# **Gelatin-based ionogel with anti-swelling properties for underwater human**

## **physiological signal detection**

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#### **Synthesis of DMAEMA–MA**

The procedure for the synthesis of DMAEMA-MA was shown in Figure S1. First, 9.43 g (0.06 mol) of 2-(dimethylamino)ethyl methacrylate was added to 15.0 mL of acetone solution. After stirring, 3.48 g (0.03 mol) maleic acid was added to the above solution until complete dissolution. Then, the solution was added to a round-bottomed flask under nitrogen protection and stirred at 50 °C for 4 hours. After completion of the reaction, the reaction solution was cooled at low temperature for 4 hours. Rotary evaporators was used for vacuum suction filtration to evaporate organic solvent. The obtained crude product was dried in vacuum at 65 ℃ for 4 hours to obtain white needlelike crystalline powder DMAEMA-MA.



**Figure S1.** Synthesis of ionic liquid DMAEMA–MA.

### **FT-IR of G-P(HEMA/BA/AA)-D/MBAA ionogel**

As shown in figure S2, FT-IR confirmed the successful preparation of G-P(HEMA/BA/AA)-D/MBAA ionogel. Comparing the FT-IR spectra of D, G and G-P(HEMA/BA/AA)-D/MBAA, it could be observed that the telescopic vibrational peak of -C=C (1642 cm-1) disappeared, demonstrating the successful introduction of DMAEMA-MA into the ionogel system. The C=O stretching vibrational peak of D at 1820 cm<sup>-1</sup> was shifted to 1720 cm<sup>-1</sup>. The vibrational peaks of N-H and C-N also shifted from 1483 cm<sup>-1</sup> and 1030 cm<sup>-1</sup> to 1554 cm<sup>-1</sup> and 1080 cm<sup>-1</sup>, respectively, which demonstrated the formation of H-bonds . The FT-IR of G and G-P(HEMA/BA/AA)- D/MBAA were compared. The amino  $(-NH<sub>2</sub>)$  and hydroxyl  $(-OH)$  groups shifted from 3550 cm<sup>-1</sup> to 3570 cm<sup>-1</sup>. This indicated that -NH<sub>2</sub> and -OH of gelatin (G) form hydrogen bonding with the polymer chain segments. Also, N-H of G shifted from 1554 cm<sup>-1</sup> to

1420 cm-1 , implying gelatin forms hydrogen bonding with other polymer chain of the system.



**Figure S2.** FT-IR of G-P(HEMA/BA/AA)-D/MBAA ionogel



**Figure S3.** (a) Tensile stress-strain curves of G-P(HEMA/BA/AA)-D/MBAA ionogel with different D content, (b) Young's modulus and toughness of G-P(HEMA/BA/AA)- D/MBAA ionogel with different D content.

#### **Rheological properties of G-P(HEMA/BA/AA)-D/MBAA ionogel**

As shown in Figure S3, the energy storage modulus (G') of the pristine ionogel and the post-soaking ionogel was higher than the loss modulus (G'') of that throughout the frequency range. And G' and G'' remained stable before and after soaking, which suggested that the network sturcture of ionogel could maintain stable after soaking for 5 days. Meanwhile, the dynamic energy storage modulus (G') and loss modulus (G'') were measured under different shear strain. As the ionogels continued to deform, G' and G'' would eventually intersect at one point, indicating that the ionogels transition from gel state to sol state due to the dissociation of hydrogen bonding and electrostatic interaction. It was obvious that the intersection point of the soaked ionogel was earlier than that of the original ionogel, which could indicate the soaked ionogel displayed weaker ability to maintain original state.



**Figure S4.** (a) Energy storage modulus and loss modulus of G-P(HEMA/BA/AA)-  $D/MBAA$  ionogel under different frequency ranging from  $10^{-1}$  to  $10^{2}$  rad/s, (b) energy storage modulus and loss modulus of G-P(HEMA/BA/AA)-D/MBAA ionogel under different deformation ranging from  $10^{-2}$  to  $10^{20}$ %.