



Title	Quantitative Observation of Electric Potential Distribution of Brittle Polyelectrolyte Hydrogels Using Microelectrode Technique
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## **Supporting Information**

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

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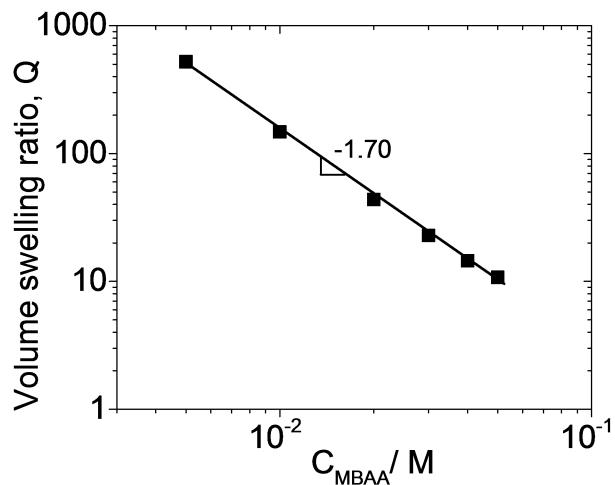
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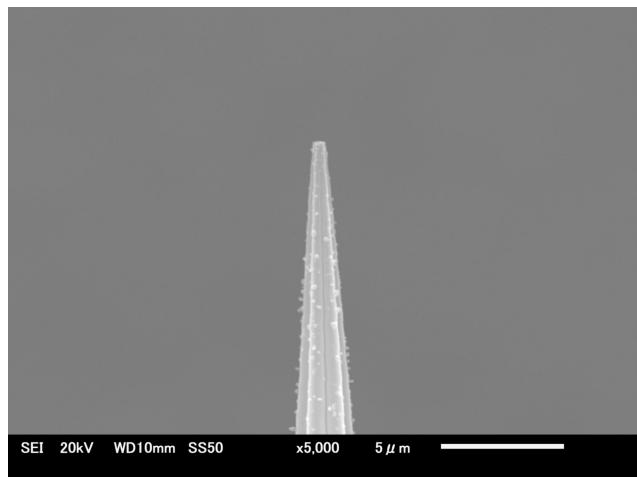
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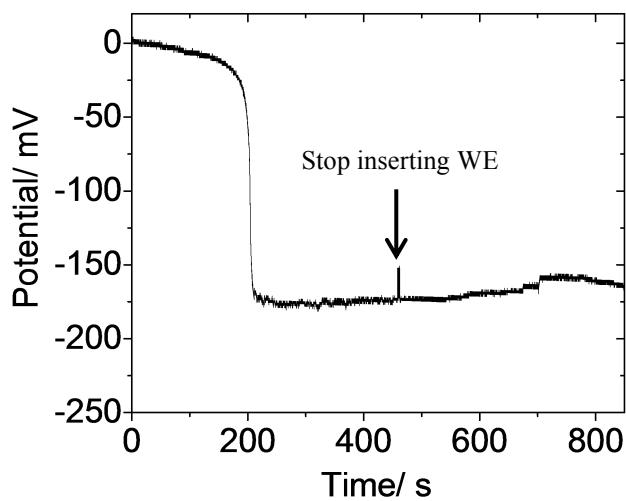
**Figure S1. Equilibrium volume swelling ratios  $Q$  of the PAMPS hydrogels synthesized with different cross-linking densities  $C_{\text{MBAA}}$ .**  $Q$  is in relative to the as-prepared state. Each data was the average of 3 measurements of length, width and height for a sample of different positions.



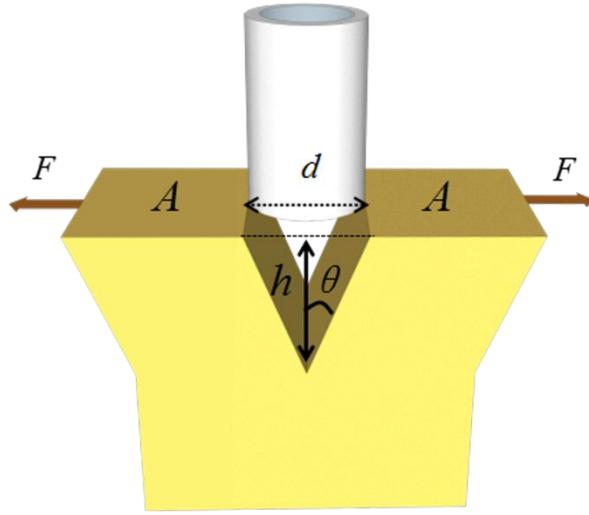
**Figure S2. Schematic diagram of microelectrode holder for TEM observation.**



**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  $\mu$ m); 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 polyelectrolyte 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:

$$G \stackrel{\text{def}}{=} \frac{F / A}{\tan \theta} \quad (1)$$

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:

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

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

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

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

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