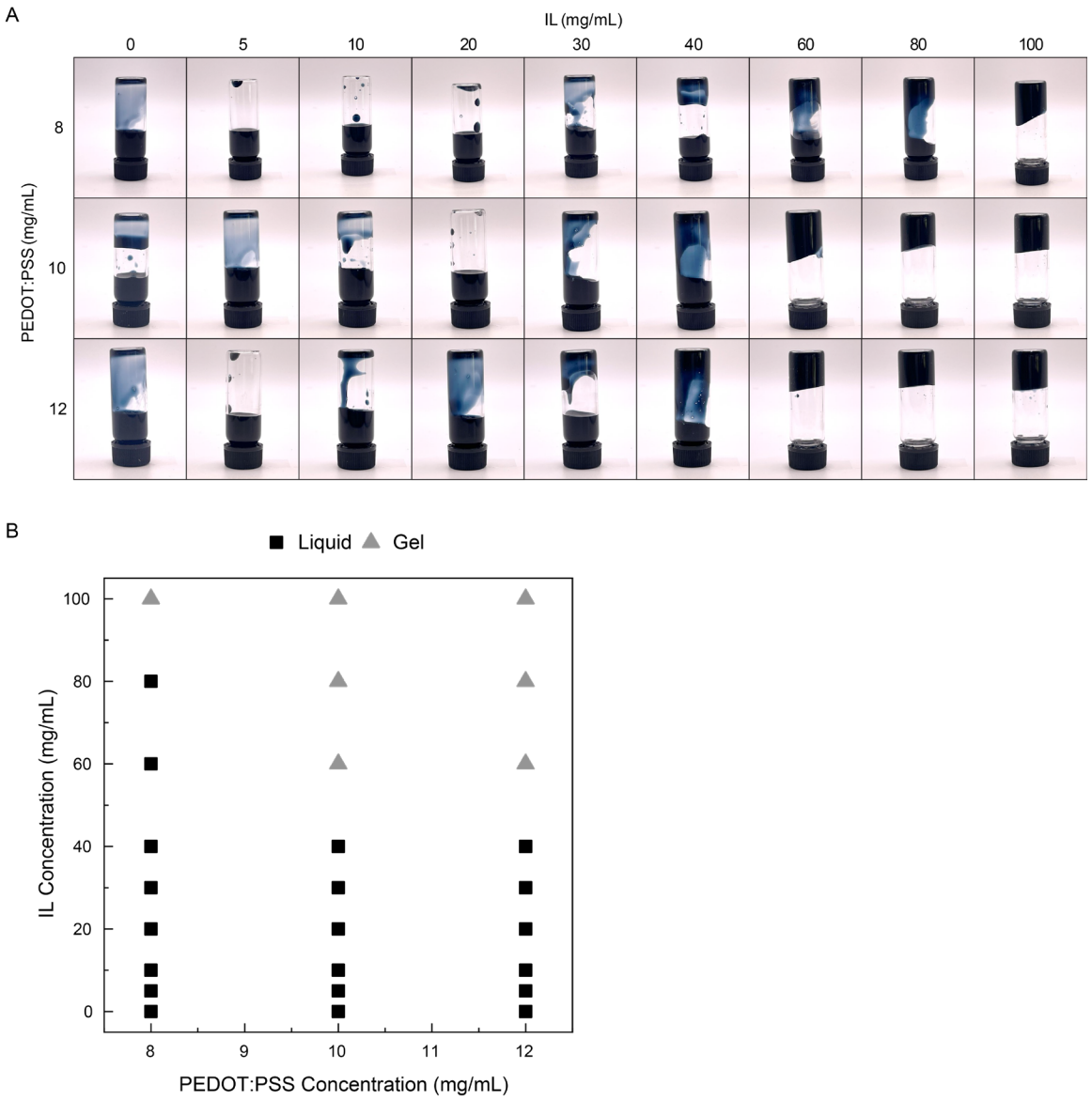
Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.



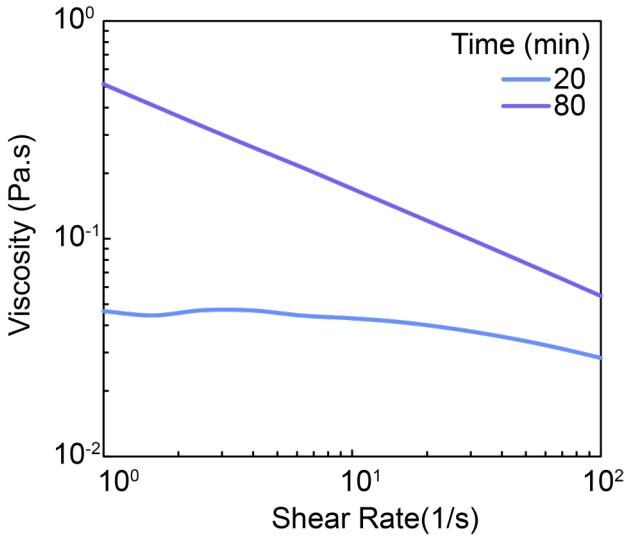
Supporting Figure 1. **Qualitative assessment of PEDOT:PSS-IL mixture phase and gelation kinetics.** A) Photos of tube inversions of 27 formulations used for generating B) a scatter plot of mixture phase, liquid or gel at 15 minutes after IL addition. The formulations with the most diluted PEDOT:PSS (8 mg/mL) were gels at 100 mg/mL IL while the formulations using 10 and 12 mg/mL PEDOT:PSS were gels at 60 mg/mL IL and above. All formulations investigated below these thresholds were liquids. N=1 per formulation.

Supporting Table 1. Table of values of PEDOT:PSS-IL mixture properties at 20 min after IL addition from representative plots of oscillatory and rotational rheology data presented in Figures 2 and 3.

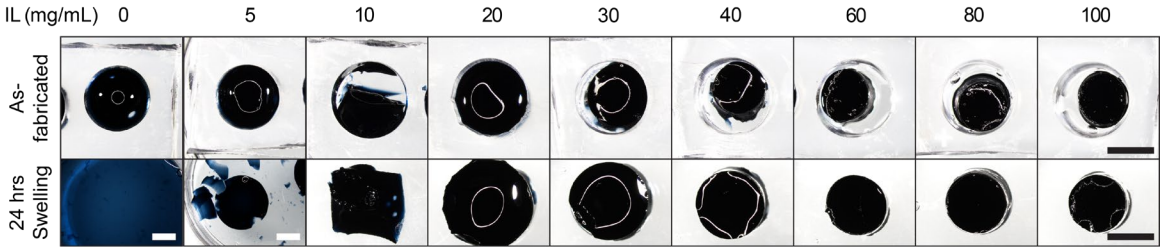
IL conc (mg/mL)	G' (Pa)	G" (Pa)	tan(δ)	Phase	G'-G'' Cros-over (min)	Viscosity (Pa.s)
10	0.00132	0.0398	30.1	Liquid	NA	0.0284
20	0.0570	0.192	3.37	Liquid	62.3	0.0530
30	0.321	0.444	1.38	Liquid	25.4	0.0726
40	0.895	0.823	0.919	Gel	19.6	0.111
60	3.63	1.60	0.440	Gel	NA	NA
80	12.8	2.39	0.187	Gel	NA	NA
100	16.4	2.93	0.179	Gel	NA	NA

Supporting Table 2. Table of values of PEDOT:PSS-IL mixture properties at 75 min after IL addition from representative plots of oscillatory and rotational rheology data presented in Figure 2.

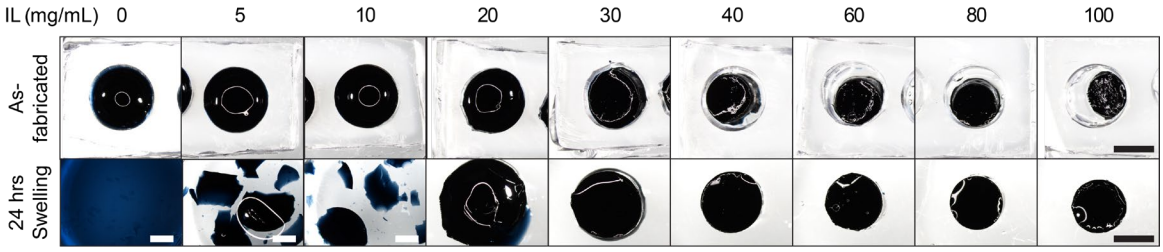
IL conc (mg/mL)	G' (Pa)	G" (Pa)	tan(δ)	Phase
10	0.0170	0.0897	5.27	Liquid
20	0.766	0.626	0.818	Gel
30	4.09	1.31	0.321	Gel
40	16.8	2.53	0.151	Gel
60	31.4	3.05	0.0968	Gel
80	72.5	4.63	0.0638	Gel
100	83.8	5.30	0.0632	Gel



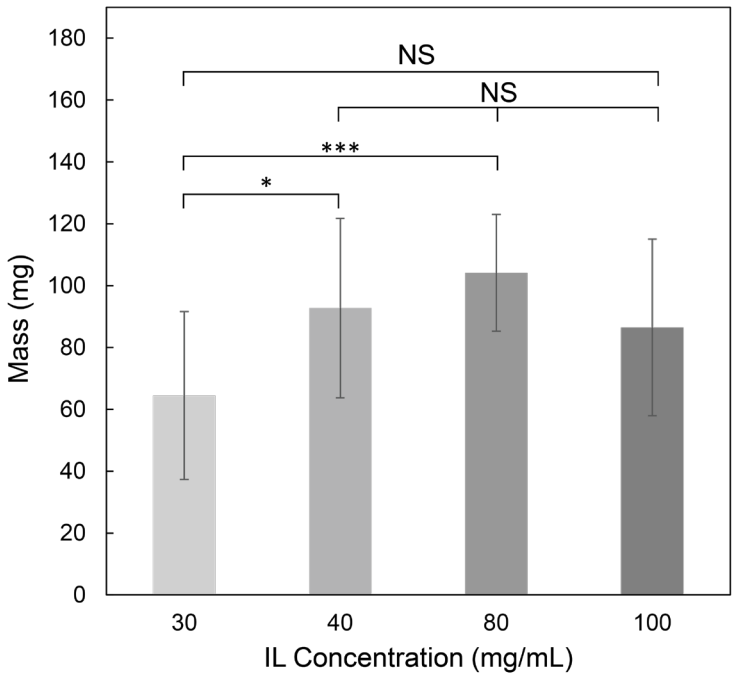
Supporting Figure 2. Rotational rheology of PEDOT:PSS-IL mixtures with 10 mg/mL IL. Representative plots of viscosity as a function of shear rate measured 20 and 80 minutes after mixing PEDOT:PSS with IL. Viscosity has increased ~92.3% at 100 1/s over the 60 min time period analyzed.



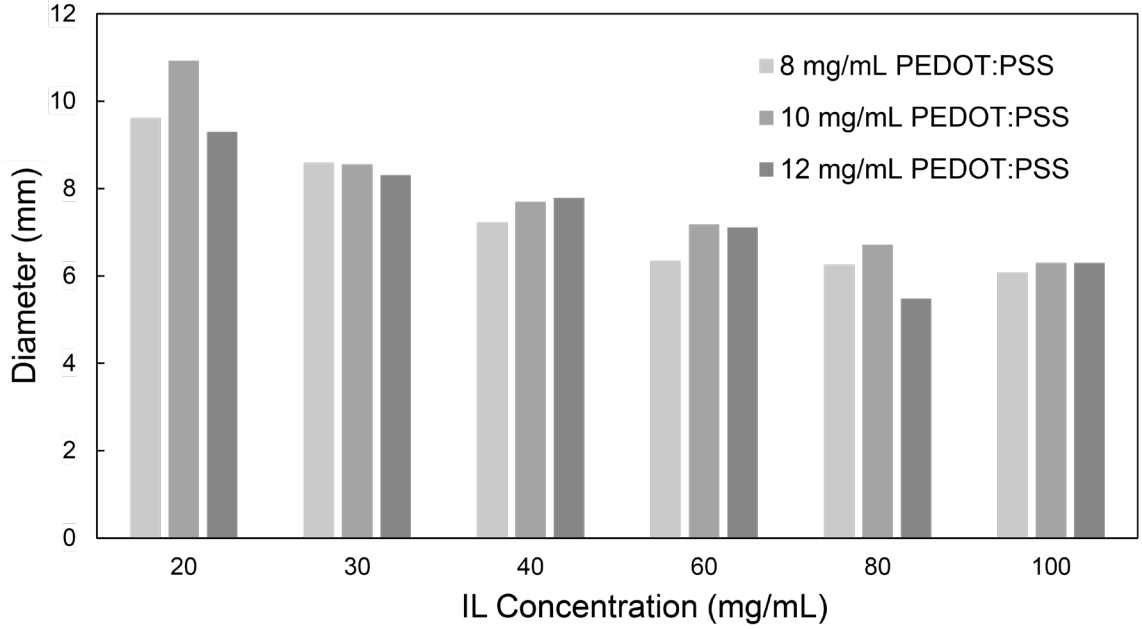
Supporting Figure 3. PEDOT:PSS hydrogels fabricated with 8 mg/mL PEDOT:PSS and nine IL concentrations were evaluated for stability after 24 hrs swelling in deionized water. N=2 per formulation. Representative images. Scale bars 5 mm; black scale bars apply to all images except those with a white scale bar.



Supporting Figure 4. PEDOT:PSS hydrogels fabricated with 10 mg/mL PEDOT:PSS and nine IL concentrations were evaluated for stability after 24 hrs swelling in deionized water. N=2 per formulation. Representative images. Scale bars 5 mm; black scale bars apply to all images except those with a white scale bar.

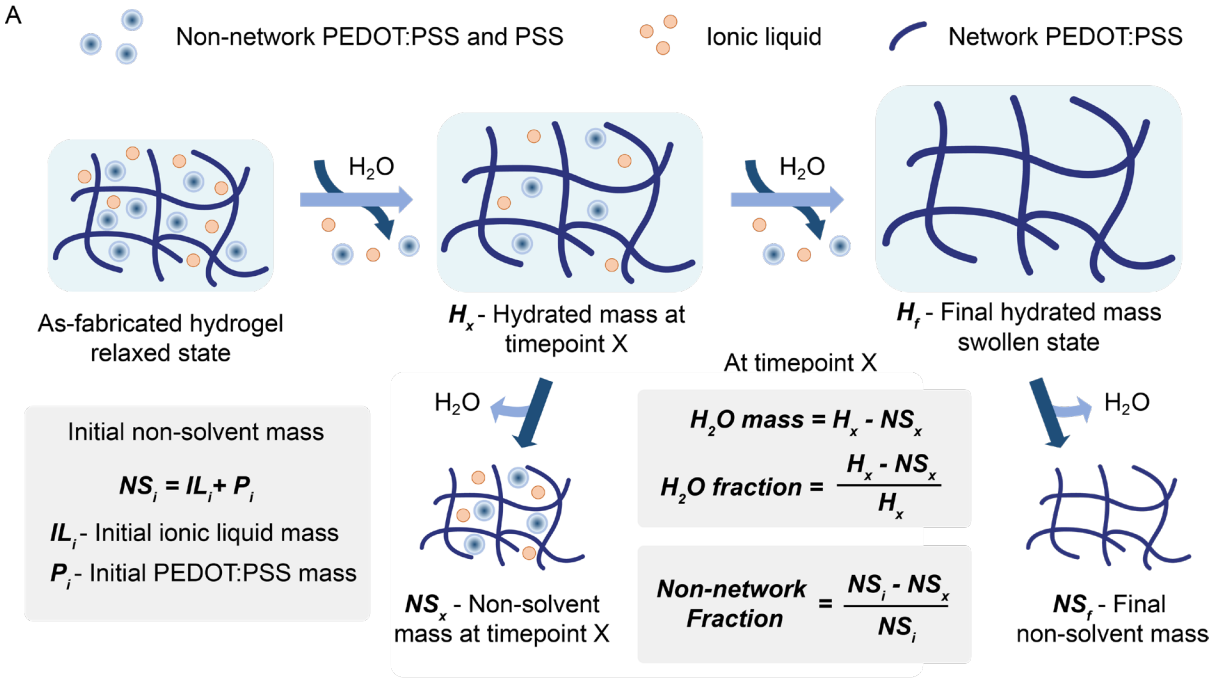


Supporting Figure 5. Masses (mg) of PEDOT:PSS hydrogels (12 mg/mL) after fabrication are less than the starting mass of the precursor mixture (~250 mg) due to water evaporation during oven incubation. The observed decreases correspond to mass losses of 58.3-74.2% of the starting mass. Mean and standard deviation presented. N≥8 per concentration. One-way analysis of variance (ANOVA) and Tukey's multiple comparison test. ****P≤0.0001 ***P≤0.001 **P≤0.01 *P≤0.05 Non-Significant (NS) P>0.05.



Supporting Figure 6. Hydrogel diameters at 24 hours swelling in deionized water were found to be similar for a given IL concentration across varying PEDOT:PSS concentrations. N=2 per formulation, mean presented.

A



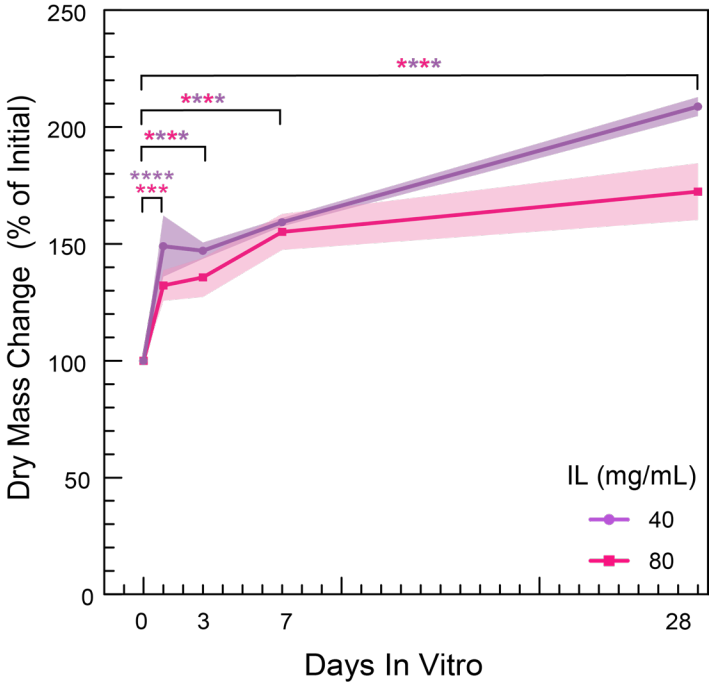
B

	% of final hydrated mass H_f		% of initial non-solvent mass NS_i			% of initial PEDOT:PSS mass P_i
IL (mg/mL)	Water fraction $H_f - NS_f$	Polymer fraction NS_f	Non-network fraction $NS_i - NS_f$	Initial IL mass IL_i	Estimated PEDOT:PSS mass loss $NS_i - NS_f - IL_i$	Estimated PEDOT:PSS mass loss $P_i - NS_f$
30	99.8	0.179	86.7	71.4	15.2	53.3
40	99.7	0.330	86.0	76.9	9.06	39.3
60	99.6	0.418	89.2	83.3	5.83	35.0
80	99.3	0.714	91.2	87.0	4.25	32.6
100	99.0	0.993	92.4	89.3	3.10	28.9

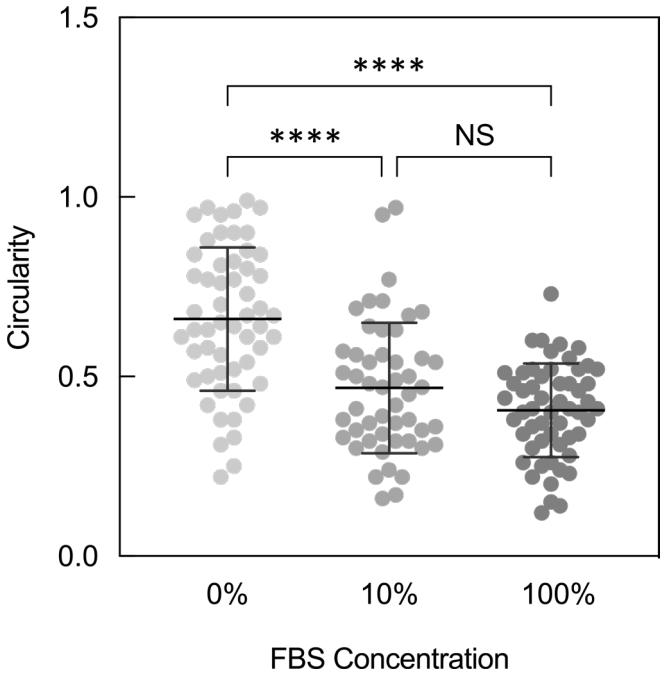
Supporting Figure 7. Water and non-network fractions of PEDOT:PSS hydrogels are dependent on IL concentration. A) Schematic and equations for calculating water mass, water fraction, and non-network fraction of hydrogels. At timepoint X, water mass is determined by taking the difference of hydrated mass H_x and non-solvent mass NS_x. NS_x is obtained from measuring lyophilized hydrogels. Water fraction is defined as the fraction of water mass to hydrated mass H_x. Non-network fraction is defined as the fraction of initial non-solvent mass NS_i that does not stay linked to the hydrogel network and is removed by washing. In this case, non-network fraction is expected to comprise of IL, excess PSS, and non-network PEDOT:PSS colloidal nanogels or larger aggregates. Initial solvent mass NS_i is the sum of initial ionic liquid mass IL_i and initial PEDOT:PSS mass P_i. NS_i was not directly measured but was assumed to be the same as that calculated from volumes and known concentrations. B) Table of mean values of final fractions and non-solvent mass changes of equilibrium swollen hydrogels fabricated with 30-100 mg/mL IL (in deionized water for at least 7 days, N=2-6 per concentration). Polymer fractions were calculated assuming that the non-solvent mass at day 7 was only network PEDOT:PSS.

Supporting Table 3. Table of values for elastic modulus and ultimate strength in compression as well as conductivity of PEDOT:PSS hydrogels. Mean and standard deviation presented. N≥7 per group.

Ionic Liquid Concentration (mg/mL)	Elastic Modulus (kPa)	Ultimate Strength (kPa)	Conductivity (S/m)
30	1.05 ± 0.343	0.11 ± 0.055	2.75 ± 3.75
40	2.91 ± 1.59	0.38 ± 0.24	7.19 ± 5.24
80	4.10 ± 1.43	0.61 ± 0.42	52.2 ± 26.4
100	4.13 ± 1.63	0.79 ± 0.49	127 ± 75.1



Supporting Figure 8. PEDOT:PSS hydrogels were fabricated with 40 and 80 mg/mL IL, washed, disinfected, and suspended in Dulbecco's Modified Eagle Medium (DMEM) with 10% fetal bovine serum (FBS) at 37 °C and 5% CO_2 for 28 days. Hydrogel dry mass (measured from lyophilized hydrogels) significantly increased in comparison to day 0 throughout the twenty-eight day period in both concentrations; 210% and 170% on day 28 in 40 mg/mL and 80 mg/mL, respectively. N=4 per concentration per time point. Plot lines denote mean and colored shaded areas denote standard deviation presented. One-way analysis of variance (ANOVA) and Dunnett's multiple comparison test with Day 0 as the control. ****P≤0.0001 ***P≤0.001 **P≤0.01 *P≤0.05 Non-Significant (NS) P>0.05. Asterisks of alternating colors indicate significance applies to both concentrations, while asteriks of one color are specific to the inidicated concentration.



Supporting Figure 9. Fibroblasts were more spread on hydrogels pre-conditioned with serum-containing media. PEDOT:PSS hydrogels were fabricated with 80 mg/mL IL, and pre-conditioned with Dulbecco's Modified Eagle Medium (DMEM), 10% fetal bovine serum (FBS) in DMEM and 100% FBS. After overnight culture, circularities of normal human dermal fibroblasts (N>50 cells) on hydrogels pre-conditioned with 10% and 100% FBS were significantly lower compared to those on hydrogels pre-conditioned with DMEM alone. Circularity is determined by $4\pi A/P^2$, in which A is the cell area and P is the perimeter. A mathematically perfect circle has a value of 1. Mean and standard deviation presented. Two-way analysis of variance (ANOVA) and Šidák's multiple comparison test: ****P≤0.0001 ***P≤0.001 **P≤0.01 *P≤0.05 Non-Significant (NS) P>0.05.

Supporting Table 4. Expansion of normal human dermal fibroblasts on tissue culture plastic (TCP) versus PEDOT:PSS hydrogels fabricated with 40 mg/mL or 80 mg/mL IL. Cell expansion is defined as the ratio of cell density to that of the previous time point. Day 1 cell density was compared to the initial seeding density (denoted as Day 0). N=1 TCP control per timepoint; N=3-4 hydrogels per condition per timepoint, mean and standard deviation presented.

Substrates\Expansion	**Day 1/Day 0**	**Day 3/Day 1**	**Day 7/Day 3**	**Day 7/Day 14**
Tissue culture plastic	1.21	4.22	3.39	1.19
PEDOT:PSS hydrogel 40 mg/mL IL	0.761 ± 0.330	6.46 ± 3.64	9.27 ± 2.85	1.02 ± 0.135
PEDOT:PSS hydrogel 80 mg/mL IL	1.05 ± 0.108	8.43 ± 0.987	5.78 ± 0.317	1.03 ± 0.103