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## Dielectric performance of new 3d printed biobased materials

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### 1. INTRODUCTION

The 3d printing market is promising and in full expansion [1]. This technology allows to accelerate rapid prototyping, which makes it more and more indispensable in the architecture, engineering and research fields [2]. Previously limited by the choice of printable materials, research is progressing, and new printable materials are available for more complex and functional products such as batteries and electronics. Biopolymers are also suitable for 3D printing field, allowing this processing technology to be the vector of innovation, eco-design and sustainable development. PLA was one of the first polymers used on a significant scale in 3d printing. Recently, new biopolymers have been added to the list of printable materials, such as PHBV and cellulose acetate (CA) [3]. Industries are still looking for the right formulation to obtain original functionalized 3d printed materials (improved stiffness, thermal conduction, tribological generator, electrical insulator).

The aim of this study is to better understand the dielectric behavior of 3d printed biopolymeric blends. We propose an original work based on the formulation of 3d printed blends of polylactic acid (PLA) and cellulose acetate (CA) and the optimization of their dielectric performances for an electrical insulating application. We focused also on the rheological properties to attest the printability of the blends. Other experiments are in progress to provide a deep knowledge and understanding of physico-chemical and mechanical performances of these 3d biobased materials.

### 2. MATERIALS AND METHODS

During this study, 2 biobased polymers are used:

- Cellulose acetate (CA) was purchased from Natureplast (FRA),
- Polylactid acid (PLA) was purchased from EURL BBFil (FRA). It is worth noting that cellulose acetate has a plasticizer content of  $W_f = 29\%$  according to the supplier.

In addition to neat biopolymers, 4 different blends were prepared and manufactured. The table below summarizes the associated formulations in weight fraction ( $W_f$ ).

	B0	B10	B20	B30	B40
$W_f$ PLA	100	90	80	70	60
$W_f$ CA	0	10	20	30	40

These blends were processed in a twin-screw extruder at 180°C and at 50 rpm. The resulting extrudates were then pelletized to a size of 4x4x4mm. A second extrusion step in a single screw extruder was carried out for a better dispersion and to obtain printable filaments. Finally, a 3D printing process was used to obtain the different samples used for physical, thermomechanical, and dielectric analysis.

### 3. MAIN RESULTS

On s'attend à des réponses pour:

- Processabilité des mélanges, paramètres optimaux, compatibilité des mélanges,
- Les mesures diélectriques, l'effet du CA sur les mesures diélectriques, la polarisation,
- Les mesures rhéologiques, l'effet du CA sur la viscosité,
- Les mesures DMA & mécaniques,

Preliminary rheological analyses showed a shear-thinning behavior of neat PLA at 180°C, with a zero-shear viscosity of 1000Pa.s<sup>-1</sup>. The addition of CA to PLA enhances the complex viscosity of the PLA:CA blend, as CA has a higher measured viscosity. However, all the blends had a viscoelastic behavior compatible with a 3D printing process and were successfully printed. The B40 specimens were the most difficult to print due to a difficulty to obtain stable printing wires. The best printing parameters were a T<sub>nozzle</sub> = 180°C, T<sub>bed</sub> = 60°C and a printing speed of 45mm.min<sup>-1</sup>. All the blends processed show a slightly compatible morphology, assessed by SEM analysis. At low CA content blends, we observed a “sea island” morphology, moving to a “co-continuous” morphology for higher CA content (40%).

The dielectric constant  $\epsilon'$  of the printed PLA and CA specimens were measured at 2.5 and 4.7 respectively for a temperature of 20°C and a frequency of 100Hz. The higher  $\epsilon'$  of CA was explained by its macromolecular chemistry obtained by FTIR. CA compared to PLA has hydroxyl and acetate functions, which enhances its polarity [1,4]. The incorporation of CA enhances the dielectric response of the blends. PLA:CA polymer blends have  $\epsilon'$  between the two components and follow a mixture law.

The electrical conductivity of the B40 shows a percolation threshold, confirming the existence of a co-continuous phase for higher CA content. CA having a higher electrical conductivity than PLA [1,5]. Maxwell–Wagner–Sillars polarization was also observed in the different blends by dielectric spectroscopy at temperatures of 150°C and measurement frequencies of 100Hz. It confirmed the existence of two phases sufficiently distinct favoring this polarization. Dielectric measurements showed that even though the incorporation of CA into PLA enhanced its permittivity, the new biomaterials produced are still fully compatible for an electrical isolation application.

Additional DMA analyses are currently in progress to determine the mechanical behaviour of all the different blends at different temperatures.

#### 4. CONCLUSION

PLA:CA blends could be successfully printed. The incorporation of CA in PLA has a dual effect. Firstly the viscoelastic properties show an enhancement of the complex viscosity was observed with the incorporation of CA in the blend. In addition, the dielectric constant and electrical conductivity enhancement increases with the proportion of CA in the blends. 3d printed PLA:CA blends are therefore perfectly compatible with electrical insulation applications.

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