

## **Supporting Information**

### **Porous Silica Nanospheres Functionalized with Phosphonic Acid as Intermediate-Temperature Proton Conductors**

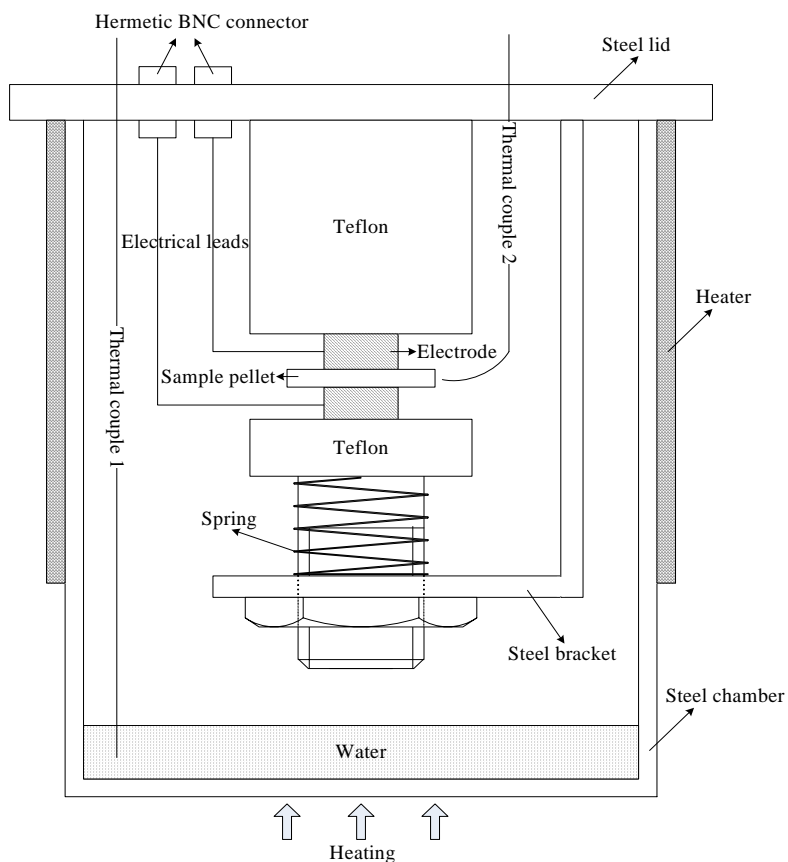
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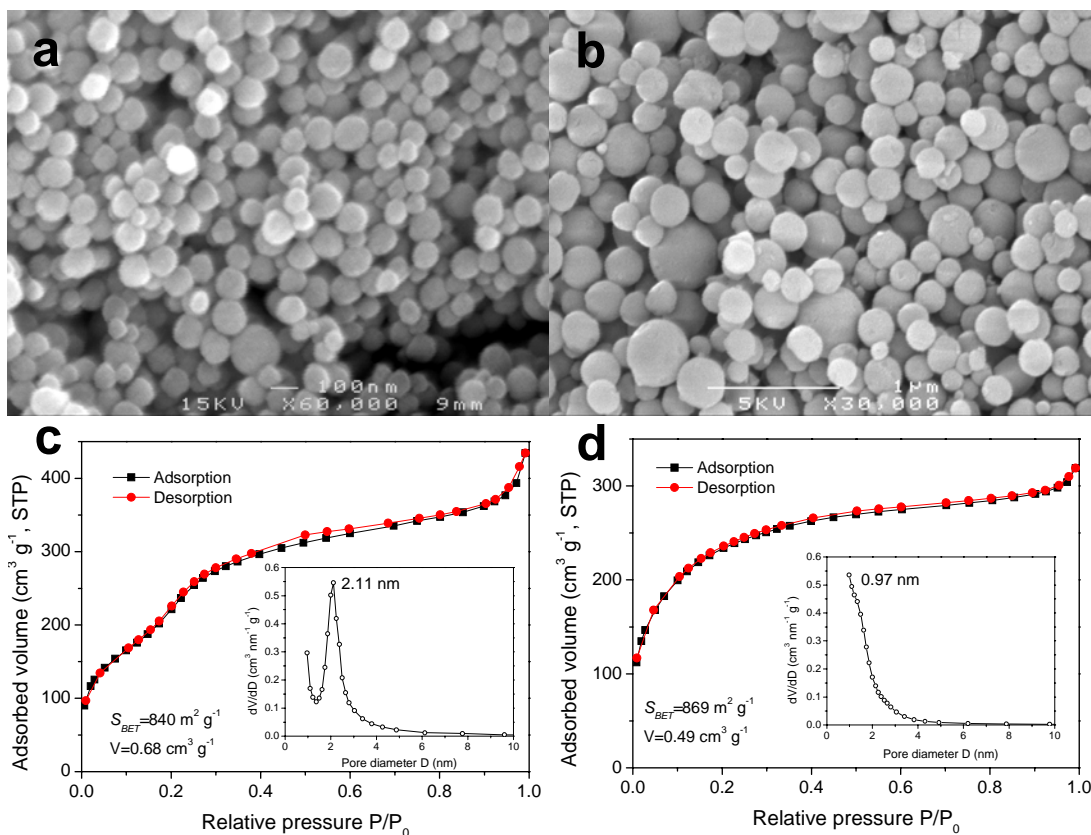
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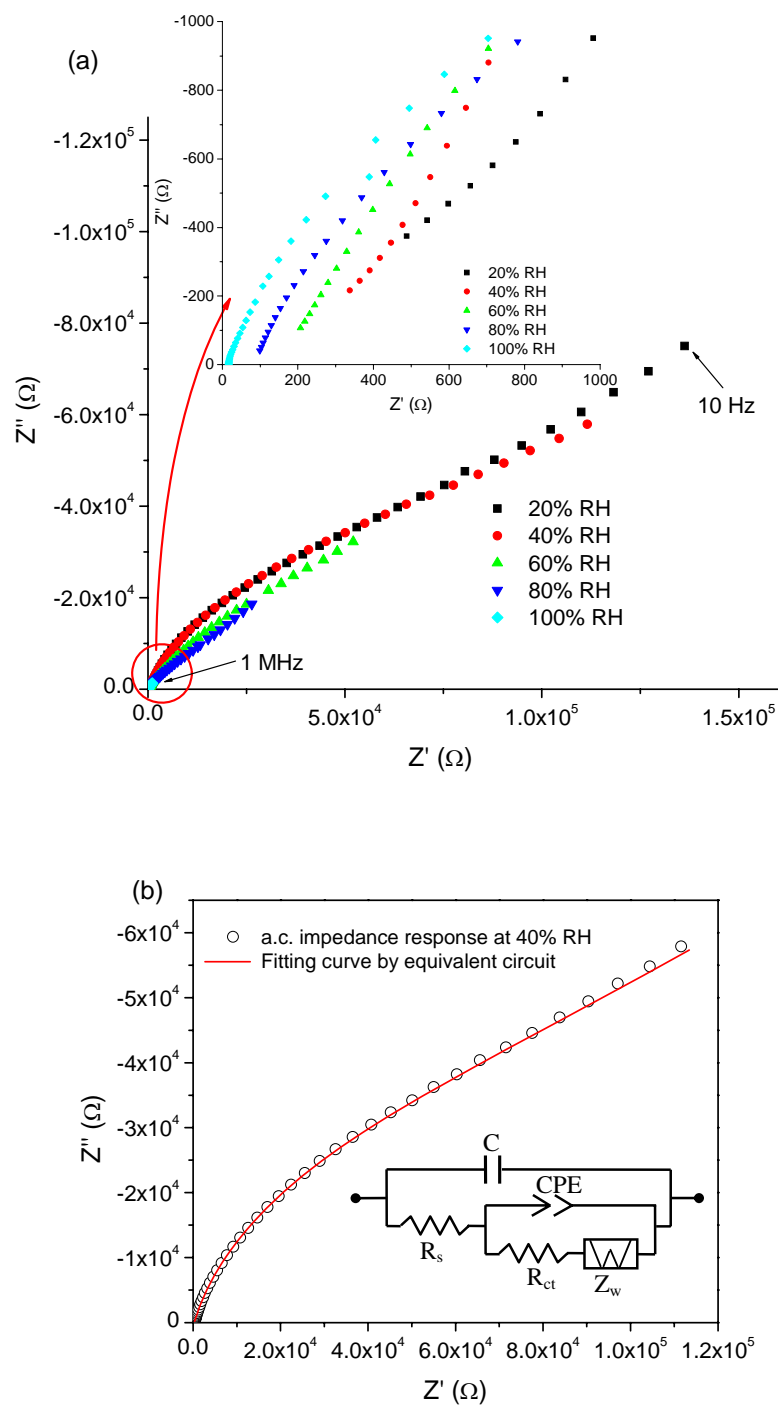
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**Scheme S1.** Scheme of the proton conductivity test cell. The relative humidity was varied by controlling the vapor pressure in the cell, which was generated by heating a small quantity of water on the bottom of the cell up to various temperatures. Water in the cell was stirred to keep homogenous temperature while heating. In addition, the temperature of the sample was independently controlled and measured adjacently to the pellet using another thermocouple.



**Figure S1.** SEM images (a,b) and nitrogen sorption isotherms (c,d) of the pressed samples pelletized with a pressure of  $150 \text{ kg cm}^{-2}$  (15 MPa) in the pellet preparation: (a,c) pressed sample NP20, and (b,d) pressed sample NP40. The insets in panel (c) and (d) represent the BJH pore size distributions of pressed NP20 and NP40. Their corresponding BET specific surface areas ( $S_{BET}$ ) and total pore volumes ( $V$ ) are listed in panel (c) and (d).



**Figure S2.** (a) Typical a.c. complex impedance spectra for NP40 measured at 130 °C and under various RH conditions. It can be seen that the high-frequency part of impedance spectra shifts to a lower resistance (the real part of a complex spectrum,  $Z'$ ) as the humidity is increased. (b) Example fitting curve of ac impedance response at 40% RH using an equivalent

circuit as shown in the inset. The equivalent circuit consists of a parallel resistor-capacitor pair ( $R_s$ -C) and a second  $R_{ct}$ -CPE (constant phase element) pair with a Warburg finite length element ( $Z_w$ ) reflecting the linear response at low frequencies. The resistor  $R_s$  represents the bulk resistance of the sample, and the second pair corresponds to the electrode/sample interfacial impedance, in which CPE is used to better present the depressed arc than the conventional RC element.