

## Drying-Induced Buckling and Liquid Crystalline Ordering in Cellulose Nanocrystal Microdroplets

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Confinement and non-equilibrium processing play a central role in the self-assembly of colloidal liquid crystals, where mechanical stresses, mass transport, and orientational ordering evolve in a strongly coupled way [1], [2]. In this contribution, we investigate drying-induced deformation and structural evolution in cellulose nanocrystal (CNC) microdroplets produced by microfluidics, with particular emphasis on how drying under confinement affects liquid-crystalline organization.

We focus on two experimentally controllable parameters: the aggregation state of the CNC suspension prior to droplet formation and the surfactant concentration in the surrounding oil phase, which regulates water transport during drying [3]. By systematically varying these factors, we examine how drying kinetics, shell formation, buckling, and optical response develop as droplets dehydrate under confinement (Figure 1). Polarized optical microscopy and rheological measurements are used to connect the internal suspension state to droplet-scale deformation and the evolution of anisotropic order. Our observations show that drying proceeds through mechanically driven instabilities rather than simple isotropic shrinkage, and that the character of these instabilities depends sensitively on both the initial dispersion quality and the external mass-transport conditions. Even after substantial size reduction, droplets remain partially hydrated, indicating that mechanical confinement, evaporation dynamics, and liquid-crystalline ordering remain coupled throughout the drying process. Importantly, we find that the final optical appearance of the dried droplets can be tuned through the surfactant concentration: increasing surfactant concentration leads to a stronger blue coloration in the dried state, as confirmed by quantitative color analysis.

Overall, this work shows how drying-induced buckling influences liquid-crystalline organization in confined CNC systems and provides insight into controlling stress, confinement, ordering pathways, and final optical properties in colloidal liquid crystals.

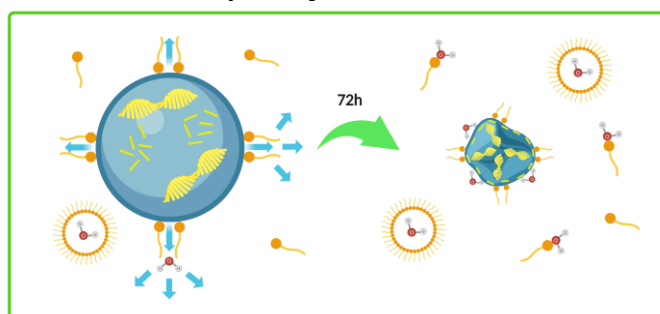


Figure 1. Illustration of the drying process in which a CNC droplet is dried in the presence of surfactant. The resulting dried droplet is later characterized based on surface morphology, internal chiral nematic structure, and structural color.

### References

- [1] M.S. Tirumkudulu, *Soft Matter* **14**, 7455-7461 (2018).
- [2] R.M. Parker, B. Frka-Petesic, G. Guidetti *et al*, *ACS Nano* **10**, 8443-8449 (2016).
- [3] I.R. Sarbandi, J.O. Zoppe, Aggregation- and Surfactant-Controlled Buckling and Structural Color in Drying Cellulose Nanocrystal Microdroplets, *submitted* (2026)

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