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## Formulation and characterization of hydrogels and films obtained from bleached Robinia pseudoacacia L. pulp

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### PURPOSE OF THE ABSTRACT

The production of biomaterials from lignocellulosic biomass is a research field of increasing interest due to its multiple advantages, such as renewability, abundance, and low cost. In this work, two different procedures are proposed to produce hydrogels and films from bleached Kraft Robinia pseudoacacia L. pulp. To produce the first gel (HGel-1), tempo-oxidated cellulose nanofibers (TOCNF) were obtained and subjected to a microfluidization [1]. The second hydrogel (HGel-2) was manufactured through the formulation of an ionogel (EmimDMP as the ionic liquid) and further complete shifting of the ionic liquid with water (checked by measuring the conductivity of the wash water) [2]. Formulation procedures were adjusted to obtain gels with a 2 % consistency. Then, both hydrogels were employed to prepare films using a Rapid-Köthen equipment by placing the sample between two SEFAR Nitex 03-10/2 fabric meshes.

The rheological behavior of both hydrogels was determined by dynamic strain and frequency sweeps. Dynamic strain sweeps shown in Figure 1a indicated that the HGel-2 exhibited a lower limit of the linear viscoelastic region (LVR), almost a decade, than the HGel-1. Rheological spectra obtained by frequency sweeps are shown in Figure 1b. The elastic modulus,  $G'$ , slightly increased as frequency was increased, whereas  $G''$  displayed a minimum, confirming the gel-type behavior of the materials. HGel-2 showed higher moduli than HGel-1, but a lower  $G'/G''$  ratio (~10 for HGel-2 and ~15 for HGel-1).

Films produced are shown in Figure 2, and their chemical structure, mechanical, optical, and barrier properties were measured. X-Ray diffraction patterns using an X'Pert Pro MPD in an angle range between  $5^\circ$  and  $50^\circ$  showed that the TOCNF film (Film-1) has characteristic peaks of cellulose I at  $16.6^\circ$  and  $22^\circ$ , while Film-2, derived from HGel-2, only has a broaden peak at  $\sim 22^\circ$ , typical of cellulose II. Mechanical properties were evaluated using a 3345 Instron Universal Testing Machine showing similar tensile strength for both films (116-108 MPa) but higher Young's Modulus (YM) and lower elongation at break (EB) for Film-1: YM of 10.9 vs 7.5 GPa and EB of 1.3 vs 3.1% for Film-1 and Film-2, respectively. Optical properties, measured using UV-1201 Shimadzu spectrophotometer from 200 to 900 nm, exhibited that both films presented high optical transparency at a wavelength of 600 nm: 80.5 % for the Film 1 and 89.6 % for Film-2. Moreover, when testing haze, Film-1 was

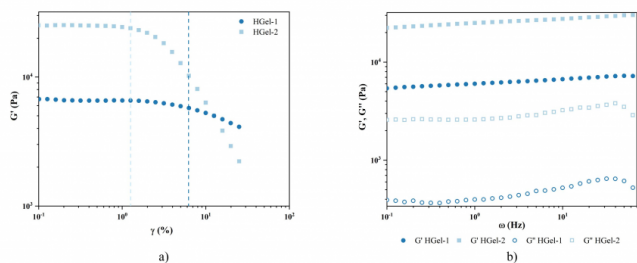
highly hazy, and Film-2 resulted in a moderated-low behavior (Figures 2c and 2d, respectively).

Finally, water vapor interactions with both films were evaluated. Adsorption/desorption isotherms were measured using a Aquadyne DVS equipment and water vapor permeability was tested following the desiccant method (ASTM E98-95 standard). Adsorption isotherms showed higher water adsorption capability for Film-1 at medium and high RH, which agrees with the presence of carboxylic groups on the surface of TOCNF nanofibers. Furthermore, water vapor transmission rate (WVTR) and water vapor permeability (WVP) were higher for Film-1 (774 g/(m<sup>2</sup>·day) and 15.6 g/(m<sup>2</sup>·day·Kpa), respectively) than for Film-2 (WVTR of 471 g/(m<sup>2</sup>·day) and WVP of 8.5 g/(m<sup>2</sup>·day·Kpa)). Thus, Film-2 showed better barrier properties.

In conclusion, the use of TEMPO-ox. followed by microfluidization and the formulation through the intermediate production of an ionogel are both valid strategies to obtain hydrogels and their subsequent films, enhancing the tenability of the manufactured material depending on the process employed. Optical and mechanical properties suggest the interest of TOCNF film (Film-1) for its application in photovoltaic cells, whereas optical and barrier properties indicate that ionogels/hydrogel-derived films (Film-2) are good candidate for its use in packaging.

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## FIGURES



**FIGURE 1**

Figure 1.

(a) Dynamic Strain sweep (yield points showed with a dash line) and (b) dynamic frequency sweep of the 2 wt.% cellulose hydrogels



**FIGURE 2**

Figure 2.

(a) Film 1 and (b) Film 2 over a sheet of paper; (c) Film 1 and (d) Film 2 at 5cm from a sheet of paper.

## KEYWORDS

Cellulose | Hydrogel | Film | Bleached pulp

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