Supporting Information

All-aqueous Electrosprayed-Emulsion for Templated Fabrication of Cytocompatible Microcapsules

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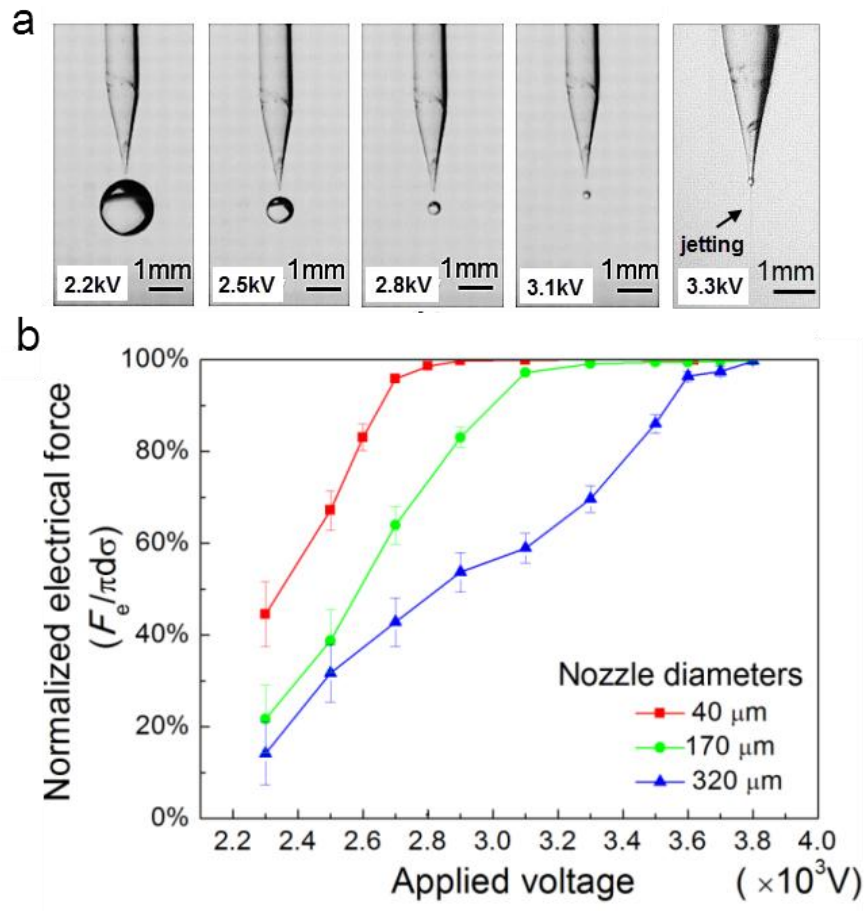

Figure S1. (a) When the applied voltage increases from 2.2 kV to 3.4 kV, breakup of the emulsion phase undergoes a transition from the dripping to the electrical-jetting mode. In the dripping regime, monodisperse droplets pinch off at the end of the capillary nozzle. In the jetting mode, a string appears and the resultant droplets are non-uniform in diameters. The nozzle diameter is 40 μm. The flow rate of the emulsion phase is 0.5 ml/h. (b) In the electrical dripping regime, the gravitational and electrical forces balance with the surface tension force. With increased applied voltages, the electrical force also increases. The transition from dripping to jetting mode starts when the applied electrical force is almost equal to the surface tension force.

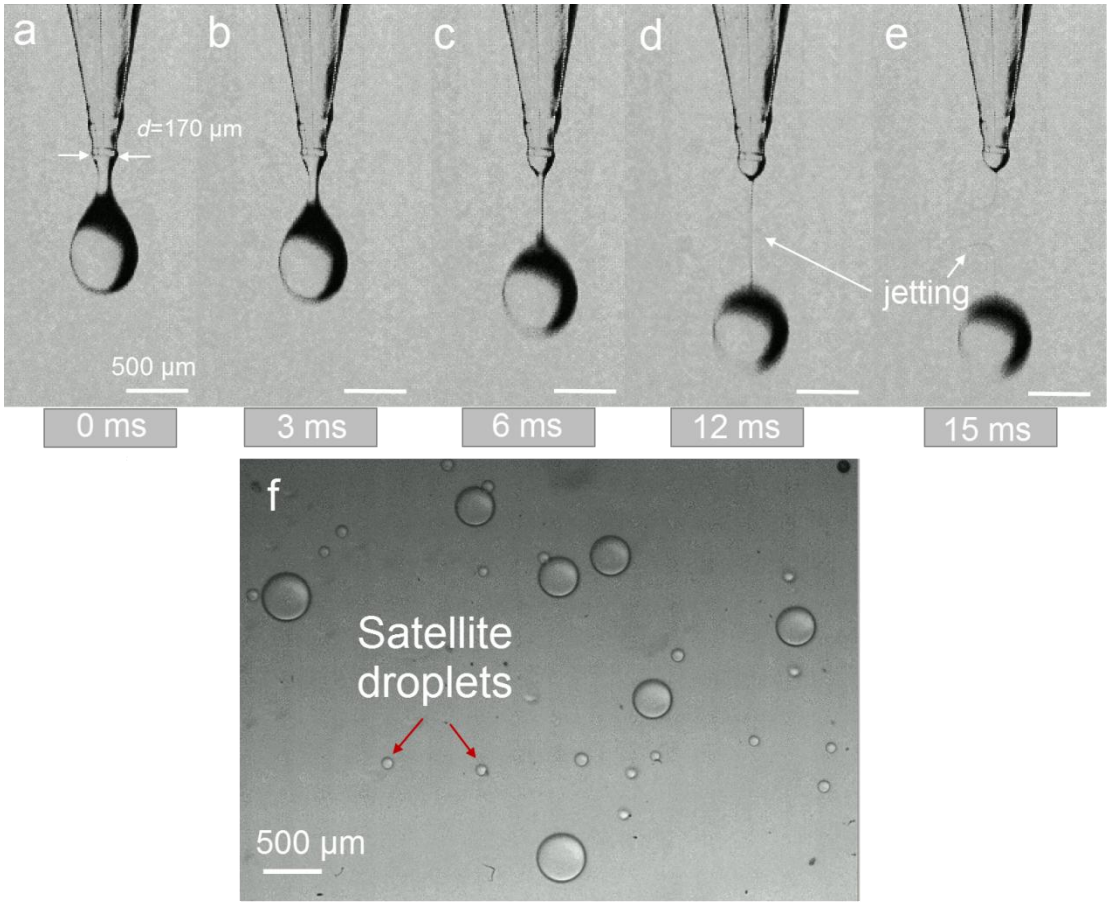

Figure S2. (a-e) Optical microscope images showing the formation of droplets in the jetting regime during the all-aqueous electrospray. The nozzle diameter is 170 µm. Scale bar is 500 µm. (f) Breakup of the jet results in polydisperse w/w emulsion droplets. Satellite droplets are indicated by the red arrows. The applied voltage is 3.9 kV.

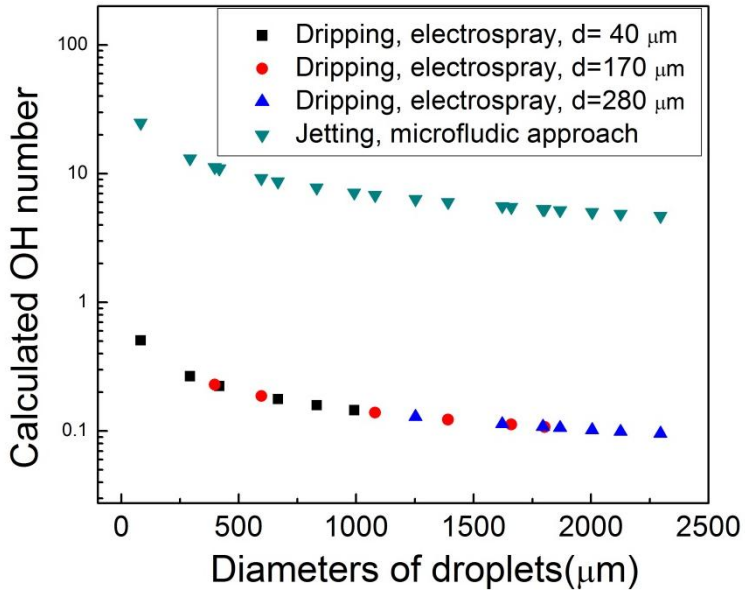

Figure S3. A plot showing the calculated Oh number as a function of droplet diameter during the electrospray. The viscosity and density of the emulsion phase (a 15 wt% dextran solution) are 40 mPa·S and 1.06 g/cm^3 respectively. The surface tension σ is around 70 mN/m. As a comparison, when the droplet-based microfluidic technique is used to generate droplets with the same diameters, the calculated Oh numbers range from 5 to 20. The interfacial tension between the dextran-rich and PEG-rich phases is about 0.03mN/m [24].

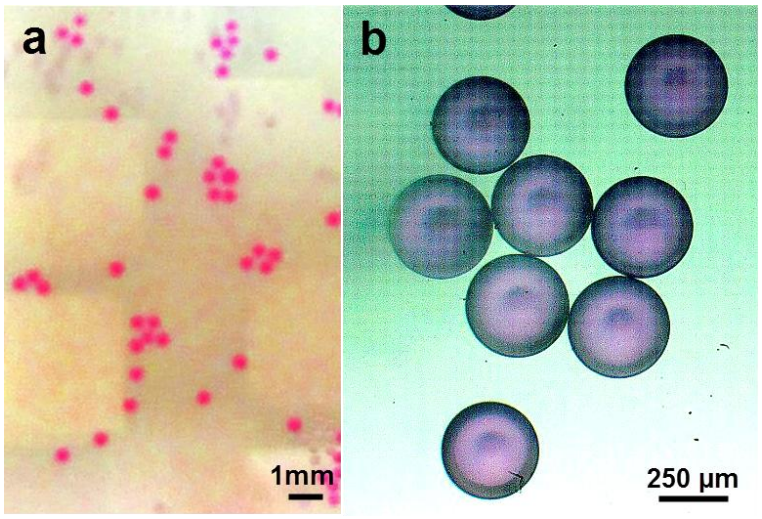

Figure S4. All-aqueous electrosprayed fabrication of PEGDA-in-K_3PO_4 emulsions. (a) A photograph and (b) an optical microscope image of PEGDA-in-K_3PO_4 emulsion droplets. The PEGDA-rich emulsion phase is stained by 0.1wt% Nile Red.

We dissolved 30% K_3PO_4 and 10% PEGDA (M_w=700) in water, forming two immiscible aqueous phases. This emulsion mixture separates into a PEGDA-rich top phase and a K_3PO_4-rich bottom phase after centrifuged at 6000 rpm for 3 minutes. (Trace amount of PEGDA droplets mixed in the K_3PO_4-rich bottom phase can be fully removed by UV-crosslinking and subsequent filtration.) The PEGDA-rich and K_3PO_4-rich phases are separately extracted from the top and bottom of the container, as the emulsion and continuous phases to be used in the all-aqueous electrospray. The electrospray setup is the same as that mentioned in figure 1. Distance between the nozzle and the collecting culture dish is 40 cm. The applied voltage is 2.4 kV. Diameter of the capillary nozzle is 50 μm. A metallic ring is put 1 mm beneath the capillary tip. Polydispersity of the emulsion droplets obtained is around 6%.

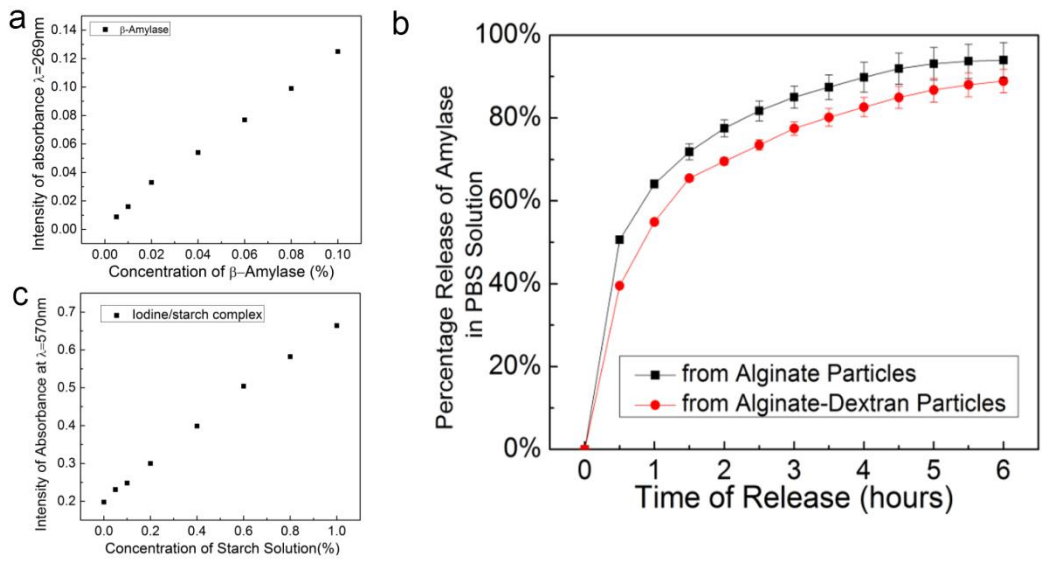

Figure S5. (a) A calibration curve showing the intensity of absorbance of β-amylase solution as a function of β-amylase concentration measured from the UV-vis spectrometer (λ=269 nm). (b) The standard release curve of the β-amylase from the alginate and alginate-dextran composite particles in the PBS solution at 37 ℃. The release efficiencies of amylase from the alginate and alginate-dextran particles are 94% and 89% respectively within the first 6 hours. After 6 hours, the amount of unreleased amylase is quantified by dissolving the particles in sodium citrate solution. (c) A calibration curve showing the intensity of absorbance of the iodine/starch complex solution (λ=570 nm) as a function of the starch concentration.

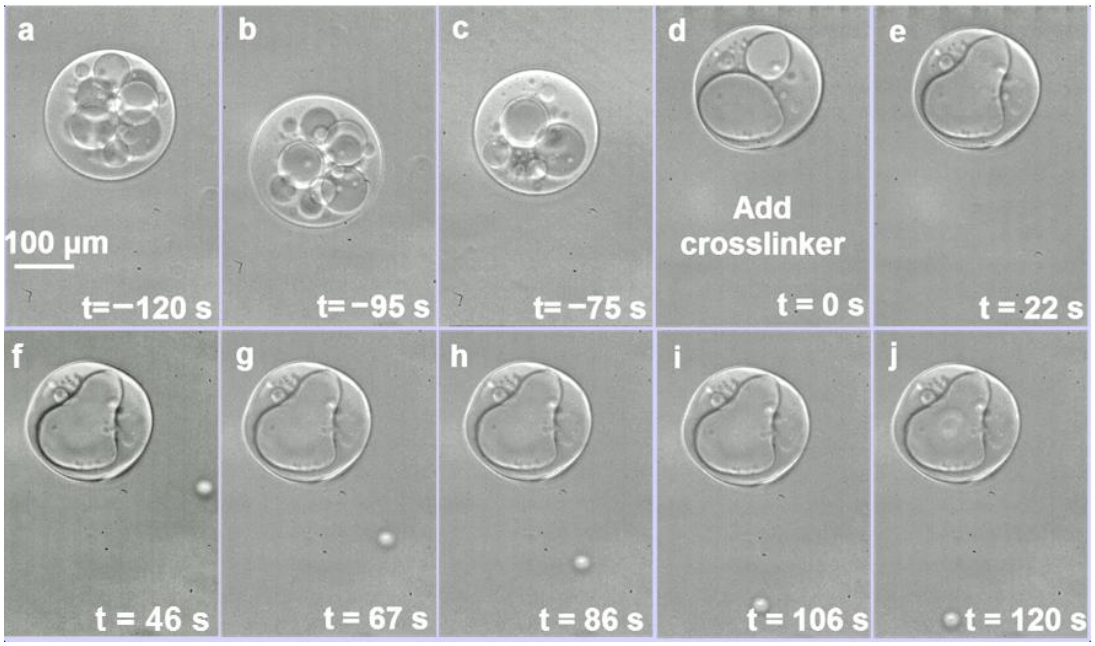

Figure S6. Optical microscope images showing that the emulsion structure can be quickly immobilized by addition of cross-linkers from the continuous phase. (a-c) before the addition of the crosslinkers, the PEG-rich inner droplets coalesce with each other, resulting in the reduced number of inner droplets. (d) The crosslinker of sodium dextran sulfate is added to the continuous phase at the time point t=0 s. (e-j) Due to the electrostatic interaction of the dextran sulfate with the collagen in the shell, the emulsion structure is quickly immobilized after about 20 seconds, as confirmed by the cessation of further coalescence as well as the maintenance of the non-spherical droplet shape.