Electronic Supplementary Information (ESI)

Mechanically Robust PEDOT:PSS Hydrogel via Mild Liquid Metal Crosslinking *

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S1. Control experiment: EGaIn-PP hydrogel prepared without DBSA

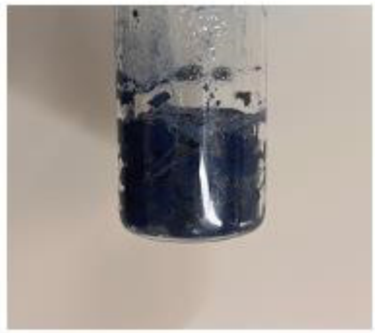

Figure S1. Photograph of the EGaIn-PP hydrogel prepared without DBSA.

S2. Effect of ultrasonication conditions on the size of EGaIn nanoparticles

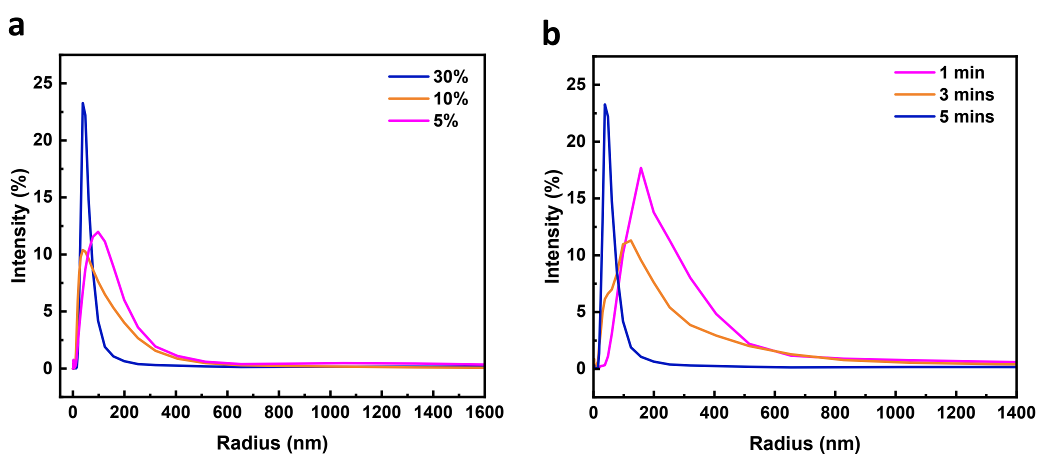

Figure S2. Effect of (a) ultrasonication amplitude and (b) ultrasonication time on the size of EGaIn nanoparticles.

S3. KWW growth model fitting of time-dependent modulus evolution

The time-dependent evolution of the storage modulus of the PEDOT:PSS and EGaIn-PP hydrogels was analyzed using a stretched-exponential Kohlrausch–Williams–Watts (KWW) growth model. The experimental data (average) were fitted using the following expression:

$$G'(t) = G'\infty - (G'\infty - G'0)exp\left[-\left(\frac{t}{\lambda}\right)^{\beta}\right]$$

where G′(t) is the storage modulus at time t, G′0 is the initial storage modulus at the beginning of the measurement, and G′∞ is the asymptotic storage modulus at long times.

The parameter λ is the characteristic time constant describing the timescale of network development, and the stretching exponent β ($0 < \beta \leq 1$) reflects the breadth of the relaxation or growth time distribution. A value of $\beta = 1$ corresponds to a single exponential growth process with a narrow and homogeneous relaxation spectrum, whereas $\beta < 1$ indicates a broader distribution of characteristic times and a higher degree of dynamic and structural heterogeneity within the evolving network.

In the context of hydrogel formation, the KWW growth model provides a phenomenological description of the progressive network reinforcement process arising from the formation and rearrangement of physical crosslinks. A smaller λ represents faster modulus evolution, while a lower β indicates a more heterogeneous and dynamically evolving network structure.

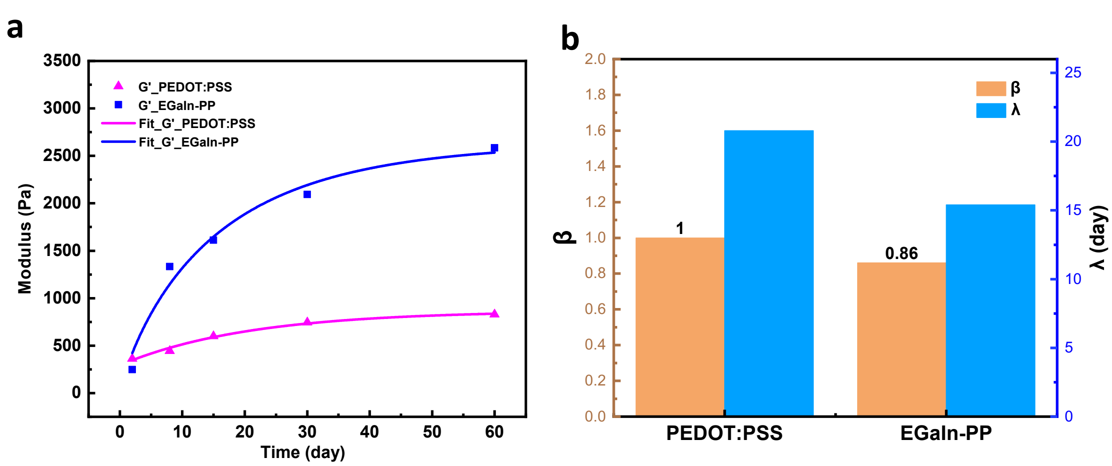

Figure S3. (a) KWW growth model fitting of the time-dependent storage modulus evolution of the PEDOT:PSS hydrogel and the EGaIn-PP hydrogel. (b) Comparison of the fitted KWW parameters, including the stretching exponent β and characteristic time λ, for the PEDOT:PSS hydrogel and the EGaIn-PP hydrogel.

S4. Cyclic compression stress–strain curves of the PEDOT:PSS hydrogel

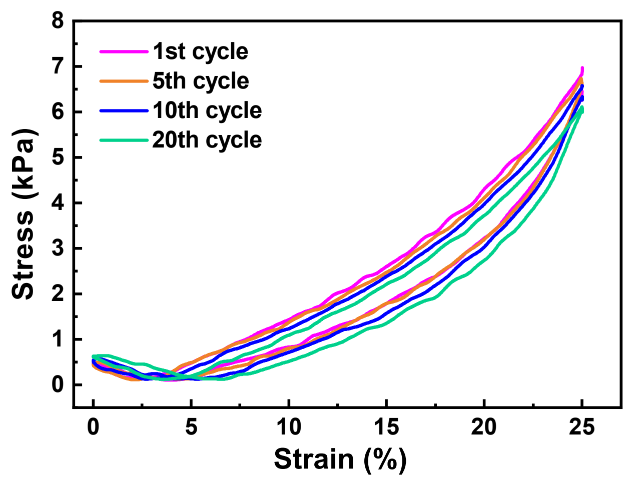

Figure S4. Cyclic compression stress–strain curves of the PEDOT:PSS hydrogel

S5. Electrical Characterization of Hydrogels

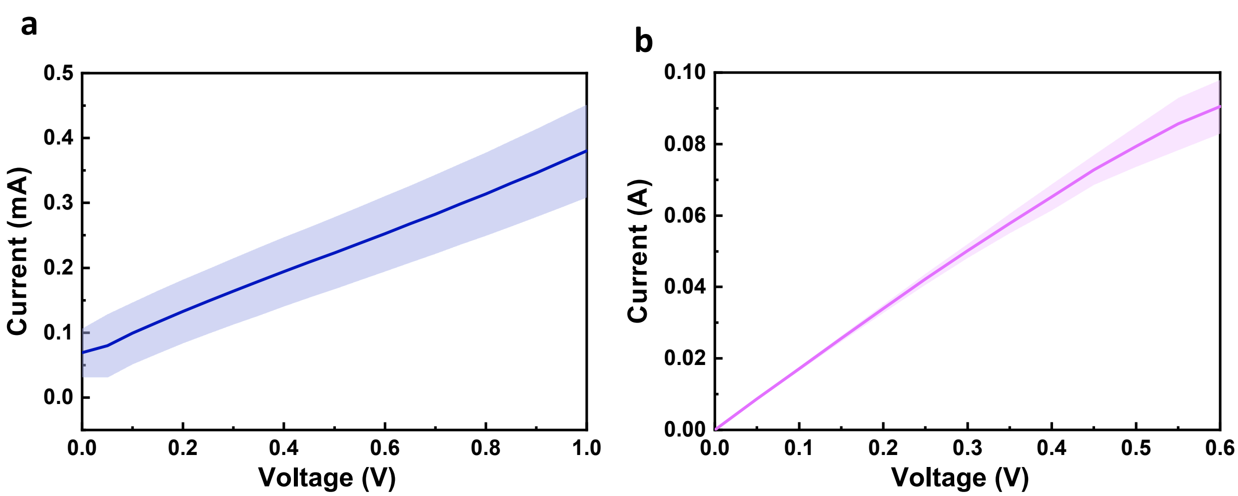

Figure S5. Current–voltage (I–V) characteristics of (a) the EGaIn–PP hydrogel and (b) PEDOT:PSS hydrogel measured using a two-electrode (parallel-plate) configuration.