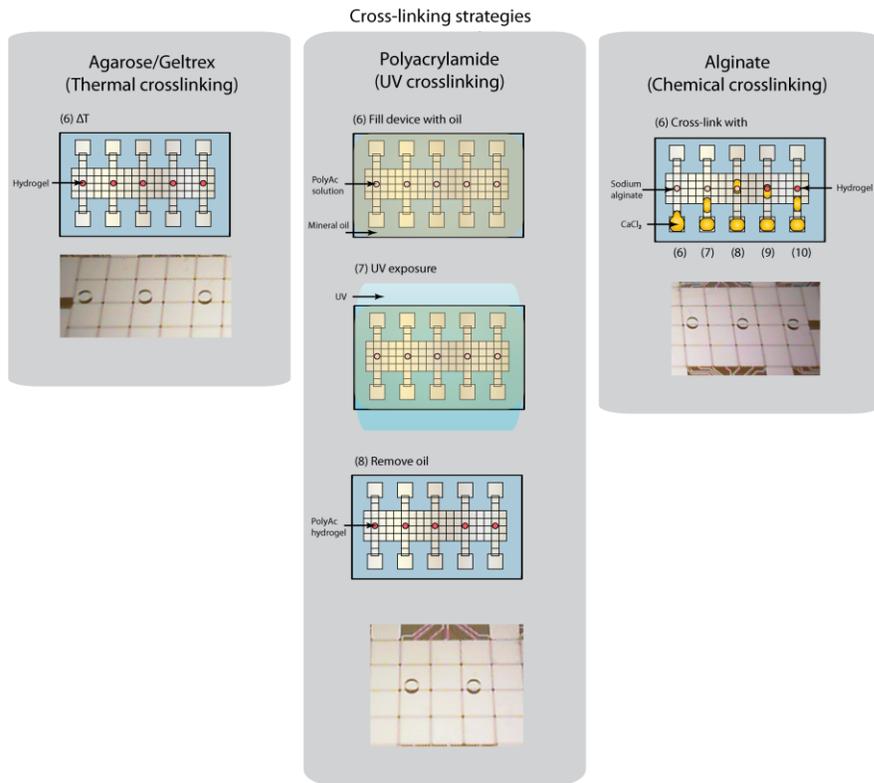
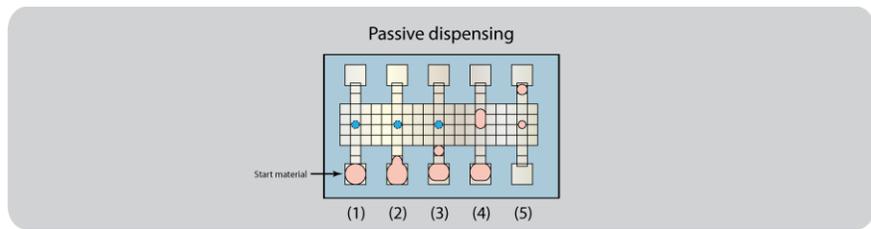
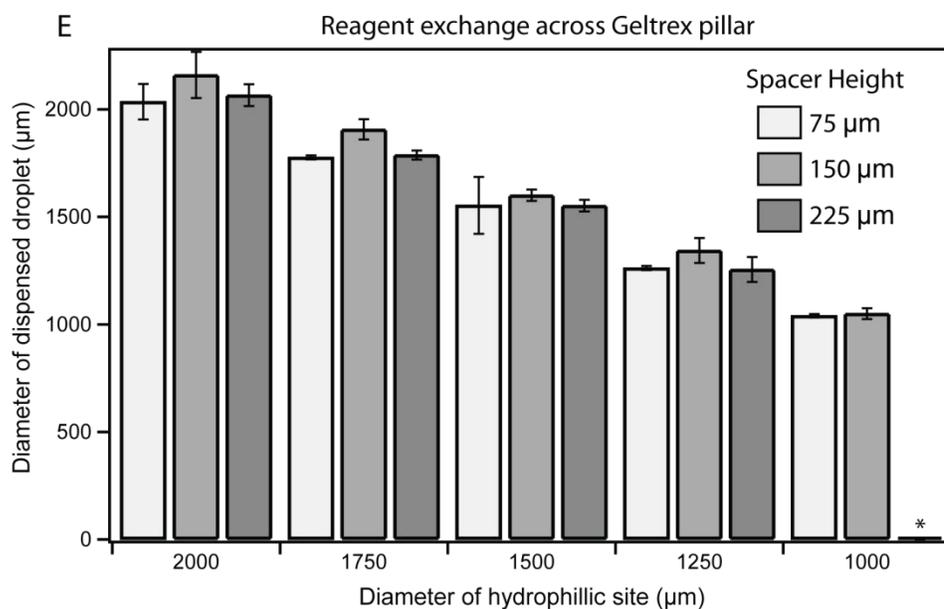
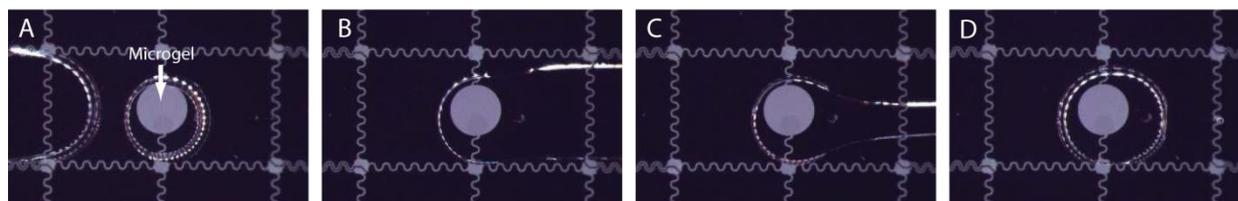


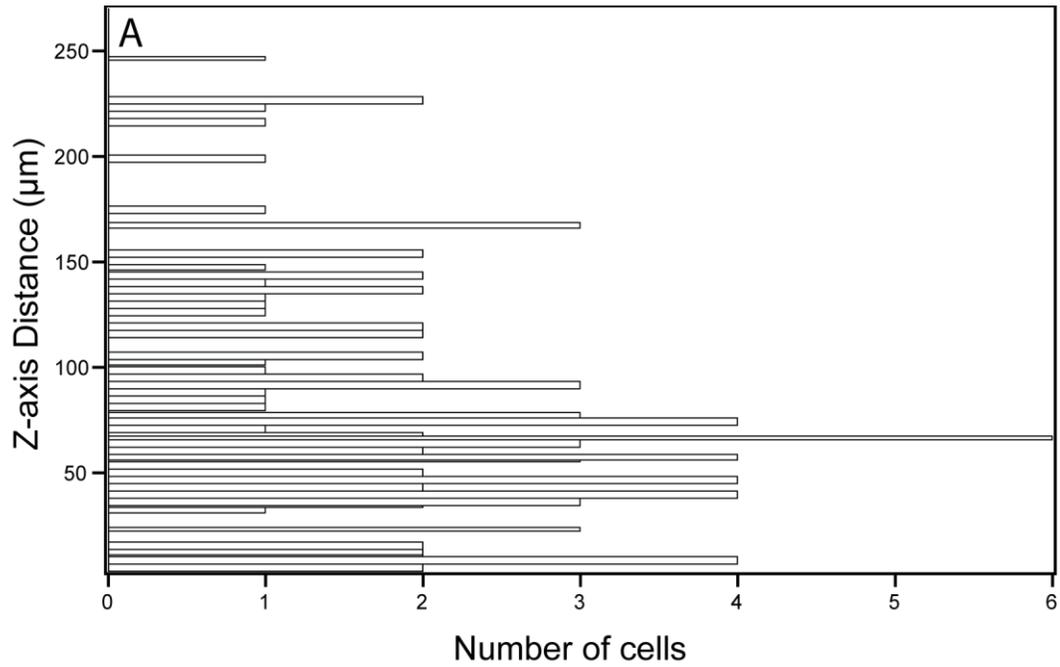
Supplementary Figure 1: *Precision of microgel on-demand formation.* Frames from a movie (A-D) depicting sol-state Geltrex being passively dispensed to form a sub-droplet. Scale bar = 2 mm. (E) The diameters of sol-state Geltrex droplets were measured by brightfield microscopy and compared to the diameters of circular hydrophilic sites. The asterisk indicates the single condition tested for which sub-droplets failed to form. Experiments were performed in triplicate and error bars indicate ± 1 S.D.



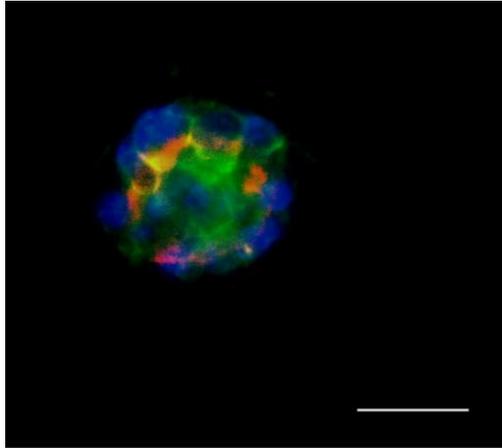
Supplementary Figure 2: *Microgel generation*. Sol-phase droplets formed on demand were crosslinked using thermal (left), UV (middle), or chemical (right) means. Photographs of microgels formed by each method are shown at the bottom of each column.



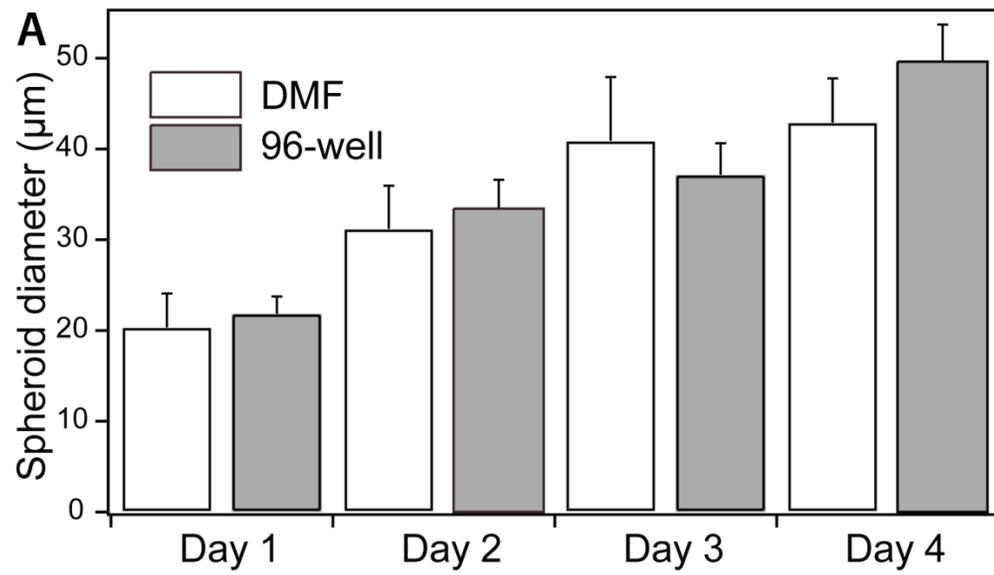
Supplementary Figure 3: *Precision of reagent exchange across microgels*. Frames from a movie (A-D) depicting a droplet of reagent passed across a microgel. (E) The diameters of dispensed droplets were measured by brightfield microscopy and were compared to the diameters of circular hydrophilic sites. The asterisk indicates the single condition tested for which sub-droplet failed to form. Experiments were performed in triplicate and error bars indicate ± 1 S.D.



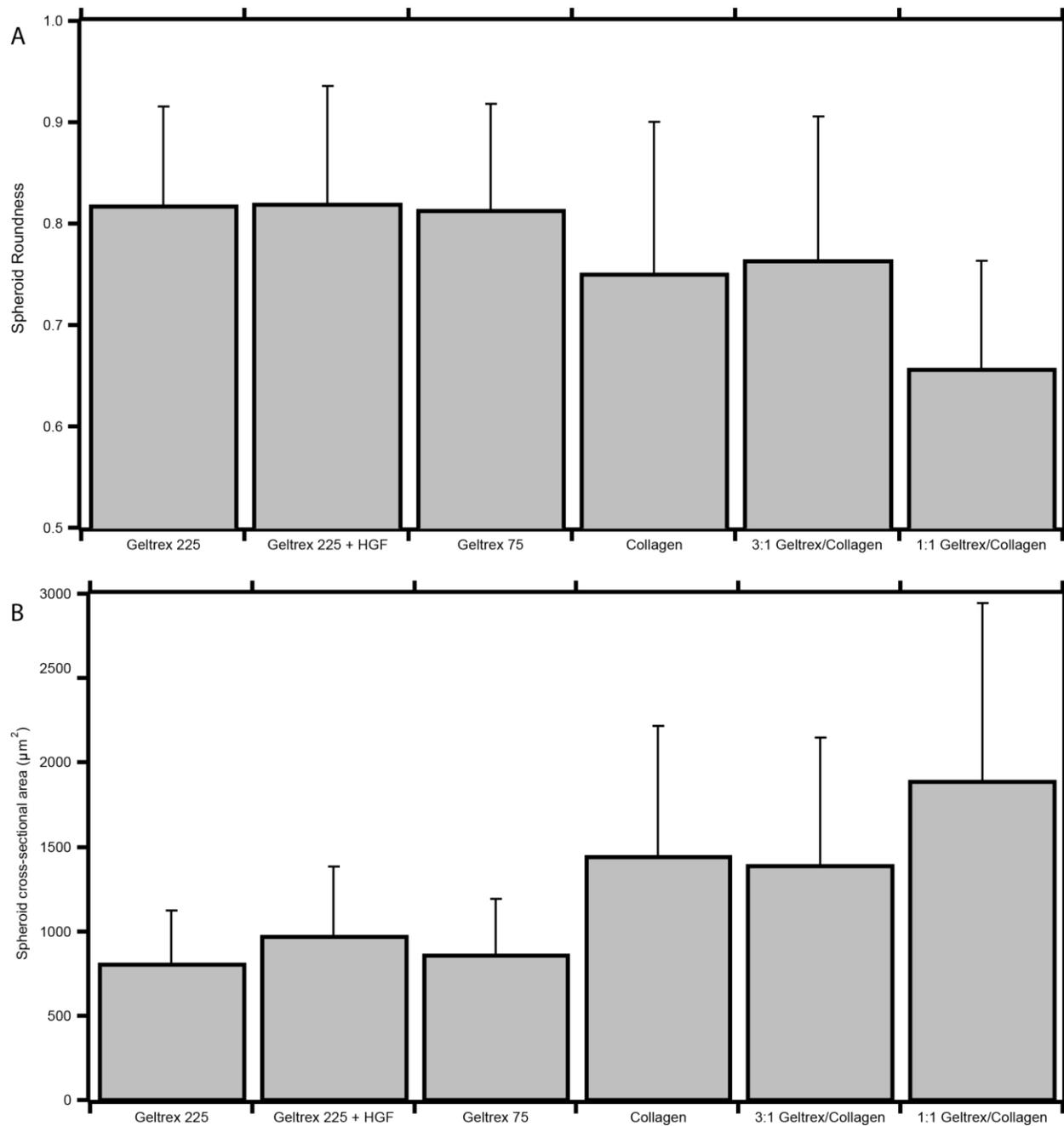
Supplementary Figure 4: *Z-axis distribution of cells in microgels*. Graph of distribution of cells in the z-axis for the data in Figure 5B in the main text. The number of cells was enumerated at each image plane (~6 μm thick).



Supplementary Figure 5: *Immunostaining on-demand and spheroid polarization*. A single spheroid grown until day 5 and then fixed and stained for ZO-1 (red), E-cadherin (green), and nuclei (blue). ZO-1 accumulation along the luminal wall indicates spheroid polarization. Scale bar = 50 μm .



Supplementary Figure 6: *Spheroid size*. Comparison of spheroid size for three-dimensional MDCK culture in microscale (DMF, white bars) and macroscale (96-well plate, gray bars).



Supplementary Figure 7: *Pilot Screen*. Comparison of spheroid roundness (A) and cross-sectional area (B) across all evaluated conditions. Error bars are + 1 S.D.