**Supporting Information**

# Quantitative observation of electric potential distribution of brittle polyelectrolyte hydrogels using microelectrode technique

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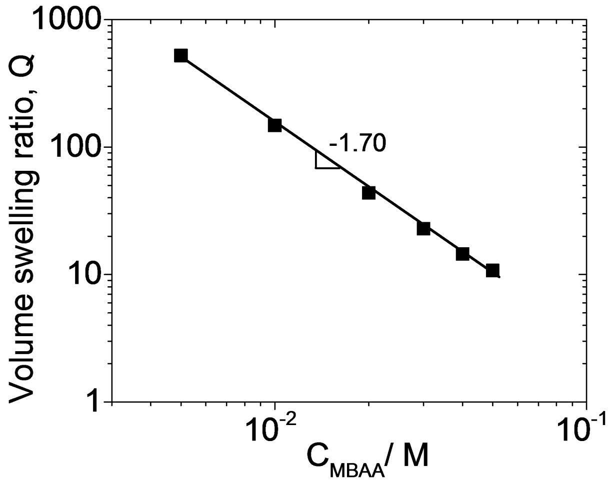



**Figure S1. Equilibrium volume swelling ratios *Q* of the PAMPS hydrogels synthesized with different cross-linking densities $C_{MBAA}$.** *Q* is in relative to the as-prepared state. Each data was the average of 3 measurements of length, width and height for a sample of different positions.

**Figure S2. Schematic diagram of microelectrode holder for TEM observation.**

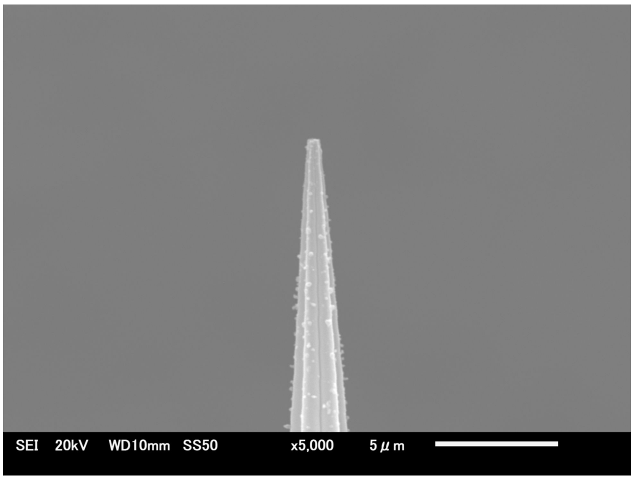



**Figure S3. Image of 7-barrel electrode observed by SEM.**

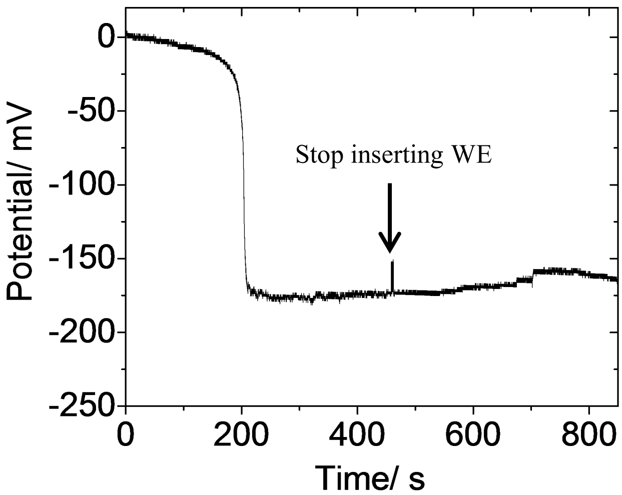
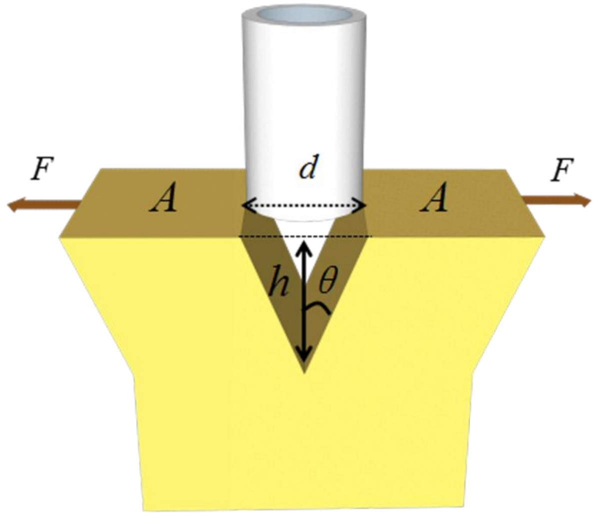



**Figure S4. Effect of dewelling of the microelectrode on the electric potential of hydrogel.** A PAMPS-4 gel was measured in $10^{-5}$ M KCl solution, the working microelectrode (WE) was inserted into hydrogel at a velocity of 395 nm/s for the initial inserting time of 450 s (corresponding to a depth of 100 μm); after that the inserting was stopped for the rest of the measurement of 390 s.




**Scheme S1. The relationship between crack length *h* and diameter *d* of microelectrode.**

As shown in Scheme S1, when a microelectrode was inserted into a brittle polyelectrolye hydrogel, a crack was generated at the tip of microelectrode. To simplify our discussion, we consider a crack with a triangle shape at the cross-section. The shear modulus $G$ can be defined by:

$$G \overset{def}{=} \frac{F/A}{\tan\theta} \qquad (1)$$

Here, $F$ is the applied force perpendicular to the microelectrode; $A$ is the deformed area with applying force $F$; $h$ and $\theta$ are the crack length and half angle of the crack, respectively. Therefore, the shear stress can be expressed by:

$$\frac{F}{A} = G \cdot \tan\theta = G\frac{d}{2h} = \frac{Ed}{4(1+\nu)h} \qquad (2)$$

Where $E$ is Young's modulus, $d$ is the crack width and $\nu$ is Poisson's ratio of elastic materials.

If the hydrogel was fractured and separated by microelectrode with outer diameter of $d$, the fracture energy $\Gamma$ is

$$\Gamma = \frac{F}{A} \cdot \frac{d}{2} \qquad (3)$$

By combining Eq. (2) and Eq. (3), we get the relationship between crack length $h$ and diameter of microelectrode $d$ as

$$h = \frac{Ed^2}{8(1+\nu)\Gamma} \qquad (4)$$