

Supporting Information

Stabilizing Aqueous 3D Printed Constructs Using Chitosan-Cellulose Nanocrystal Assemblies

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Video contents

Video S1. A droplet of 0.5 wt.% chitosan in dextran aqueous solution ($\text{pH} = 3.5$) was injected into the 1 wt.% CNC in PEG aqueous solution ($\text{pH} = 2.5$). When the volume of the droplet was decreased, wrinkles appeared immediately, and as the volume continuously decreased, the number of wrinkles increased. When the volume of the droplet was increased again, all vestiges of the folds vanished.

Video S2. The high-speed images of a dextran aqueous solution without chitosan falling in CNC/PEG aqueous solution at an injection speed of 1.5 ml/min. The concentrations of CNCs in PEG aqueous solution is 1 wt.%. The pH of the PEG aqueous solution and dextran phase are 2.5 and 5.0.

Video S3. The high-speed images of a chitosan/dextran aqueous solution falling in PEG aqueous solution without CNCs at an injection speed of 1.5 ml/min. The concentrations of chitosan in dextran aqueous solution is 0.5 wt.%. The pH of the dextran aqueous and PEG aqueous solution are 3.5 and 3.5.

Video S4. The high-speed images of a chitosan/dextran aqueous solution falling in CNC/PEG aqueous solution at an injection speed of 0.05 ml/min. The concentrations of CNCs in PEG and chitosan in dextran aqueous solution are 1 wt.% and 0.5 wt.%. The pH of the PEG aqueous and dextran aqueous solution are 2.5 and 3.5.

Video S5. The high-speed images of a chitosan/dextran aqueous solution falling in CNC/PEG aqueous solution at an injection speed of 1.5 ml/min. The concentrations of CNCs in PEG and chitosan in dextran aqueous solution are 1 wt.% and 0.5 wt.%. The pH of the PEG aqueous and dextran aqueous solution are 2.5 and 3.5.

Video S6. The formed liquid tubules in **Video S5** rest at the base of the cell and do not coalesce.

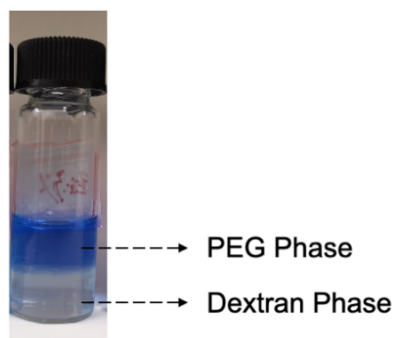


Figure S1. Phase separation of 15 wt.% PEG and 20 wt.% dextran aqueous two-phase system.

The concentration of Nile blue A is 0.03 mg/ml.

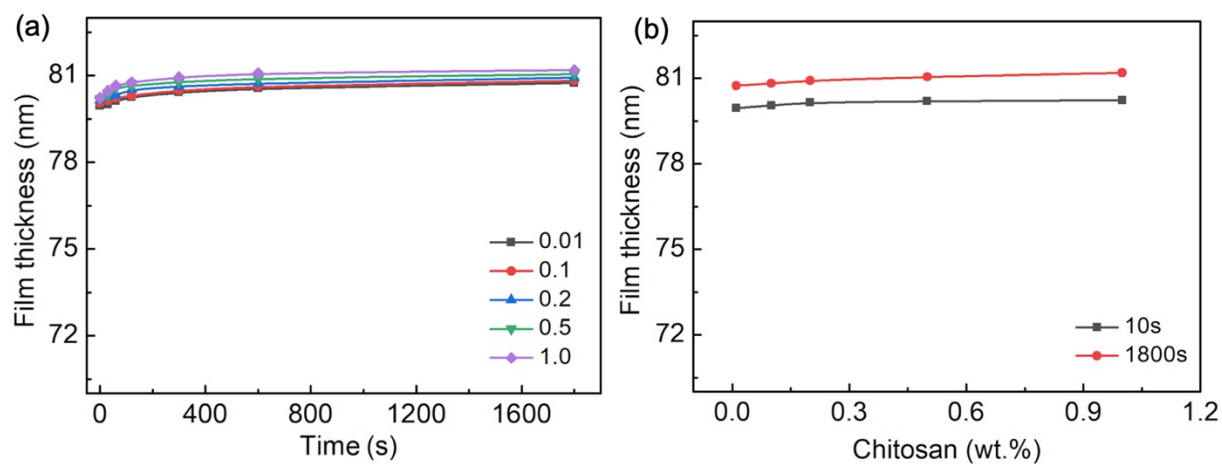


Figure S2. (a) With different chitosan concentration, film thickness as a function of assembly time. (b) At assembly time of 10 s and 1800 s, film thickness as a function of chitosan concentration. The concentration of CNC in PEG aqueous phase is 1 wt.% and the pH of PEG phase is 2.5.

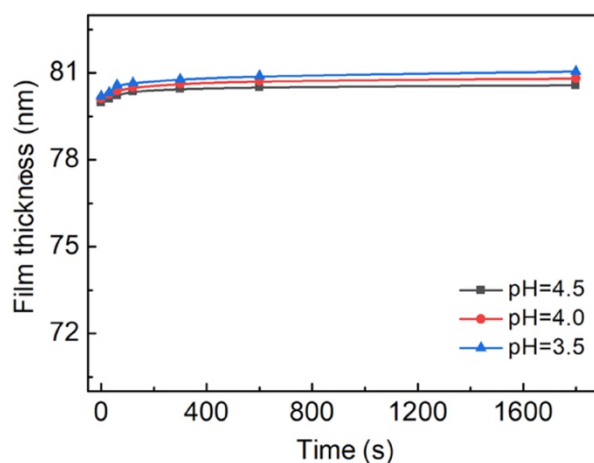


Figure S3. At different pH of the dextran solution, film thickness as a function of assembly time. The concentrations of CNCs in PEG and chitosan in dextran aqueous solution are 1 wt.% and 0.5 wt.%.

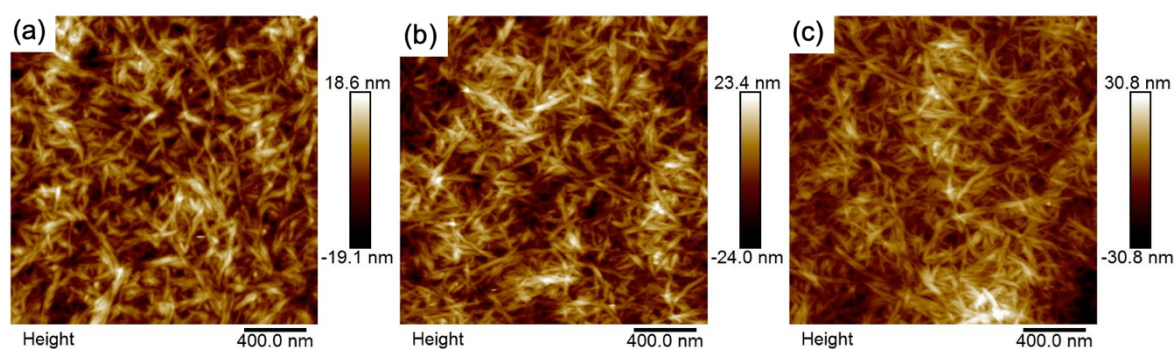


Figure S4. AFM height images of the CNC/chitosan assembly film formed at water-water interface. (a), (b), and (c) are height images of the generated films at the chitosan concentration of 0.1 wt.%, 0.5 wt.% and 1.5 wt.%. The concentration of CNC is 1 wt.% and pH of PEG solution is 2.5.

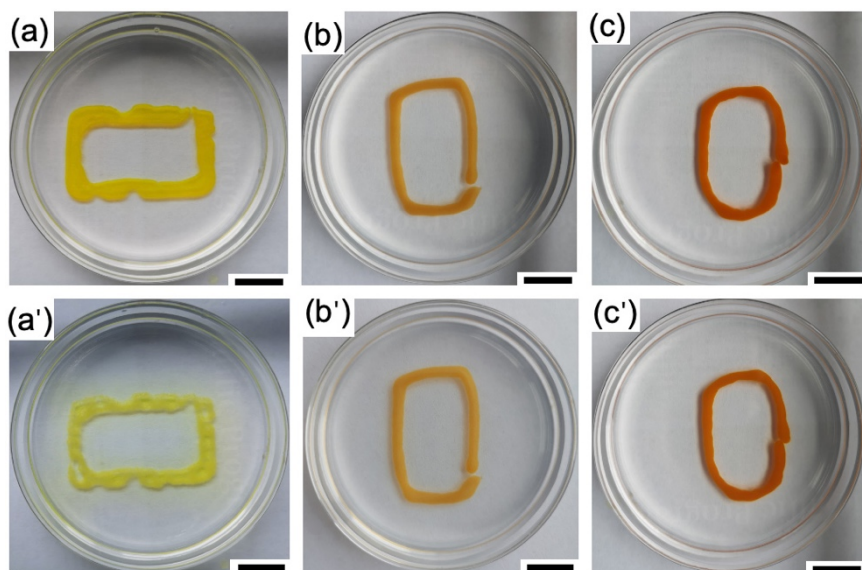


Figure S5. The effect of chitosan concentration on the shape and stability of the printed structures. At a chitosan concentration of (a) 0.1wt.%, (b) 0.5 wt.%, and (c) 1 wt.%, the shape of printed structure after printing for 1 min (a, b, c) and 30 min (a', b', c'). The concentration of CNC is 1 wt.% and the pH of PEG solution is 2.5. The scale bar is 1 cm. Fluorescein sodium with a concentration of 1.2 mg/ml in dextran solution was used to increase the image contrast.



Figure S6. The printed spiral structure after printing for 1 h. The concentrations of CNC and chitosan are 1 wt.% and 0.5 wt.%. The pH of the dextran and PEG aqueous solution are 3.5 and 2.5. The scale bar is 1 cm. Fluorescein sodium with a concentration of 1.2 mg/ml in dextran solution was used to increase the image contrast.