MATRIX –FREE ALL CELLULOSE COMPOSITES

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Abstract
Matrix-free all cellulose composites are produced by combining different types of cellulosic materials with vastly different sizes to produce high performance hierarchical composites, without cellulose regeneration and, therefore, no chemical waste is produce. The matrix-free all cellulose composites are manufactured in a simple way based on paper making processes.

1. Introduction

Sustainable composites such as natural fibre composites are growing in demand due to the fact that they can be reused and/or recycled or be disposed of through composting (in the case of green natural fibre composites) or burned for energy recovery. One of the main problems in natural fibre composites is the adhesion between the fibres and the matrix, which is thought to be due to the fact that natural fibres are composites themselves (the cellulose fibrils are embedded in hemicellulose and lignin matrix) and, therefore, have different parallel and transverse linear thermal coefficients of expansion (LTCE)[1]. The difference in the LTCE makes the fibres shrink differently from the matrix, and therefore, the stress transfer is affected within the composite. These issues could be addressed by using only cellulosic materials within the composite. By doing so, the adhesion problem between the fibre and the matrix is considerably reduced.
Matrix – free all cellulose composites are produced by combining natural fibres, pulp, nanofibrillated cellulose and bacterial cellulose. Cellulosic materials of vastly different sizes are combined to utilize the space within the macro fibres and thus, to reduce the composites´ porosity and enhance its mechanical properties. The matrix-free all cellulose composites are manufactured in a simple way based on paper making processes. Cellulose hornification allows the formation of a cellulosic network which enables stress transfer between the fibres and holds them together, therefore no matrix is needed. The novelty of the matrix-free all-cellulose composites lies in the fact that no dissolution and cellulose regeneration is needed and therefore no chemical waste is produced, which is one of the drawbacks of all-cellulose composites[2]. The properties of the matrix-free all cellulose composites are characterized.

2. Experimental methods

2.1.Materials

Loose Flax fibres (25mm length) were kindly supplied by S.A.R.L. Novalin, France. Bacterial cellulose was extracted from commercially available nata de coco (CHAOKOH coconut gel in syrup, Ampol Food Processing Ltd., Nakorn Pathom, Thailand) and treated according to the method reported by Lee et al.[3] Chlorine free pulp was kindly supplied by EMPA Dübendorf. Nanofibrillated Cellulose was grinded from the above mentioned pulp by using a Matsuko grinder and after a Microfluidizer using 400µm, 200µm, 100µm, 75µm chambers as reported in literature.[4]

2.2.Composite production

The matrix free all cellulose composites are made by mixing the cellulosic fibres in an aqueous suspension and by filtering it under vacuum adapting a paper making process, as reported by Lee et al[5, 6]. The wet cake will be then hot pressed within a mould, to avoid the non- homogeneity of the composites. The composites were hot pressed under 2 tons of weight at 120°C for 2 hours. The mixing process of the cellulosic fibres and the reproducibility of the experiments will be optimized within further experiments. A main problem of natural fibre composites is the fibre distribution due to the non-homogeneity of the fibres (due to processing and agricultural factors) and the difficulty to process them in a way to achieve a compact packing, leading to porosity problems [7]. By arranging the cellulosic materials of different sizes and combining them to minimize porosity and utilize the nanofibres as matrix, we increase the density, which should lead to increased mechanical properties of the composite. As well as utilizing the different sizes of the cellulosic materials, we use different types of mixing/process to achieve composites with better mechanical properties, such as filtration per layers or the consolidations of the layers within the mould.

2.3. Characterization

The density of the flax fibres, NFC, BC, pulp and its composites was measured using Helium Pycnometry with an AccuPyc II 1340 (Micromeritics). The envelope density of the composites was measured using a GeoPyc 1360 (Micromeritics) . The weight of the samples was measured prior to both tests. In future studies we will test the mechanical properties of the composites to determine their mechanical performance. The porosity was calculated in two different ways; by measuring the envelope density of the samples measuring its apparent volume (Porosity) and by measuring the envelope density of the samples with the GeoPyc (Porosity GeoPyc).

3. Results and discussion

The matrix-free all –cellulose composites were made by adding to a constant amount of Flax fibres, a percentage of other cellulosic fibres to see its effect. Composites were made by adding 10%, 20% and 30% w/w of bacterial cellulose to Flax fibres; by adding 10%, 20% and 30% w/w of nanofibrilated cellulose to Flax fibres and by adding 10%, 20% and 30% w/w of pulp. Additional experiments were done to achieve a more homogeneous composite (especially to achieve an homogeneous thickness). Matrix- free all cellulose composites with 10% BC loading, were manufactured in two additional ways. The fibre suspension was

filtered in layers, so we could assure a good distribution of the bacterial cellulose trough the composite (10%BC layered) and then it was hot pressed within the mold. While filtering, it could be observed that every time a new layer of solution was poured into the funnel, reabsorption of the filtered fibres occurs. To avoid this issue, another type of composite was produced, 10%BC layered consolidated, which was manufactured by filtering one quarter of the solution each time and pile the four preforms together in the mold to press them.
It has to be taken into account that the hornification of the nanofibres occurs during the pressing step.

On Figure 1, a picture of a 10%BC composite can be seen and its flexural strength appreciated.

Figure 1. 10%BC composite parts (top and side view)

Since the mechanical properties are determined by the porosity of the composites, as well by its components and distribution, the porosity of the composites was calculated, as it can be seen in table 1.

Sample (matrix amount)	**Porosity [%]**	**Porosity GeoPyc [%]**
10%BC	63 ±4	66 ±1
10%BC Layered	60±3	65±2
10%BC Layered Consolidated	62±2	66±1
20%BC	60 ±2	68±1
30%BC	59 ±2	64±1
10%Pulp	65±2	78±1
20%Pulp	63±3	80±1
30%Pulp	70±3	80±3
10%NFC	59±3	58±1
20%NFC	57 ±2	59±2
30%NFC	58±5	60±1

Table 1. Porosity results of matrix-free all-cellulose composites.

As it can be seen in Table 1, the porosity of the composites seems to increase when increasing the matrix loading, but due to the non-homogeneity of the samples, it cannot be affirmed until further mechanical tests are performed.

As expected, the porosity of pulp composites is higher than for bacterial cellulose or nanofibrillated cellulose composites. This is due to the fact that pulp fibres are much larger in size and their surface area is smaller, providing less mechanical stability as matrix.

The method of manufacturing the composites seems not to have a significant effect on the porosity, but it has an effect on the thickness of the samples, as it can be seen in Figure 2, the thickness of the 10%BC Layered Composite is constant through the length of the sample, while the 10%BC composite thickness varies through it.

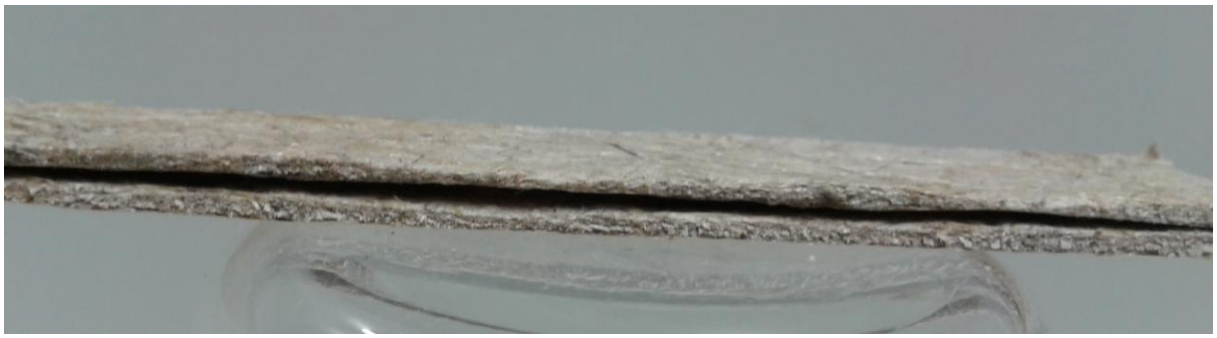

Figure 2. Thickness of 10%BC composite (top) and 10% BC Layered (bottom)

Mechanical tests, such as tensile test, were performed on some of the composites. As preliminary results, the tensile strength of the composites increased approximately by double every time the loading of BC (as it can be seen in Figure 3) was increased by 10%. The properties of pure flax preforms could not be determined due to the lack of binding within the loose fibers. The BC and NFC composites resulted in much higher tensile properties than the pulp composites, due to the better hornification of the nanocellulose fibres on the flax fibres.

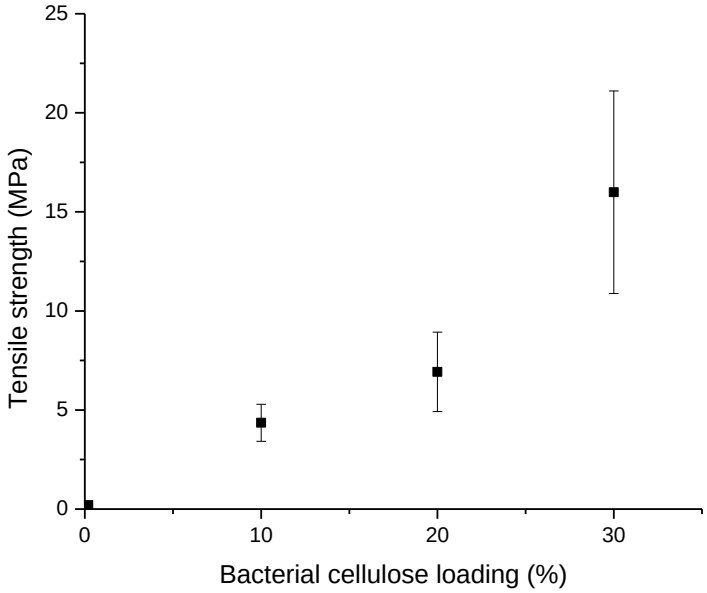

Figure 3. Preliminary tensile test results.

The composites were tried to be compacted by increasing the pressure when hot pressing, but there was no increase in the mechanical properties of these composites compare to the ones pressed under 2 Tons of weight.

4. Conclusions

Matrix-free all-cellulose composites were produced by utilizing different types of cellulosic fibres adapting a paper making process. The porosity of the different composites was calculated and compared. The tensile strength was increased nearly by double every time the loading of BC was increased by 10%. Further mechanical testing will be performed to investigate the potential of this type of composites.

5. References

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