Supplemental Figures & Legends

Formation of Core-Sheath Polymer Fibers by Free Surface Spinning of Aqueous Two-Phase Systems

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Supplemental Figure 1. Path of the microneedle (captured after withdrawal). The deflection of the dextran-PEO interface is visible in the bulk PEO phase several minutes after microneedle withdrawal. Scale bars are 100 µm.
### Supplemental Figure 2. Epifluorescence images of fibers formed from the dextran-PEO interface.

The localization of FITC-dextran and pyrene-PEG-rhodamine supports a core-sheath structure when the dextran solution is placed at the front of the reservoir and the PEO solution is placed at the back of the reservoir. A thin fiber with discontinuous FITC-dextran and pyrene-PEG-rhodamine signal is formed when the PEO solution is placed at the front of the reservoir and the dextran solution is placed at the back of the reservoir. Scale bars are 20 µm.
**Supplemental Figure 3.** (A) Proposed mechanism for fiber formation when the dextran solution is placed at the back of the reservoir and the PEO solution is placed at the front of the reservoir. (B) A single frame from a 60 frame/s video after needle withdrawal, showing the expected needle path into the dextran phase, the phase boundary between the dextran and PEO phases, and the presence of a cone-like structure in the PEO phase. (C) Movement of the microneedle through the reservoir containing PEO and dextran. Scale bars in (B) and (C) are approximately 500 µm.
Supplemental Figure 4. Localization of dextran and PEO phases after needle withdrawal when the dextran solution is placed at the back of the reservoir and the PEO solution is placed at the front of the reservoir. Scale bars are 100 µm.
Supplemental Figure 5. Fibers formed from 10 wt% and 15 wt% PEO for a dextran concentration of 54 wt%. Scale bars are 50 µm.
**Supplemental Figure 6.** Fibers formed from either two solution of dextran or two solutions of PEO of varying concentration and viscosity. Scale bars are 50 µm.