Construction of binder-free PANI-CQD-Cu electrode by electrochemical

method for flexible supercapacitor applications

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1. Material Characterization

CQD absorbance and fluorescence behaviour are investigated by JASCO NIS 670 (liquid) UV-Visible spectrophotometer and JASCO FP-8200 Spectro fluorometer (200 to 800 nm). The 1 mL CQD solution was diluted in 100 mL of deionized water. Then, 2mL of diluted CQD was used for analysis. The functional group was analysed by JASCO 4600 Fourier transform infrared spectrometer (FT-IR) in Attenuated Total Reflectance (ATR) Accessories mode (Resolution: 0.7 cm^{-1}) the range is 400-4000 cm⁻¹. The surface morphology of the CQD is identified by the high-resolution transmission electron microscope (HR-TEM)-JEOL-2100.The Zeta potential of CQD was investigated with the help of Dynamic Light Scattering (DLS) analysis. The phase and crystal structure of CQD and polymer electrodes were investigated by X-ray diffraction spectrometer-XRD (XRD - X' Pert Pro – PANalytic; source - Cu K α , wavelength - 1.54 Å, scan step time -10.7950s, scan step size-0.0500°). The functional groups were identified by XploRATM PLUS Raman Spectrometer - Confocal Raman Microscope (wave number -100 cm⁻¹ to 4000 cm⁻¹, wavelength range - 200-1050 nm, spot size-20X (NA=0.4 WD=1.3 mm), scan step size – 10 nm, power-reflection (LED eqv 100 W)/ transmission (Halogen 30 W), repeatability-1 μ m; resolution - 100 nm, FLAT fluorescence subtraction). The Raman data was analyzed by LabSpec6 spectral software. The element composition was analyzed by X-ray photoelectron spectroscopy (XPS) spectroscopy (PHI - VERSAPROBE III instrument using Al Ka- 1486.6 eV). The prepared electrodes surface morphology was investigated by Field emission scanning electron microscopy (FESEM-SU08010) equipped with energy dispersive X-ray analysis (EDAX), the accelerating voltage – 0.1 ~ 30 kV and current 300 μ A, photo magnification ×20 ~ ×800,000 (Photo magnification) × 60 ~× 2,000,000 (Display magnification), Resolution-1.0 nm@ 15 kV /1.3 nm @1 kV. The surface area and porous nature were investigated by Brunauer-Emmett-Teller (BET) techniques using the Nova 2200e model.

2. Electrochemical Characterization

Electrochemical analysis cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS) and charge-storage performance galvanostatic charge-discharge (GCD) were evaluated by Metrohm electrochemical analyzer MULTIAUTOLAB-M204 (NOVA software) using a three-electrode system. The CC, CC coated PANI, PANI-CQD, PANI-Cu and PANI-CQD-Cu electrodes, Ag/AgCl and platinum electrode were using working, reference and counter electrodes, respectively.1 M H_2SO_4 used as an electrolyte. The EIS was done by open potential with 10 mV of AC amplitude and the frequency range was 0.01 Hz -100 kHz. The GCD measurement was carried out within the potential range of -0.2-0.8 V for the three-electrode system and the cyclic stability was investigated by the GCD measurement at the current density at 5 mA cm⁻² (1 A g⁻¹). PVA- H_2SO_4 gel was used as an electrolyte for asymmetry device fabrication. Before the electrochemical test, the mass of the active material is measured by analytical semi-micro balance.



Scheme S1. The photograph and SEM image of carbon cloth and PANI-CQD-Cu coated carbon cloth.



Fig. S1. EDAX spectra for the CC, PANI, PANI-CQD, PANI-Cu and PANI-CQD-Cu electrode materials.



Fig. S2. Comparative Ragone plot—areal energy density (μ Wh cm⁻²) vs. areal power density(mW cm⁻²) of AC/PVA-H₂SO₄/PANI-CQD-Cu flexible device with other PANI-Carbon based flexible supercapacitor device.