Electronic Supplementary Information†

Fully-hydrophobic ionogel enables highly efficient wearable underwater sensor and communicator

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1. Materials

N,N-Dimethylacrylamide, (DMAA), tert-Butyl acrylate (t-BuA), hydrophobic silicon dioxide (SiO_2, ~15 nm), ethylene glycol dimethacrylate (EGDMA), 2, 2-azobisisobutyronitrile (AIBN) were purchased from Aladdin Industrial Co., 1-Butyl-3-methylimidazolium Bis(trifluoromethanesulfonyl)imide ([BMIm]TFSI), 1-Butyl-3-methylimidazolium tetrafluoroborate ([BMIm]BF_4) were purchased from Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences.

2. Preparation of the ionogel

Typically, the fully-hydrophobic ionogel Pt-BuA-[BMIm]TFSI was prepared by the following method: Firstly, 200 uL t-BuA, 1 uL EGDMA, 5 mg AIBN were dissolved in [BMIm]TFSI. Then a certain mass of hydrophobic SiO_2 NPs (0, 5, 10, 15, 20, 25, 30 mg) were dispersed in the above solution under ultrasound for 5 minutes, forming a precursor solution with various mass fraction of SiO_2 (0, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0 wt%). The Pt-BuA-[BMIm]TFSI ionogel was formed by thermal-initiation polymerization at 60 °C for 90 min.

The PDMAA-[BMIm]TFSI ionogel was prepared by the following method: 200 uL DMAA, 1 uL EGDMA and 5 mg AIBN were dispersed in 794 uL [BMIm]TFSI under ultrasound for 5 minutes to form

an uniform precursor solution, and the PDMAA-[BMIm]TFSI ionogel was formed by placing the precursor solution in oven at 60 °C for 90 min.

The Pt-BuA-[BMIm]BF_4 ionogel was prepared by the following method: Firstly, 200 uL t-BuA, 1 uL EGDMA and 5 mg AIBN were dispersed in 794 uL [BMIm]BF_4 under ultrasound for 5 minutes to form an uniform precursor solution, and the Pt-BuA-[BMIm]BF_4 ionogel was formed by placing the precursor solution in oven at 60 °C for 90 min.

The PDMAA-[BMIm]BF_4 ionogel was prepared by the following method: Firstly, 200 uL DMAA, 1 uL EGDMA and 5 mg AIBN were dispersed in 794 uL [BMIm]BF_4 under ultrasound for 5 minutes to form an uniform precursor solution, and the PDMAA-[BMIm]BF_4 ionogel was formed by placing the precursor solution in oven at 60 °C for 90 min.

3. Preparation of the fully-hydrophobic ionogel sensor

The fully-hydrophobic ionogel was obtained after soaking the 2 wt% SiO_2 based Pt-BuA-[BMIm]TFSI ionogel in deionized water for 3 days.

4. Characterizations

4.1. Mechanical measurement

The tensile stress-strain properties of the ionogel were measured by an electronic universal testing machine (Z1.0, Zwick, Germany). The strip samples, 2 mm in width and 1 mm in thickness, were carried out at the speed of 50 mm/min.

4.2. Ionic conductivity measurement

The ionic conductivity (σ) of the ionogel were measured by AC impedance technique. In this experiment, the cylindrical electrolyte samples were sandwiched between two stainless steel electrodes and firm contact was ensured. The measurements were carried out by an electrochemical workstation (CHI 660E, CH Instruments Co., Ltd) in the frequency range of 100 kHz to 10 Hz. The intersection of the curve at the real part is taken as the bulk resistance of the hydrogel electrolyte (R_b), and the ionic conductivity of the sample is calculated according to the following equation:

$$\sigma = \frac{L}{AR_b} \qquad \text{[Equation S1]}$$

where L is the thickness of the hydrogel electrolyte and A is the electrode area.

The ionic conductivity of soak solution was measured by a conductivity meter (FE38, METTLER TOLEDO).

4.3. Water contact angle measurement

The water contact angles were measured on the OCA25 Contact Angle Measuring System (Dataphysics, Germany). A 2 μL water droplet was carefully deposited ionogel surfaces using a syringe.

4.4. Swelling behavior measurement

The swelling behavior was studied by soaking the ionogels in deionized water and weighing the ionogel samples every day. The swelling ratio (δ) is calculated by the following equation:

$$\delta = \frac{W_s - W_0}{W_0} \qquad \text{[Equation S2]}$$

where Ws is the swollen weight and W_0 is the initial weight of the ionogels.

4.5. Adhesion force measurement

The adhesion force was measured by 90° peel-off method using an electronic universal testing machine (Z1.0, Zwick, Germany). The ionogel samples were prepared with film shape (2 cm in width, 8 cm in

length). Before test, the ionogel samples were adhered to various substrate (including glass, plastic, silicone, textile, steel, copper, nitrile rubber and skin) under a small pressure. Then the ionogel sample and substrate were transferred to air environment or underwater environment. Finally, the test samples were fixed by electronic universal testing machine and carried out at the peeling speed of 50 mm/min. For the long-term adhesion test, the ionogel adhered to silicone was soaked in water for 5 days before peeling. For the repeated adhesion test, the ionogel was adhered to skin.

4.6. Sensing performance measurement

The Real-time resistance (R) of the ionogel sensor was calculated the I−t curve, that was measured by an electrochemical workstation (CHI 660E, CH Instruments Co., Ltd) at a constant voltage of 1 V. The relative resistance ($\Delta R/R_0$) of the ionogel sensor is calculated by the following equation:

$$\Delta R/R_0 = \frac{R - R_0}{R_0} \quad \text{[Equation S3]}$$

where R_0 is the resistance of the ionogel under a strain of 0%.

The gauge factor (GF) is defined as follows:

$$GF = \frac{\Delta R/R_0}{\varepsilon} \quad \text{[Equation S4]}$$

Where ε is the strain of the ionogel sensor.

When the ionogel was soaked into salt solution, the GF value can be deduced through the following process:

$$R_0 = \frac{R_{water} R_{ionogel}}{R_{water} + R_{ionogel}} \quad \text{[Equation S5]}$$

$$\Delta R = \frac{R_{water} \lambda R_{ionogel}}{R_{water} + \lambda R_{ionogel}} - \frac{R_{water} R_{ionogel}}{R_{water} + R_{ionogel}} \quad \text{[Equation S6]}$$

$$GF = \frac{\Delta R/R_0}{\varepsilon} = \frac{\lambda - 1}{\varepsilon} \times \frac{1}{1 + \frac{\lambda R_{ionogel}}{R_{water}}} \quad \text{[Equation S7]}$$

Where R_{water} is the resistance of the soak solution, $R_{ionogel}$ is the resistance of the ionogel sensor under a strain of 0%, and the $\lambda R_{ionogel}$ is the real-time resistance of the ionogel sensor under the strain of ε.

5. Figures

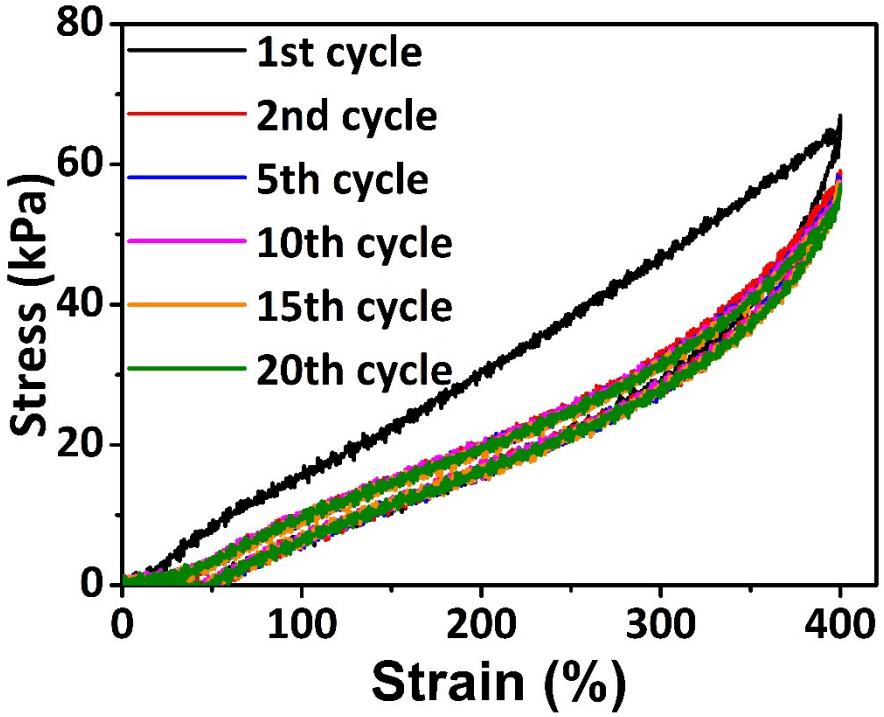

Fig.S1 Cyclic tensile property of the fully-hydrophobic ionogel. Cyclic tensile curves of the 2 wt% SiO_2 based Pt-BuA-[BMIm]TFSI ionogel at the speed of 50 mm/min.

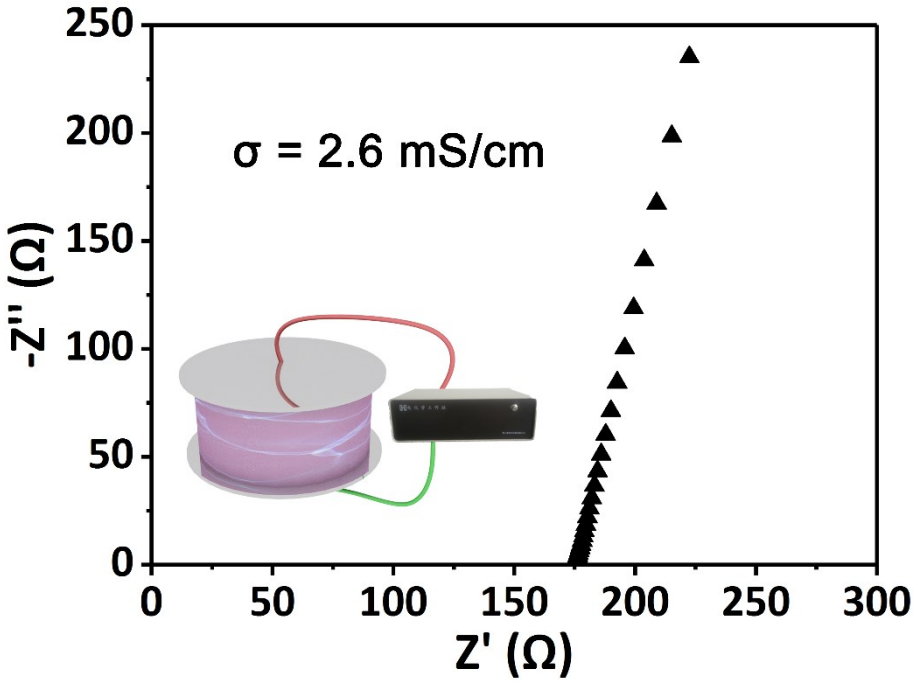

Fig.S2 Ionic conductivity of the fully-hydrophobic ionogel. Nyquist plot of the 2 wt% SiO_2 based Pt-BuA-[BMIm]TFSI ionogel after soaking for 3 days.

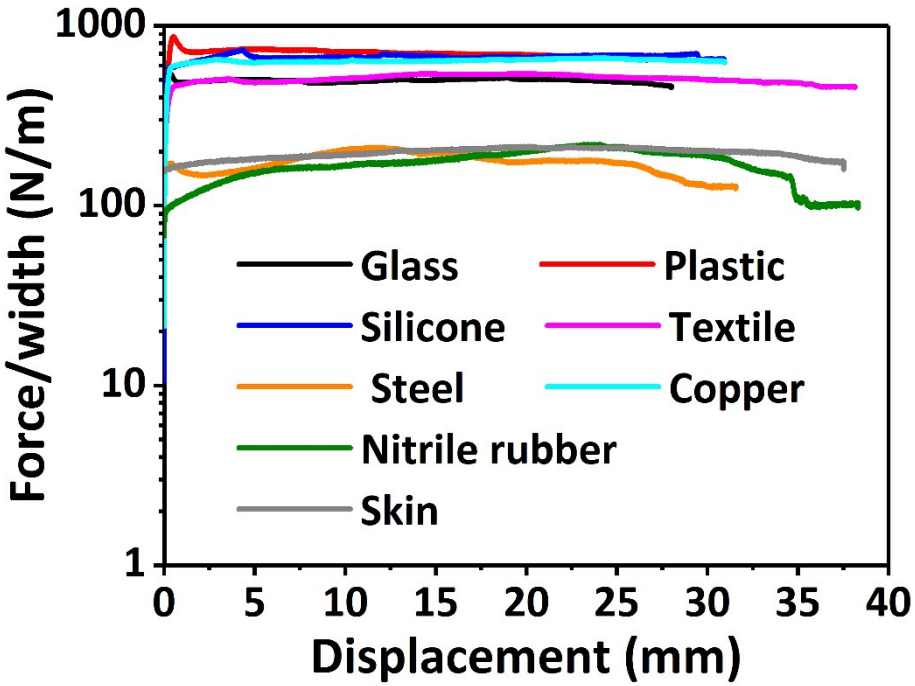

Fig.S3 Adhesion property of the fully-hydrophobic ionogel in air. Peeling curves of the 2 wt% SiO_2 based Pt-BuA-[BMIm]TFSI ionogel to various substrates in air.

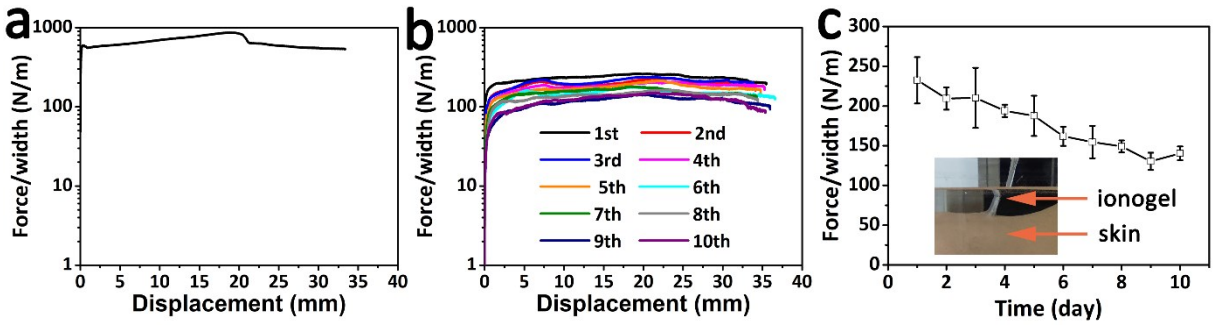

Figure S4 Adhesion performance under water. (a) Peeling curve of the ionogel adhered to silicone after soaking for 5 days. (b) Peeling curves of repeated adhesion for skin in the underwater environment. (c) Repeated adhesion ability of ionogel in the underwater environment.

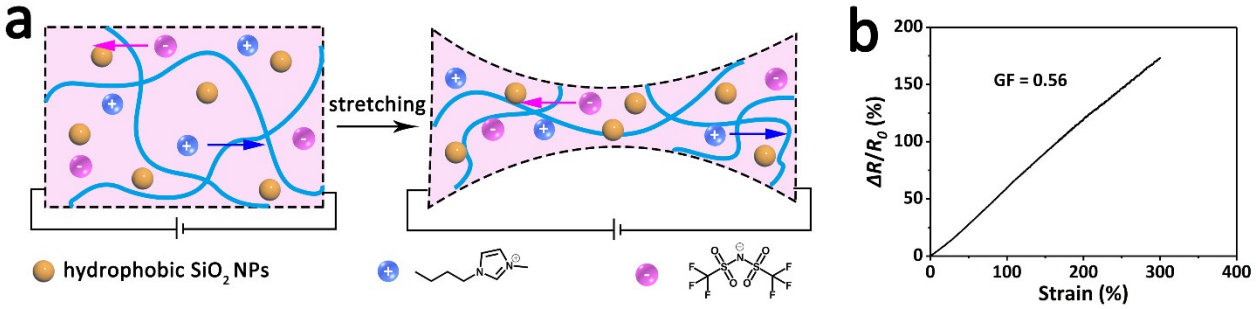

Fig.S5 Schematic of the SiO_2 based ionogel sensor during stretching process. (a) The aggregation of SiO_2 NPs within ionogel sensor. (b) Gauge factor of the ionogel sensor without SiO_2.

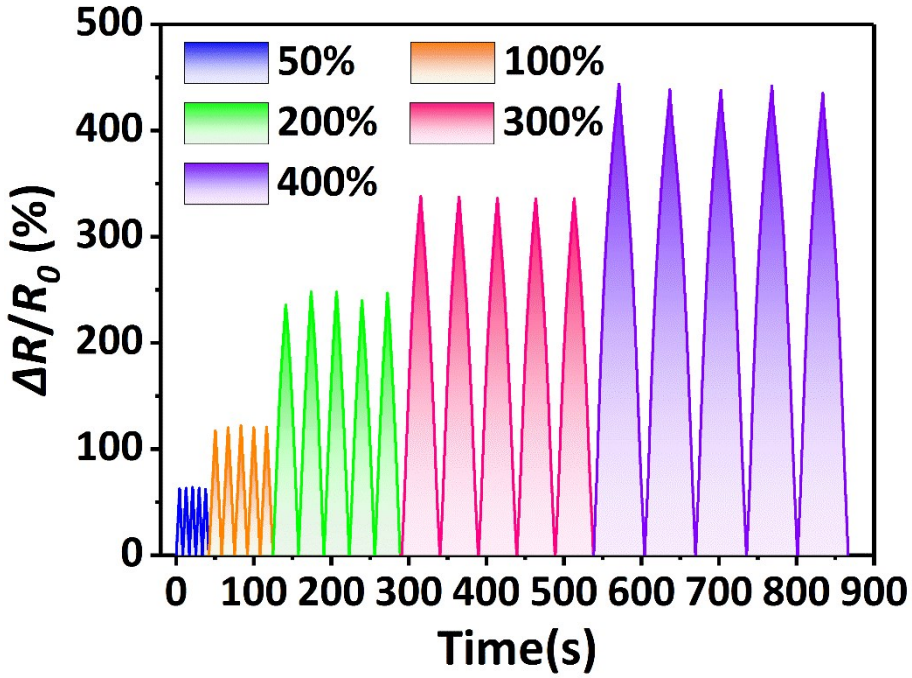

Fig.S6 Underwater sensing performances under large strain. Relative resistance variation of the ionogel sensor under large strains of 50%-400%.

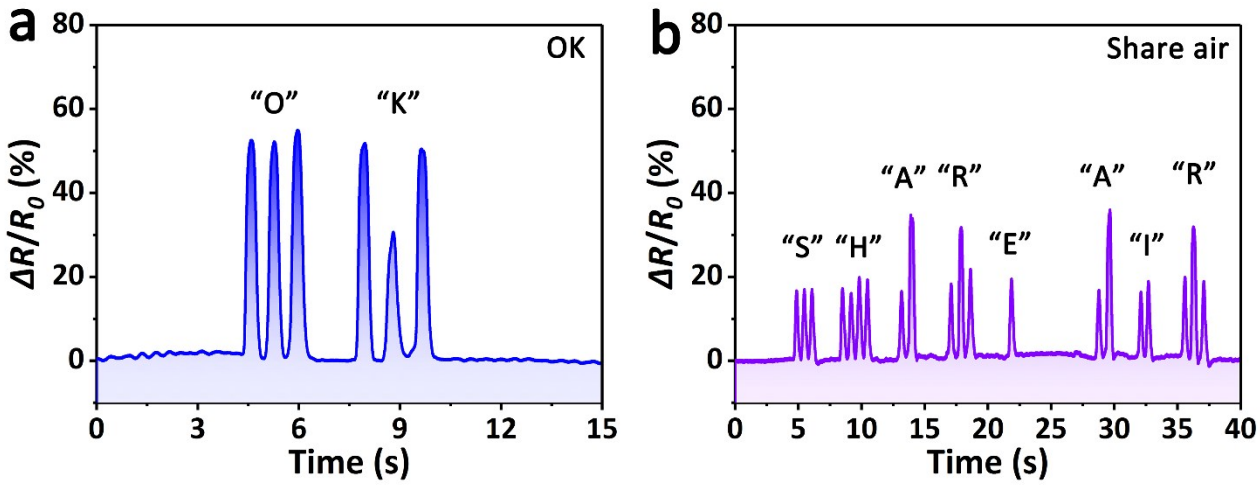

Fig.S7 Underwater communication application of the fully-hydrophobic ionogel sensor. (a) The sensing signal for the message of "OK". (b) The sensing signal for the message of "Share air".

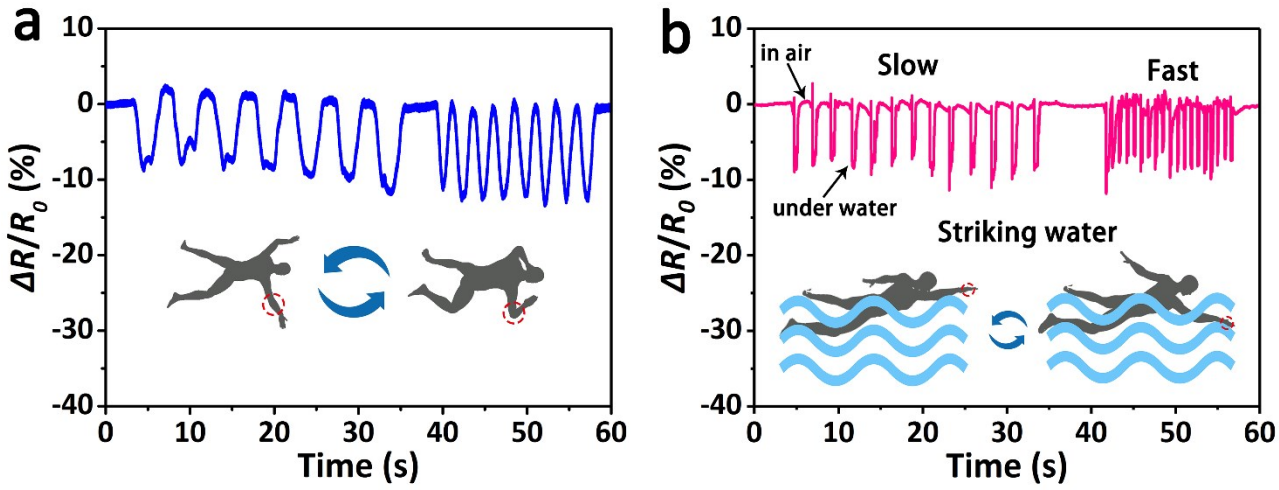

Fig.S8 Motion monitoring application of the fully-hydrophobic ionogel sensor. (a) Relative resistance variation of the ionogel sensor monitoring the arm movement during the swimming by fixed ionogel sensor on elbow. (b) Relative resistance variation of the ionogel sensor monitoring the striking water action during the simulated swimming by fixed ionogel sensor on the back of hand.

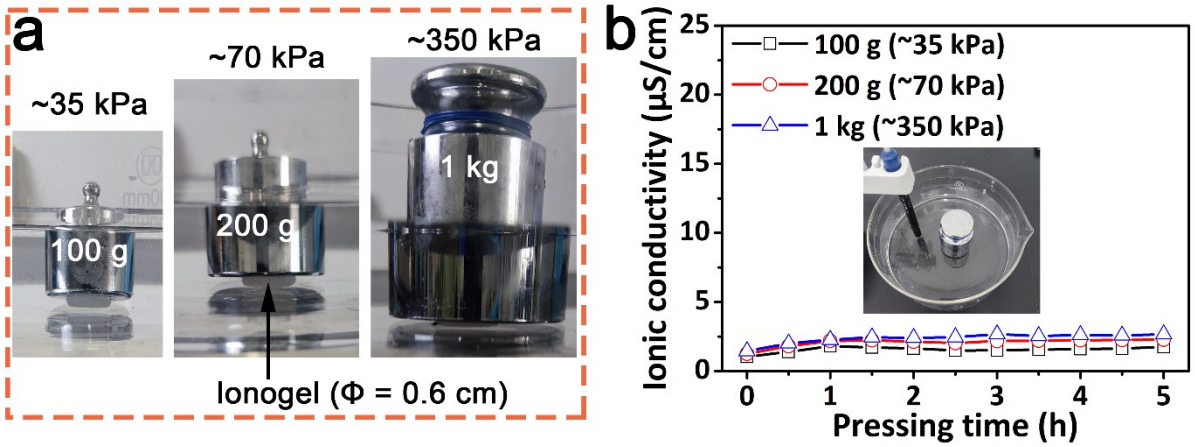

Fig.S9 Anti-leakage performance of ionogel in the simulated high-pressure environment. (a) Digital photos of the simulated high-pressure environment for ionogel in the underwater environment, the diameter of ionogel is 0.6 cm. (b) Ionic conductivity changes of water during the simulated high-pressure process with different pressure.