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Electronic Supplementary Information

Electrolyte imprinted graphene oxide-Chitosan chelate with copper crosslinked composite electrodes for intense cyclic stable flexible supercapacitors

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1. Materials used

Chitosan, Copper chloride di hydride ($CUCl_2.2H_2O$), 1-butyl-3-methyl imidazolium tetrafluoroborate ($BMIMBF_4$,) ionic liquid, N, N-dimethylformamide, sulfuric acid (H_2SO_4), Phosphoric acid (H_3PO_4) acetic acid, were purchased from Sigma Aldrich and also, the ITO-PET film with surface resistivity 100 Ω/sq was supplied by Sigma Aldrich. Ethanol was provided by Samchun chemicals.

2. Preparation of graphene oxide-chitosan-copper composite

First graphene oxide was prepared from W. S. Hummers and R.E. Offeman method using graphite[1] and then equal 1.0 Wt% of prepared graphene oxide (GO), chitosan, $CUCl_2.2H_2O$ and 1% of $BMIMBF_4$ ionic liquid were added in a Teflon-lined stainless steel autoclave. The well-sealed autoclave was placed in hydrothermal reactor unit (Hanwoul eng. Reactor Control System). The reaction temperature was set at 100°C for 24h. After that, the autoclave was allowed to cool gradually and to collect an obtained gel-like composite materials. This gel was cast on the surface of ITO-PET film (the ITO-PET film was pre-cleaned before casting process using acetone, ethanol and D.I. water under sonication[2]). This film material was allowed to heat at 120°C for the removal of absorbed moisture and other impurities. Similar types of experiments were conducted through adjusting the temperatures as follows 50, 75, and 100°C and these electrodes were labeled as CC1, CC2, and CC3 respectively.

3. Characterization techniques

The crystallinity of prepared composites were studies by XRD patterns, which were analyzed on a Powder X-ray diffraction of XPERT-PRO X-ray diffractometer with Cu-Kα anode radiation (λ=1.5418 Å) in the 2θ range of 10-90°and with a scan rate of 2°/min. The XPS spectra carried out by Theta Probe AR-XPS System (Thermo Fisher Scientific-U.K; X-ray source: monochromated Al Ka, hv=1486.6eV; X-ray energy: 15kv, 150W; Spot size: 400µm). Surface morphology of materials such as SEM analyzed by FE-SEM- Hitachi: S-4200 and TEM examined by JEOL TEM: JEM 2011 (Vac : 200 kV; CCD camera 4k x 4k -Ultra Scan 400SP). All the electrochemical analyses were done using BioLogic-SP150 workstation with two electrode setup.

4. Fabrication of high-performance supercapacitors

The CC1, CC2, and CC3 composite electrode supercapacitors were fabricated as symmetric type capacitors with a size of 5 cm x 2.5 cm of composite coated ITO-PET flexible electrodes. The bare graphene oxide and chitosan were also fabricated for comparison using the same condition under the ionic liquid electrolyte medium. The other composite lectrodes are systematically imprinted with BMIMBF$_4$ electrolyte. The Whatmann filter paper was used as a separator. All the electrochemical performances were analyzed BioLogic SP-150 electrochemical workstation.

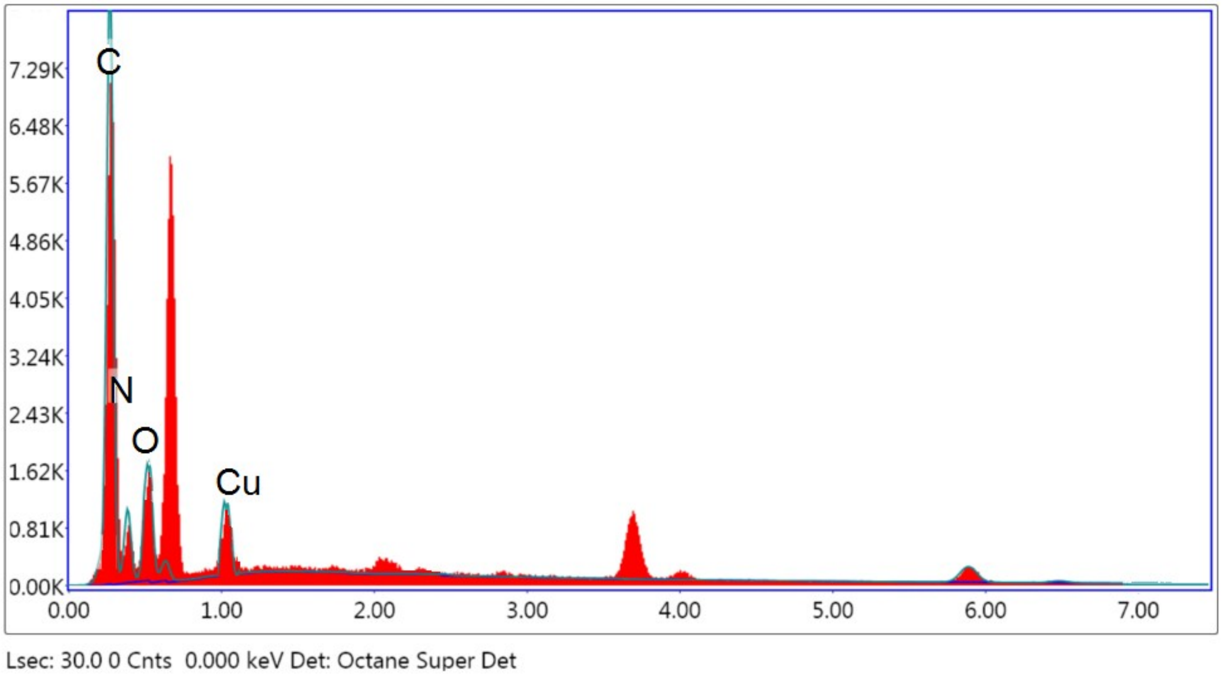

Fig. 1. EDX analysis of graphene oxide-chitosan-cupper crosslinked composite.

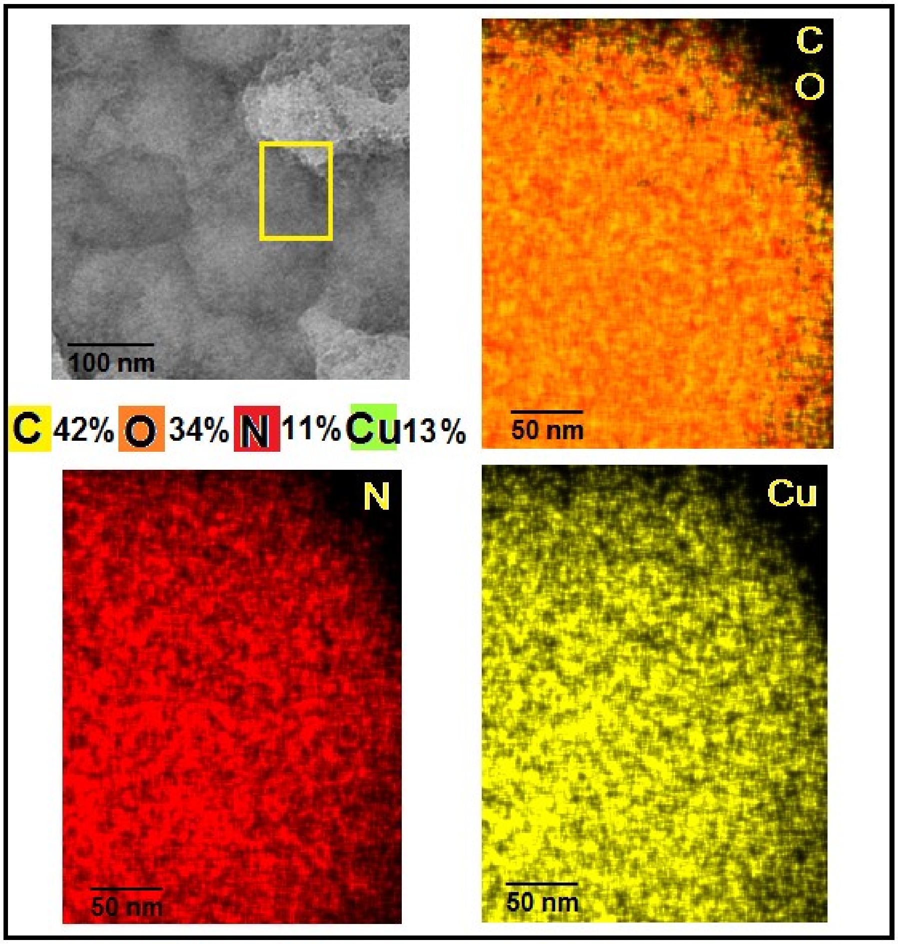

Fig. 2. STEM-EDS images of graphene oxide-chitosan-cupper crosslinked composite.

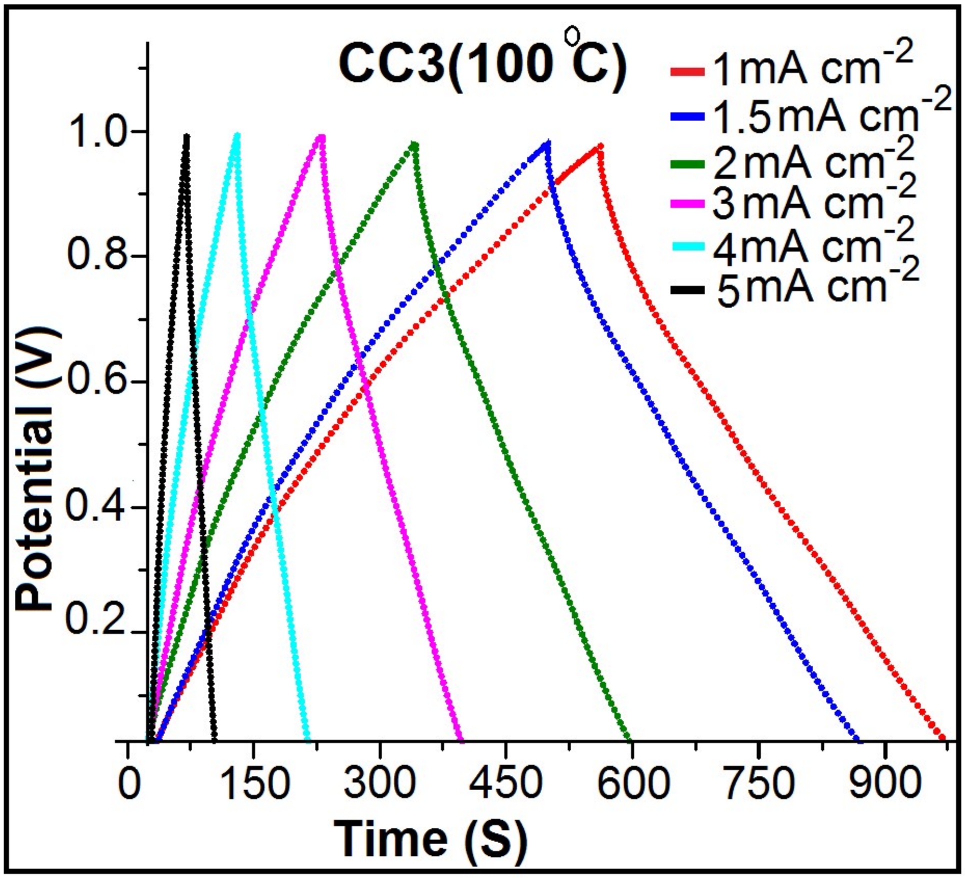

Fig. 3. Charge-discharge profile CC3 supercapacitor at increased current density.

Table. 1. Specific capacitance, Energy density and Power density performances of CC1. CC2, and CC3 supercapacitors.

Electrodes	**Specific capacitance (Fg^{-1})**	**Energy density (Wh Kg^{-1})**	**Power density (Wkg^{-1})**
CC1 (50°C)	356	178	1200
CC2 (75°C)	273	137	1198
CC3 (100°C)	175	88	905

Calculations

Capacitance could also be calculated from the galvanostatic discharge curves, using the following equation 1.

$$C = \frac{I\Delta t}{m\Delta V} \quad .....1$$

Where *I* (A) is charge or discharge current, Δ*t* (s) is the time for a full charge or discharge, *m* (g) indicates the mass of the active material, and Δ*V* represents the voltage change after a full charge or discharge.

The energy density (E) calculated by equation 2 and power density (P) were calculated by equation 3.

$$E = \frac{C(\Delta V)^2}{2} \quad ......2$$

$$P = 3600\frac{E}{\Delta t} \quad ......3$$

Where C is the specific capacitance of the active materials, and ΔV is the potential window of discharge.

The equivalent series resistance (ESR) and columbic efficiency (ɲ) were calculated by following equation 4. [3, 4]

$$ESR = \frac{V_{IR}}{2I}\Omega \quad ....4$$

Where, ESR is the equivalent series resistance, V_{IR} is the IR drop from charge-discharge current and I is the current.

References

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