necessary (i) to characterize the obtained structures at different scales, namely the micro-level and the macro-level, (ii) to evaluate the influence of the fiber properties on the physical properties of the fibrous structure.

Materials and method

Handsheets constituted of softwood pulp were successively dried and repulped, considering ten cycles. Mechanical and structural properties were evaluated following standards in controlled temperature (23°C) and humidity (50 HR) conditions [3]. The length of the sample for mechanical tests is 10 cm and its width is 15 mm. The Tensile machine was an Instron 5965. Each property was tested 10 times, if not other information is given. The morphology of the fibers was studied on a commercial equipment (MorFi).

X-ray microtomography refers to the visualization of 3D objects. Micro-tomography (µCT) concerns the 3D acquisition of samples of millimetric scale. The imaging mode used here is based on the absorption of X-Ray. The preparation of the samples does not modify the structure. Therefore, this technique is called non destructive [4, 5]. The samples are glued on a capillary, which is then fixed on the sample holder. The sample holder is placed in the measuring chamber. Both temperature and humidity are controlled. As the sample holder is rotated, an image is captured for each position. This series of projections is then filtered back projected to obtained 2D images. Finally, the 3D volume is built from the 2D data.

Results

The results concern the mechanical and the structural properties of the fibrous structures. The morphological properties of the fibers and the 3D microstructures complete the presentation of the results.

1 – Mechanical properties of the fibrous structure

The mechanical properties were first evaluated. - The evolutions of the tensile index, the deformation and the Young modulus, for the successive cycles, are presented in Figure 1a, 1b and 1c, respectively.

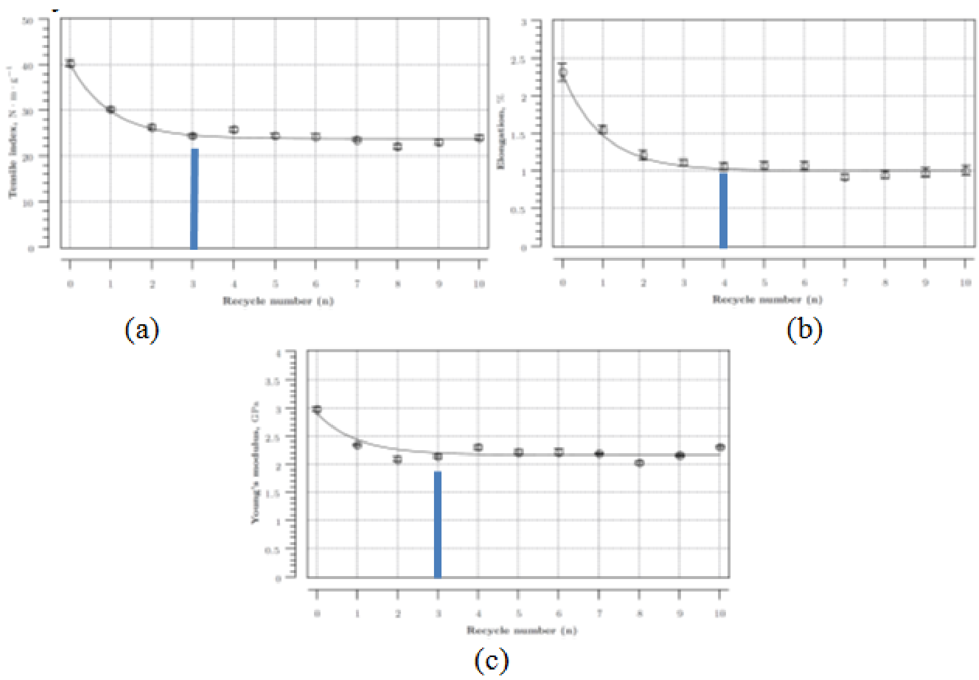

Figure 1: Tensile index (a), deformation (b), Young modulus (c) of handsheets made from successive cycles of drying

The tensile index is decreasing essentially during the first two cycles (37 %), before reaching a plateau. After an initial decrease of the deformation (56 %), a plateau is reached after four cycles. The decrease in elongation indicates brittleness of fiber network. The plateau is reached for the Young modulus after two cycles with a decrease in the initial value of 30 %.

2 – Structural properties of the fibrous structure

The structural properties were then characterized. The density of cellulose is roughly 1540 $kg.m^{-3}$ as the one for a paper varies from 300 to 1000 $kg.m^{-3}$ depending on the porosity [6]. Bulk is defined as the inverse of density: Bulk $[cm^3 . g^{-1}] = e / G$ where e [µm] and G [$g.m^{-2}$] represent the thickness and the basis weight, respectively. The evolution of the bulk vs. recycling is presented in Figure .

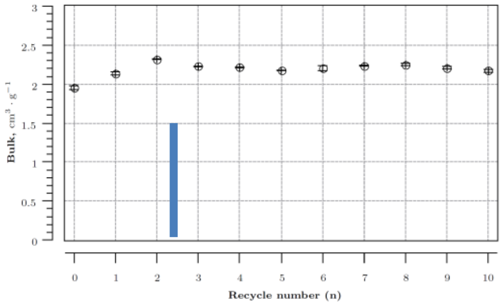

Figure 2: Bulk of handsheets made from recycled pulps

Bulk increases during the first two cycles, before reaching a plateau. In other words, the structure became more porous (15%). The results represent here are a mean of six values for three different handsheets obtained for the same level of recycling.

3 – Properties of the fibers

The evolution of the fiber morphology was characterized. Indeed, the evolution of the contact between two fibers explains the observed evolution in the different structures [7]. The results concerning the fiber widths are presented in Figure . It has to be noted that both the pulp and the fibers (eliminating the fines from the pulp) were considered. The decrease (12 %) of the width of the fibers may be observed until the fifth cycle.

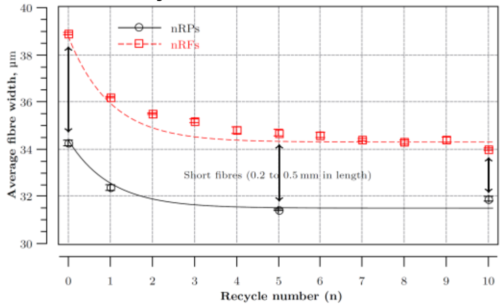

Figure 3: Average fiber width for both pulp (nRP) and fibers (nRF) vs. number of cycles

4 – 3D microstructure of the fibrous networks

The 3D characterization of the fibrous networks allows quantifying different structural properties at the fiber scale. The global porosity may be obtained as well as its evolution through the material thickness is obtained. An example, Figure , illustrates the fibrous structures that may be obtained with the µCT.

Figure 4: Microtomographs of handsheets produced from never dried (left) and ten times recycled (right) pulps. The sample size is (105 µm x 350 µm x 350 µm)

Profiles of porosity are illustrated in Figure 5.

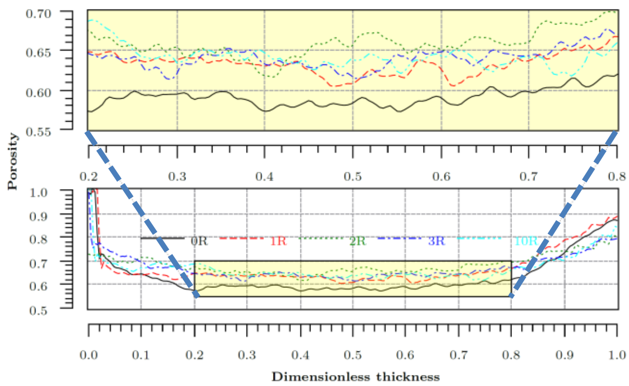

Figure 5: Porosity profiles of handsheets made from recycled pulps (N cycles)

It appears that the first cycle modifies the global porosity as well as its profile in the thickness direction.The evolution of the porosity clarifies the trends observed for both the macroscopic bulk (increase) and the mechanical properties (decrease) due to a decrease of the efficiency of the fiber bonds.

Conclusion

The effect of drying cycles on natural fibers was demonstrated considering both the mechanical and structural properties. Measurement of the fiber morphology proved that the width decreased during recycling. The X-ray synchrotron microtomography was used to quantify the evolution of the fibrous structures obtained for different number of drying cycles. The global porosity and its profile through the thickness were analyzed. For all the studied properties, a stabilization is reached after a maximum of five cycles of drying. The evolution of the microstructure explained convincingly the modification of the macroscopic properties, in terms of structure and mechanics.

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