

## Anisotropic Cellulose-Based Films for Humidity-Driven Soft Actuation and Water Harvesting

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Moisture-responsive mechanisms are widespread in nature, particularly in plants exhibiting hygroscopic motion. Inspired by these systems, recent research has focused on designing materials that replicate humidity-driven movements for various applications [1]. Liquid crystalline cellulose-based films can reversibly deform in response to changes in humidity, enabling anisotropic movements similar to those observed in *Erodium* awns [2]. In this study, hydroxypropyl cellulose (HPC) films obtained from 60% (w/w) aqueous solutions (within the liquid-crystalline concentration regime) [3] were crosslinked with citric acid (CA) and processed by shear casting followed by thermal treatment. Eight samples were produced: two pure HPC films, dried at room temperature and at 150 °C, respectively, and six crosslinked films with variable CA content, all treated at 150 °C. Structural and mechanical characterizations using SALS, POM, SEM, and tensile testing revealed that temperature and CA concentration strongly influence the films' anisotropy. Optimal anisotropic properties were observed at 7.5 wt. % CA, where POM also identified topological defects, indicating a phase transition from nematic to a cholesteric ordering. Upon exposure to moisture, the films exhibited reversible helicoidal twisting closely resembling the motion of *Erodium* awn. These findings demonstrate that citric acid crosslinking effectively modulates the hierarchical structure and moisture responsiveness, paving the way for their application as humidity sensors and water-harvesting materials.

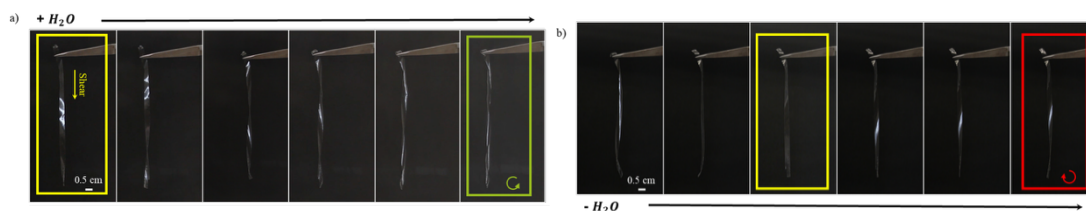


Figure 1. Reversible helicoidal actuation-controlled humidity: a) twisting of the film from a flat configuration to a fully coiled right-handed helical configuration and b) reverse motion where the film passes through its original flat configuration and adopts a left-handed helical configuration.

### References

- [1] F. Sousa *et al.*, *Adv. Mater. Technol.*, vol. e01426, pp. 1–17, 2025.
- [2] A. P. C. Almeida *et al.*, *Soft Matter*, vol. 15, no. 13, pp. 2838–2847, 2019.
- [3] M. H. Godinho *et al.*, *Liq. Cryst.*, vol. 44, no. 12–13, pp. 2108–2120, 2017.

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