Supporting Information for

Biopolymer-Chitosan based supramolecular hydrogels as solid state electrolyte for electrochemical energy storage

Lujie Cao^{ab}, Mingyang Yang^{ab}, Dong Wu^a, Fucong Lyu^a, Zhifang Sun^a, Xiongwei Zhong^{ab}, Hui Pan^b, Hongtao Liu^c, and Zhouguang Lu^a*

Address:

^a Department of Materials Science & Engineering, South University of Science and Technology of China, Shenzhen, 518000, China

E-mail: luzg@sustc.edu.cn

^b Institute of Applied Physics and Materials Engineering, University of Macau, Taipa, Macau, China

^c School of Chemistry and Chemical Engineering

Central South University, Changsha 410083, Hunan, China

Keywords: Supercapacitor, Li/Na ion batteris, Supramolecular hydrogels, Solid state electrolyte, Chitosan

Electrochemical characterization

Electrochemical characterization of the the MnO₂/SHE/KBC asymmetric supercapacitor were carried out by cyclic voltammetry CV, electrochemical impedance spectroscopy EIS and galvanostatic charge-discharge studies. Specific capacitance and charge-discharge efficiency values for the ESs were also obtained from chronopotentiometric data recorded galvanostatically at a current density of 1.8 mA cm⁻² in the potential range between 0 and 1.6 V on a Neware (CT- 4008) cycler. The specific capacitances were evaluated from discharge curves of the charge–discharge plots using the equation $C = 2I\Delta t/(S \Delta V)$, in which I is the constant input current and Δt is the time required to change the potential by ΔV . The chargedischarge efficiencies η of the ESs were calculated using the equation $\eta = t_d \times 100/t_c$, in which t_c and t_d are the respective times required for charging and discharging the electrochemical supercapacitors. All the electrochemical measurements except the temperature dependence of ionic conductivity were conducted at room temperature. The electrochemical impedance spectrum (EIS) were measured on an electrochemical station (VMP3, Bio-Logic). The bulk ionic conductivity of the supramolcular hydrogel electrolytes were determined from the complex impedance spectra in the frequency range between 200 Hz and 1 MHz with a perturbation of 5 mV by using the equation $\sigma = L/RA$, in which L, A, and R are the thickness, area, and bulk resistance calculated from high-frequency intercept on the real impedance axis of the Cole-Cole plot of the MnO₂/SHE/KBC asymmetric supercapacitor, respectively. In our experiment, the L is 0.1 cm in thickness and the S is 1.1 cm². The activation energy E_a is determined by the equation $\sigma = \sigma_0 / [KT^* exp(-E_a/KT)]$.



Figure S1 The process of preparation of solid polymer superamolecular hydrogel electrolyte (SHE).



Figure S2 The prepared SHE.



Figure S3 The SEM image of cathode of MnO₂-CNT composite.



Figure S4 The SEM image of anode of KBC(ketjen black carbon)-CNT composite.



Figure S5 The capacitance retention of MnO₂/SHE/KBC electrochemical supercapacitor.



Figure S6 CV curves under different temperature at 100 mV/S (up) and 200 mV/s (down) scan rate.



Figure S7 The EIS spectra of MnO₂/SHE/KBC asymmetric supercapacitor before (black curve) and after (red cureve) 10000 charge-discharge cycles at 303 K.



Figure S8 Schematic representationofpolymer-network hydrogelscross-linked by ultrafast complexation of metal ions and chitosan chains in water (A). Chemical structures of chitosan and their inter-woven networks driven by the complexation between metal ions and -OH and -NH₂ groups in the chitosan chains (B).



Figure S9 The SEM and EDAX mapping of the dried hydrogel with element C, N, O and Ag.



Figure S10 The XPS curve of the dried CS-Ag⁺ hydrogel.



Figure S11 The XRD patterns of hydrogel before and after 10000 charge-discharge cycles.