**Supporting Information**

# Compound Droplets Derived from a Cholesteric Suspension of Cellulose Nanocrystals

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Fig. S1 shows the schematic of the MF device with dimensions of the microchannels and an optical microscopy image of the compound droplets formed by MF emulsification.

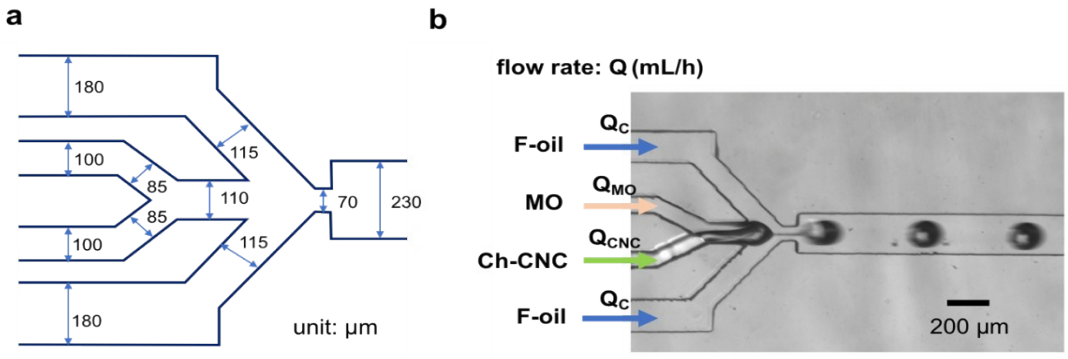



**Figure S1.** (a) Schematic of the MF device for the generation of compound droplets. The height of microchannels is 180 μm. (b) Optical microscopy image of the preparation of Ch-CNC/MO compound droplets.

Fig. S2 shows the variation in volume fractions of the MO and Ch-CNC components in the compound droplets that were prepared by MF emulsification at varying ratios of flow rates, of the Ch-CNC and MO phases $\alpha=Q_{Ch\text{-}CNC}/Q_{MO}$ and constant $\beta=Q_d/Q_c=0.1$ mL/h).

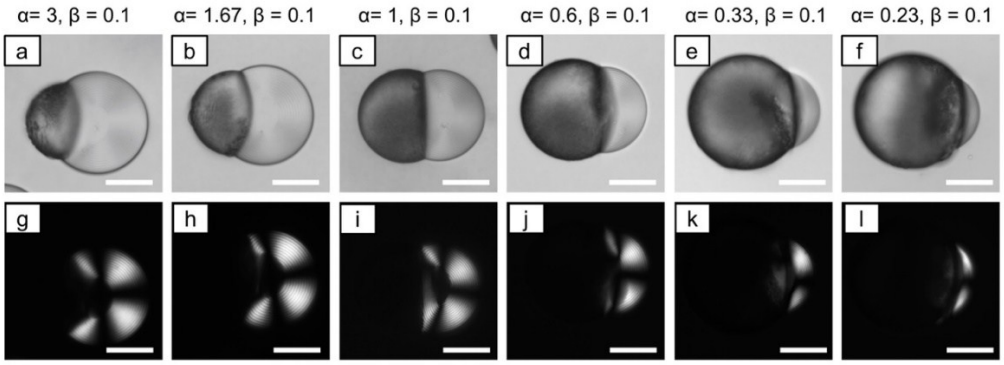



**Figure S2.** Variation in volume fractions of the MO and Ch-CNC phases of the droplets. BF (a–e) and corresponding POM (f-j) images of the Ch-CNC/MO compound droplets prepared by MF emulsification at varying α and β = 0.1 mL/h. The images were taken after 13 days from the emulsification. Scale bars are 50 μm.

Fig. S3 illustrates the formation of small droplets of the Ch-CNC suspension at the interface between the MO and Ch-CNC phases immediately after MF emulsification and after 20 day droplet equilibration. With equilibration time, small droplets diffuse to the interior of the MO phase.

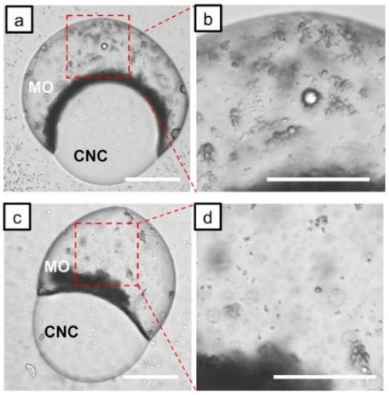
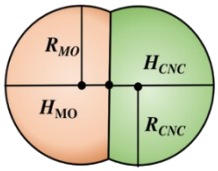



**Figure S3**. Formation of small Ch-CNC droplets at the Ch-CNC/MO interface

(in the MO phase). BF images of the droplet immediately after emulsification (a, b) and after 20 day equilibration (c, d), imaged at low (a, c) and high (b, d) magnifications. Scale bars are 100 μm.

Fig. S4 shows the calculation of the volumes of the MO and Ch-CNC phases in the compound droplets, along with notations used for these calculations.




$$V_{CNC} = \frac{\pi}{3} H_{CNC}(3R_{CNC}H_{CNC} - H_{CNC}^2)$$

$$V_{MO} = \frac{\pi}{3} H_{MO}(3R_{MO}H_{MO} - H_{MO}^2)$$

$$\frac{V_{CNC}}{V_{MO}} = \frac{H_{CNC}^2(R_{CNC} - 2H_{CNC})}{H_{MO}^2(R_{MO} - 2H_{MO})}$$

**Fig. S4.** Calculation of the volumes of the MO and Ch-CNC phases ($V_{MO}$ and $V_{Ch\text{-}CNC}$, respectively) in the compound droplets.

Fig. S5 shows the effect of the ratio of flow rates of the droplet-to-continuous phase, $\beta = Q_d/Q_c$ on the droplet size and morphology.

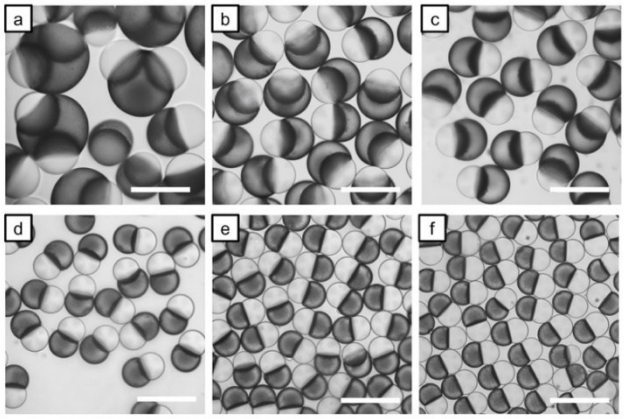



**Fig. S5.** Effect of the ratio of flow rates of the droplet-to-continuous phase on droplet size and morphology. BF images of Ch-CNC/MO droplets prepared at α=1.0 and β of 0.8 (a), 0.4 (b),

0.2 (c), 0.1 (d), 0.08 (e), and 0.05 (f). All images were taken immediately after emulsification. Scale bars are 200 μm

Fig. S6 shows the variation in droplet morphology with an increasing concentration of the surfactant Tween 80 added to the Ch-CNC phase.

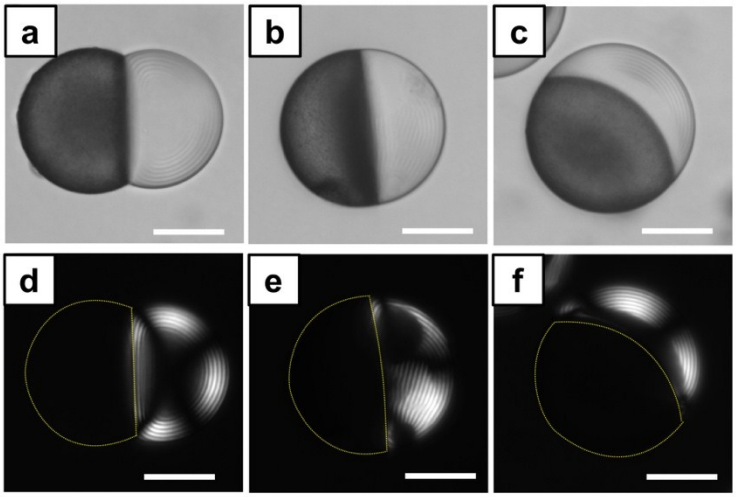



**Fig. S6.** Variation of morphology of the droplets with increasing concentration of Tween 80 in the Ch-CNC phase. BF and corresponding POM images of the Ch-CNC/MO droplets with $C_{Tw}$ of $1.3 \times 10^{-3}$ wt. % (a), (b) $1.3 \times 10^{-2}$ wt.% and 0.26 wt. % (c). The images were taken 1 day after emulsification. Scale bars are 50 μm**.**

Fig. S7 shows the change in the shape of compound droplets following their equilibration for various time intervals after MF emulsification.

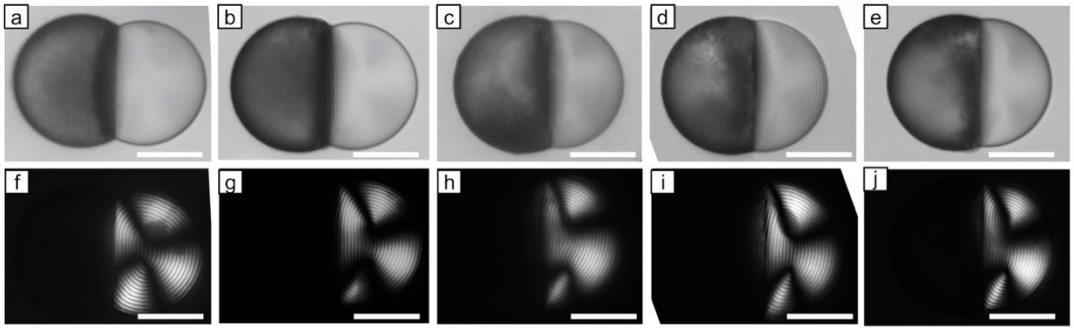



**Fig. S7.** Variation in the morphology of the droplets with time. BF (a–e) and corresponding POM (f-j) images of the Ch-CNC droplets prepared at $\alpha = 1.0$ and $\beta = 0.1$ and imaged

immediately after emulsification (a), and 3 (b), (c) 6 (c), 10 (d), and 19 (e) days after emulsification.

Fig. S8 shows compound droplets containing a monomer mixture in the Ch-CNC phase.

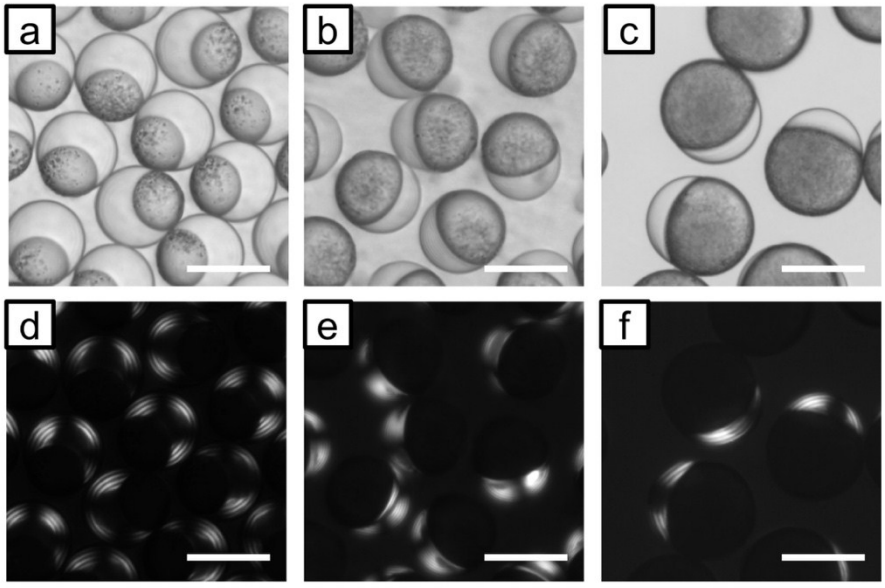



**Fig. S8.** Compound droplets with a monomer mixture introduced in the Ch-CNC phase. BF (a–c) and corresponding POM (d-f) images of the Ch-CNC/MO droplets containing a monomer mixture and prepared at β=0.1 and α of 3.0 (a, d) 0.8 (b, e) and 0.33 (c, f). All images were taken after 6 days after emulsification. Scale bars are 100 μm.