

## **New Insights into Pure Zwitterionic Hydrogels with High Strength and High Toughness**

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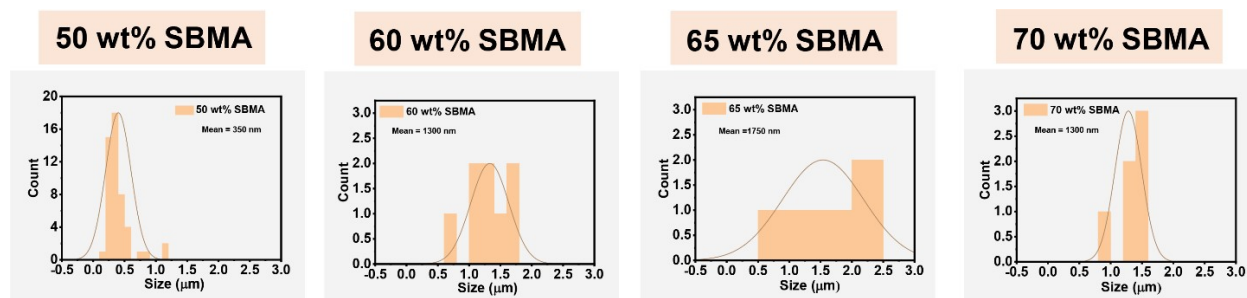
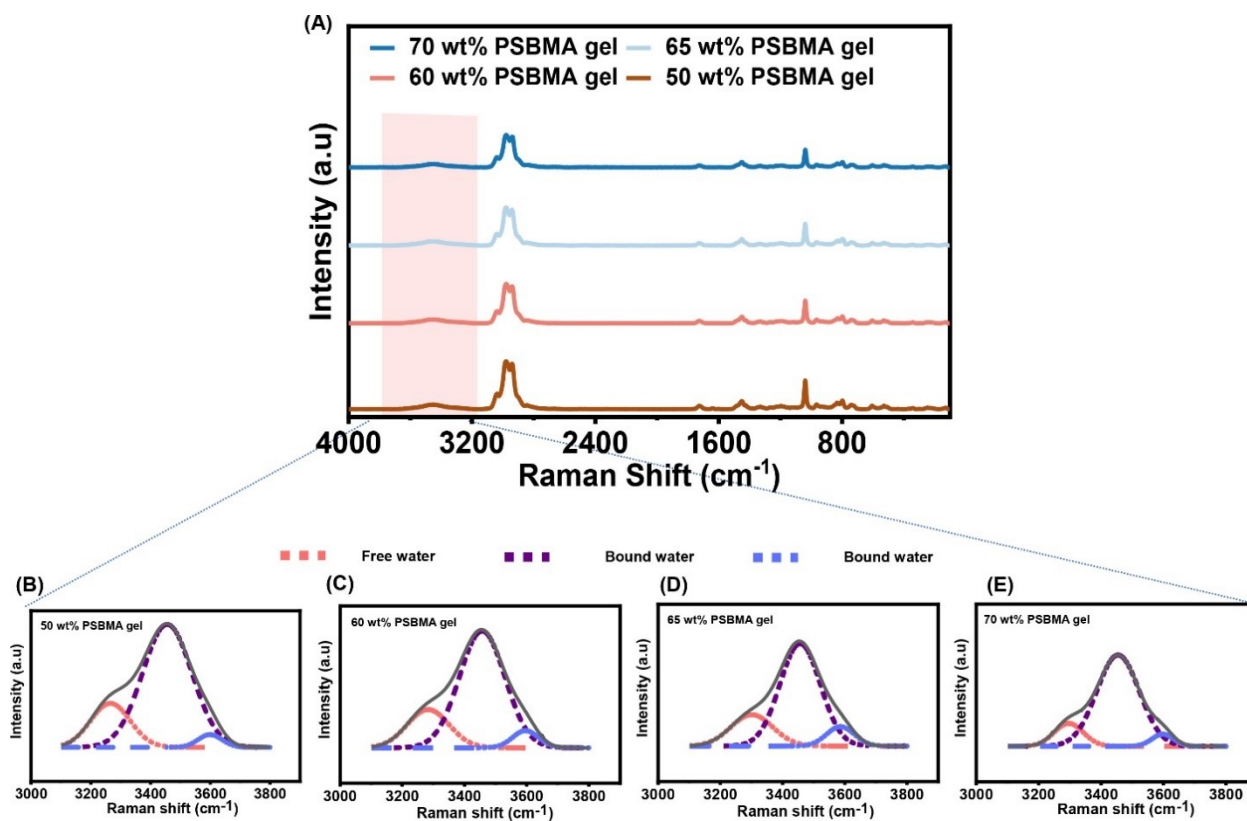


Figure S1. The related liquid phase condensate size distributions from the TEM data.



**Figure S2.** (A) Raman spectra of various concentrations of PSBMA hydrogels with crosslinker and initiator concentrations of 0.1 mol%. Pink sections of the spectra with SBMA concentrations of (B) 50 wt%, (C) 60 wt%, (D) 65 wt%, and (E) 70 wt%.

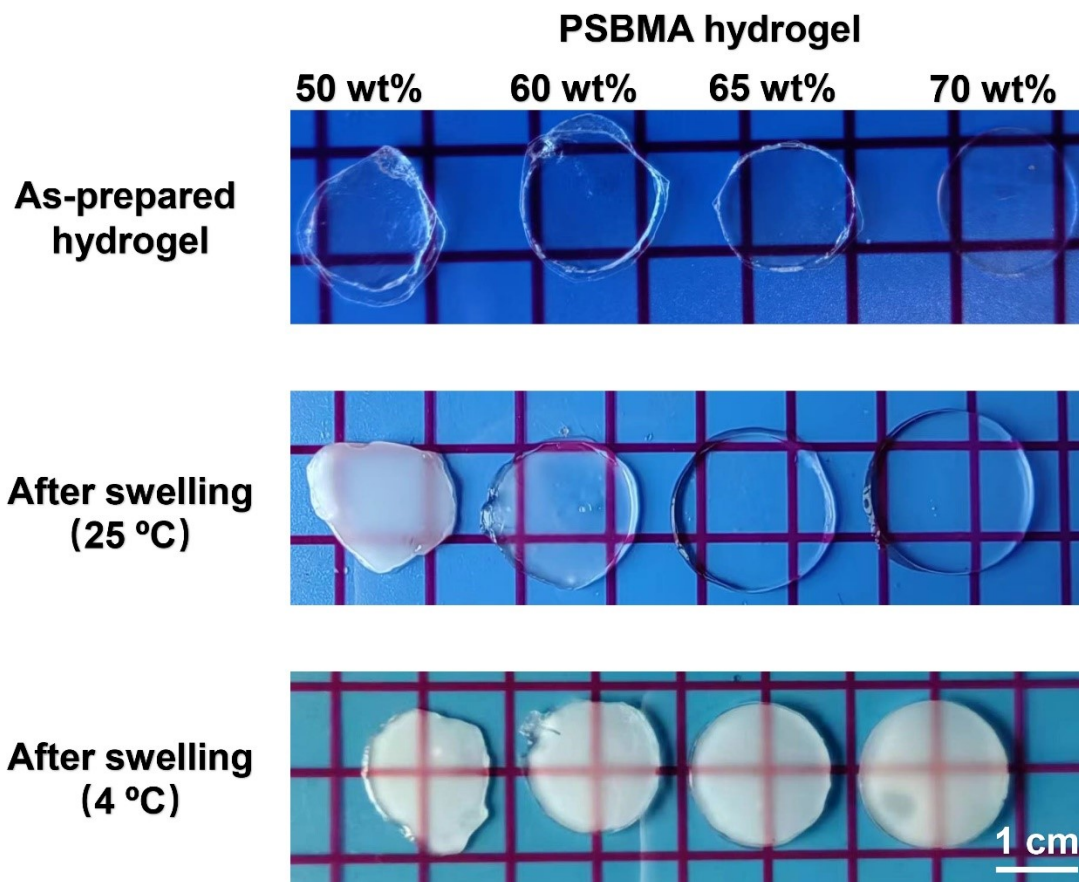
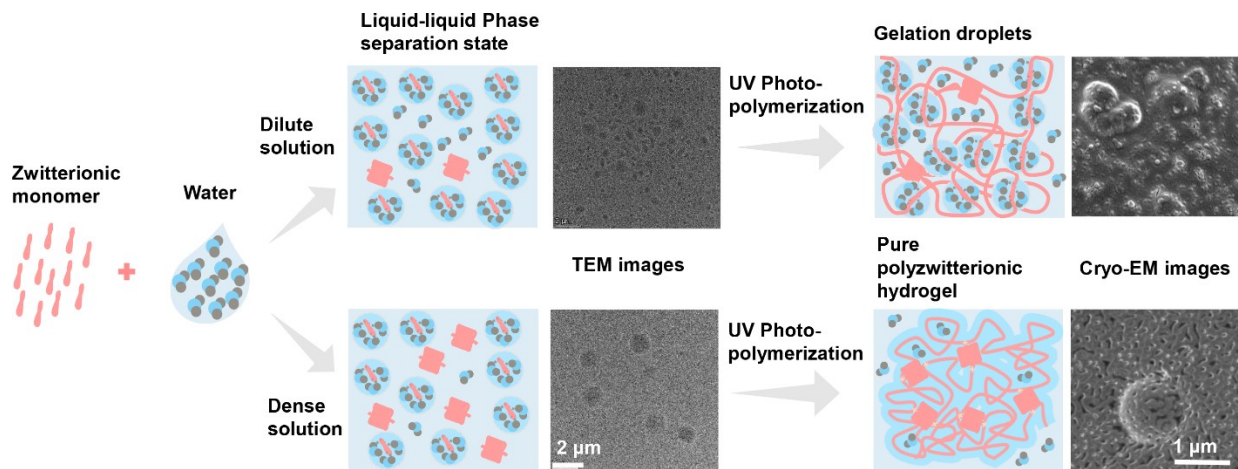
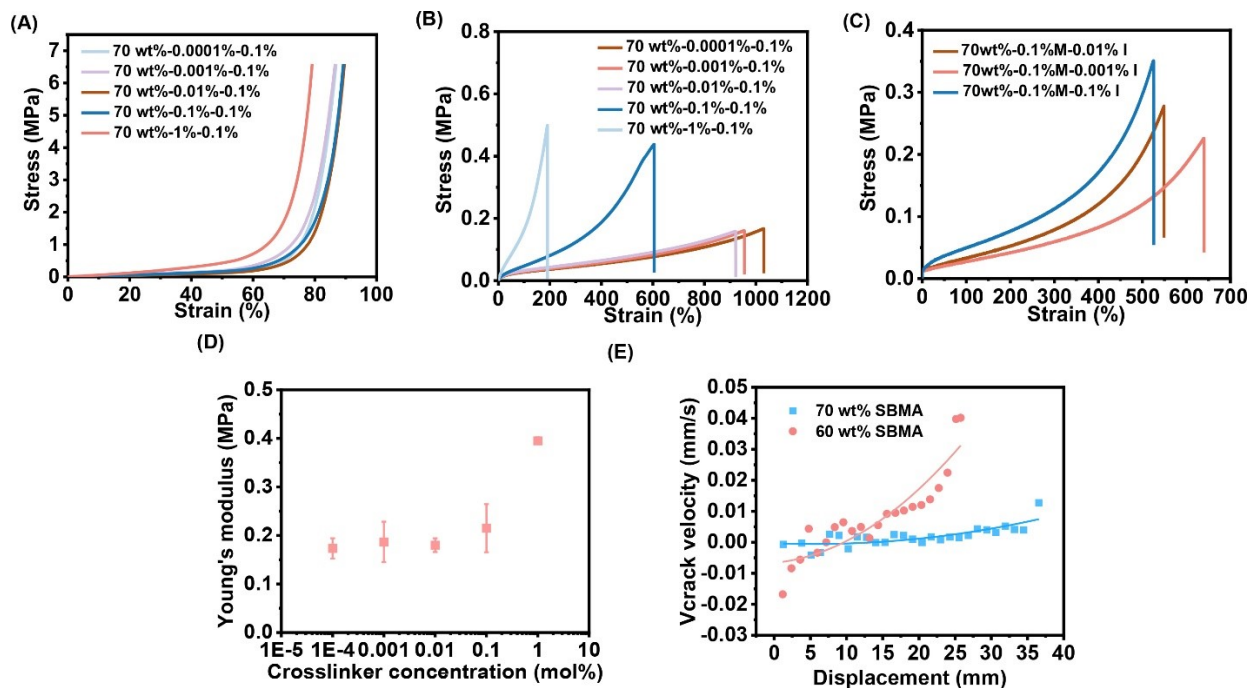


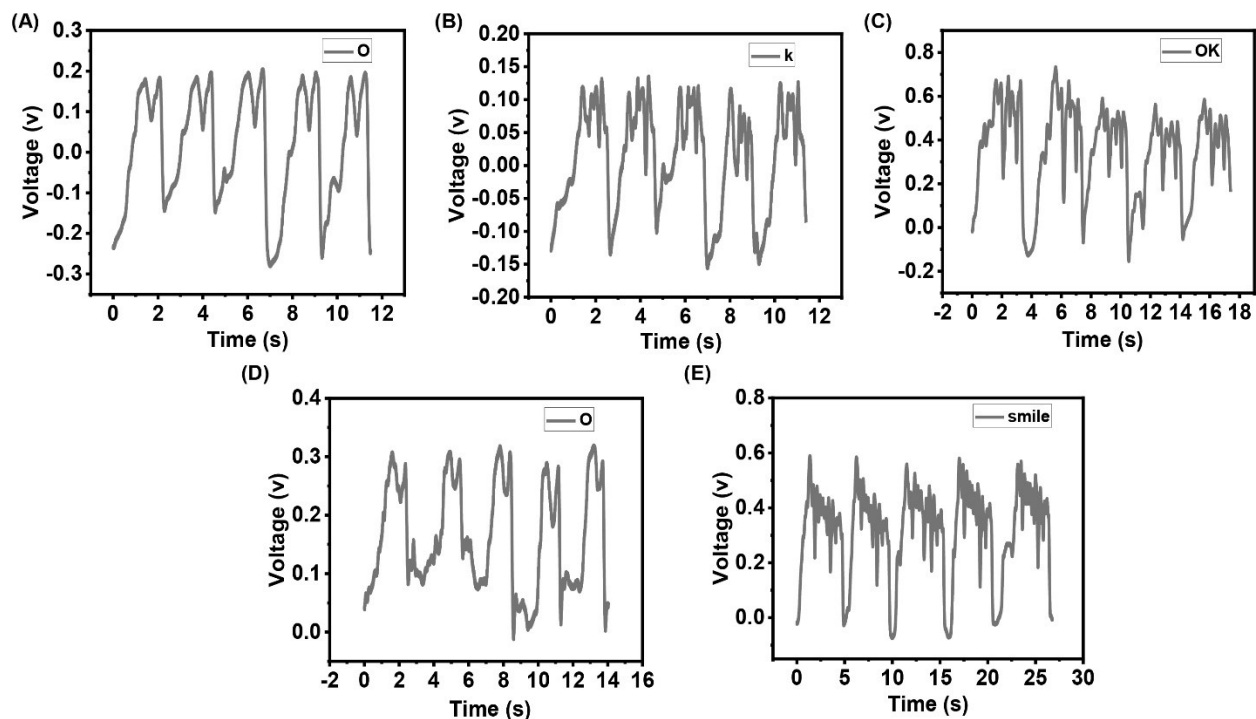
Figure S3. UCST behaviors of as-prepared hydrogel and after swelling the PSBMA hydrogel with different SBMA concentration.



**Figure S4.** Mechanism of liquid-liquid phase separation of solution state to different network structure of PSBMA hydrogels.



**Figure S5.** Mechanical properties of PSBMA hydrogels with various crosslinker and initiator concentrations at a monomer concentration of 70 wt%. Impact of crosslinker concentration on (A) compression testing results and (B) tensile testing results. (C) Impact of initiator concentration on tensile testing results. (D) Young's modulus with crosslinker concentration at an initiator concentration of 0.1 mol% condition. (E) Relationship between the displacement and crack velocity of PSBMA hydrogel with various SBMA concentration as 60 wt% and 70 wt%.



**Figure S6.** Voltage signals output by the rough-surfaced PSBMA-based TENG under different handwritten content: (A) letter “o”; (B) letter “k”; (C) word “ok”; (D) letter “o”; (E) word “smile”.

Table S1. Comparing the mechanical properties of various polyzwitterionic hydrogels

<b>Polymer composition</b>	<b>Tensile Strength (MPa)</b>	<b>Toughness (KJ/m<sup>2</sup>)</b>	<b>Compressive Strength (MPa)</b>	<b>Ref.</b>
<b>PMAA-co-DMAPS</b>	0.3	-	-	Wu et al. ACS Nano 2018, 12, 12860–12868.
<b>P(SBMA-co-AM)</b>	0.03	-	-	Yuan et al. ACS Appl. Mater. Interfaces 2019, 11, 40620–40628
<b>PCB</b>	<0.01	-	<0.45	Ma et al. Biomaterials 230 (2020) 119640
<b>FxSyR Hydrogel</b>	<0.12	-	<1.6	Fu et al. ACS Appl. Mater. Interfaces 2020, 12, 52307–52318
<b>PMPC</b>	<0.15	-	6	Norioka et al. NPG Asia Materials (2021) 13:34
<b>PVIBPS</b>	0.2	0.46	-	Yang et al. Chem. Mater. 2021, 33, 8418–8429
<b>PSP</b>	<0.05	-	<0.2	Jia et al. Adv Funct Mater. 2023, 33 2302846
<b>PSBMA</b>	0.4	2.12	6	This work