Electrorheology as a Support Tool for Understanding the Electrotribological Behavior of Nanocellulose-Based Biolubricants

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Abstract. This research aims to explore the behavior of sustainable electrorheological lubricant fluids. Rheological data allows for the analysis of the strength of a chain-like structuration stimulated by an electric field, as well as the angular orientation of nanocellulose particles' suspensions. Commercial fibrillated (CNF) and crystalline (CNC) nanocelluloses were dispersed in castor oil at concentrations of 1 and 4 wt.%. Oscillatory flow tests were performed within the linear viscoelastic range, following a pre-shear step, and under the effect of an electric field. The consequent structuration of nanocellulose particles were visualized using a microscope attached to the rheometer. Squeeze flow test and tribological analysis were also performed. Storage and loss moduli were found to increase with increasing values of the electric field, and high pre-shear caused structures to break and form new chains, reducing storage and loss moduli at low values of the electric field. Regarding the squeeze flow tests, dispersions were found to exert an opposite force to compression which grew up with increasing values of electric field and concentration. Finally, friction coefficient values were found to be electro-responsive while wear was found to decrease when applying an electric potential.

Keywords: electrorheology · nanocellulose · biolubricant

1 Introduction

Growing emphasis in the search of environmentally aware formulations entails a remarkable source of research nowadays. Alongside, smart materials have also attracted great interest and there exist numerous references of innovation. In general, these can be described as materials which respond or adapt to a specific external stimulus. Among these, electro-rheological fluids are of special relevance in the fields of composites, clutches, valves, dampers, shock absorbers or lubricants. Upon activation of an electric potential, particles align yielding chain-like structures parallel to the direction of the electric field and apparent viscosity of these ER suspensions can be increased by several orders of magnitude [1]. In relation to lubricity, previous studies have pointed out that the use of electrorheological fluids may affect both friction and wear [2, 3].

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We herein present pioneering research on the electrorheological behavior of sustainable electro-active lubricating fluids, which is intended to assist in improving comprehension of their friction and wear performance when used in electrified tribological contacts.

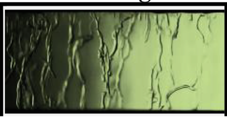
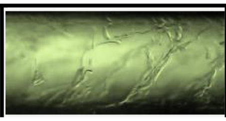
2 Methodology

An ARES G2 (TA Instruments, USA) controlled stress rheometer featured with a portable optical microscope allowed for the analysis of both strength and angular orientation of the chain-like structures formed by the nanocellulose particles subjected to an external electric field [4]. Commercially available fibrillated (CNF) and crystalline (CNC) nanocelluloses were dispersed in castor oil at concentrations of 1 and 4 wt.%. Small amplitude oscillatory shear (SAOS) tests were performed within the linear viscoelastic (LVE) range. Targeted analysis on the balance between dielectrophoretic and hydrodynamic forces affecting the fluids was conducted by pre-shearing (PS) at 0.1 s^{-1} and 30 s^{-1} before the oscillatory shear tests took place, under electric fields of 0.16, 0.8, 2.4 and 4 kV/mm. Squeeze flow tests under an electric potential of 300 V were performed at constant compression rate of $5 \cdot 10^{-4}$ mm/s and starting at a gap distance of 0.5 mm until reaching an opposing force of 20 N. Finally, the tribological performance was assessed by using a ball on three plates tribology cell coupled to a MCR 501 (Anton Paar, Austria) rheometer.

3 Results and Discussion

As can be appreciated in Table 1, the optical analysis revealed that at low pre-shear, the dielectrophoretic forces exerted a significant influence on the polarized nanocelluloses, such that the particles aligned forming chain-like structures within the electric field direction. However, a high pre-shear broke these chain-like structures and polarized nanocellulose particles aligned parallel to the flow direction, to such an extent that they suddenly re-arranged into new chains when halted the pre-shear. Regarding nanocellulose concentrations, at 1 wt%, thinner CNC-based chains were observed, which angular orientation (due to electric field-induced polarization) increased with the electric field at low pre-shear. At 4 wt.%, an entangled network was perceived, and nanofiber orientation was completely restrained.

Table 1. Different structuration of CNC particles under the influence of an electric field of 0.8 kV/mm, after a pre-shear of 0.1 s^{-1} (low PS), and 30 s^{-1} (high PS).

1 wt.% Low PS	1 wt.% High PS	4 wt.% Low PS	4 wt.% High PS
			

Regarding the SAOS test, the highest pre-shear rate carried out weaker column structures within the electric field direction, which caused a reduction in both storage and loss moduli (Fig. 1). These results reveal the competition between hydrodynamic (flow) and dielectrophoretic forces (polarization and structuring) on both electric field induced structuring and the rheological behavior of nanocellulose-based dispersions**.** This competition was more noticed at the lowest values of the electric field (e.g. below 0.8 kV/mm), when comparing low and high pre-shear. This was observed at both 1 and 4 wt.% concentrations.

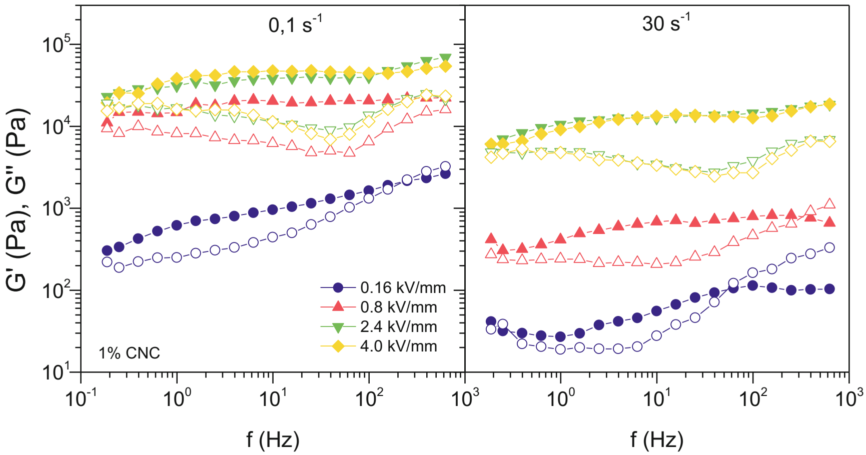


Fig. 1. Storage (filled symbols) and loss (empty symbols) moduli of SAOS tests after a pre-shear of 0.1 s^{-1} (left) or 30 s^{-1} (right) of CNC-based dispersions at 1 wt.%

In order to evaluate the strength of the electric-field induced microstructure of the nanocellulose-based dispersions studied, squeeze flow tests were also performed. In these compression tests at constant rate, the monitored normal force was found to increase at larger plate-to-plate distances under the effect of an electric field, and progressively with increasing values of concentration. Moreover, it was found by optical microscopy that polarized nanoparticles were retained within the gap between upper and lower electrified plates. This fact was found to enhance the load supporting capacity of these lubricants.

Finally, the tribological behavior was assessed under full sliding conditions in an electrified ball-on-three plates tribology cell. Friction coefficient values were found to be electro-dependent and, wear was also found to decrease when applying an electric potential. Therefore, it is worth pointing out that an electro-active control of lubrication may be possible with nanocellulose-based biolubricants.

4 Conclusions

Electrorheological behavior of the formed structures inside the nanocellulose-based biolubricants matrix was analyzed. Storage and loss moduli were found to increase with increasing values of the electric field, although they were significantly affected by the pre-shear. Different structuring of the nanocellulose particles was observed when low or high pre-shear was applied, due to the competition between hydrodynamic (flow) and dielectrophoretic forces (polarization) on electric field-induced structuring. Thus, the high pre-shear reduced both the storage and loss moduli, mainly at low values of the electric field. Furthermore, applying an electric field leaded to the accumulation of polarized nanocellulose particles between electrified plates, which exerted an increasing opposing force to compression with increasing values of electric field and concentration. Finally, friction coefficient values were found to be electro-responsive, while wear was found to decrease when applying an electric potential.

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