Electronic Supplementary Material (ESI) for RSC Advances.

Designing binder-free, flexible electrodes for high-performance supercapacitor based on pristine carbon nano-onions and its composite with CuO nanoparticles

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Supporting Information

S1 Fabrication of free-standing, flexible and binder-free electrodes

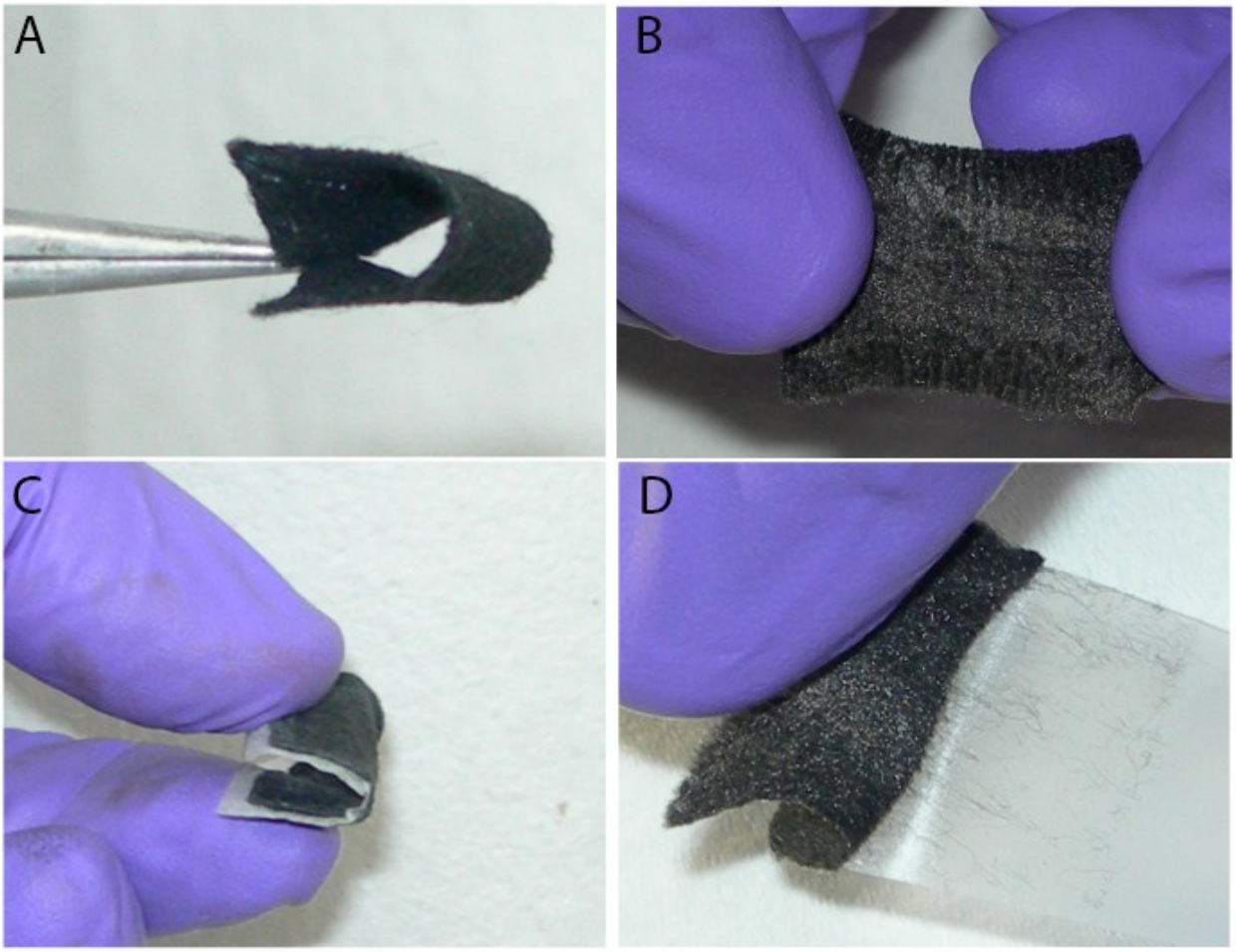

Fig. S1 Photograph demonstrating the composite electrode **(A)** flexibility **(B)** mechanical strechability **(C)** complete foldable device and **(D)** adhesion test.

The adhesion tests were performed by the standard scotch tape test (Fig. S1D). During the peel off only fibers in the cotton wipe came out, and there was no trace of active material particles on the scotch tape. This indicates a strong adhesion between the fibers of the wipe and the CNO and CNO-CuO particles. The strong adhesion of active materials to the fiber was also confirmed by repeatedly washing the electrodes in water. Even after frequent washing in DI water, the composite particles adhere well to the wipe without dissociating, and no precipitation was observed in water. These tests suggest that CNO-CuO composite particles adhered very strongly to the wipe fibers, which is critical for wearable electronic and power devices. It is essential to note here that binder and surfactants were not used in the electrode preparation. Such strong adhesion is due to the large Van der Waals forces and hydrogen bonding between CNOs and the wipe textile fibers. Superior mechanical adhesion of CNOs and composite nanoparticles on cotton is essential for high-speed roll-to-roll fabrication and stability of the energy storage device.[28]

S2 Morphology of CNO

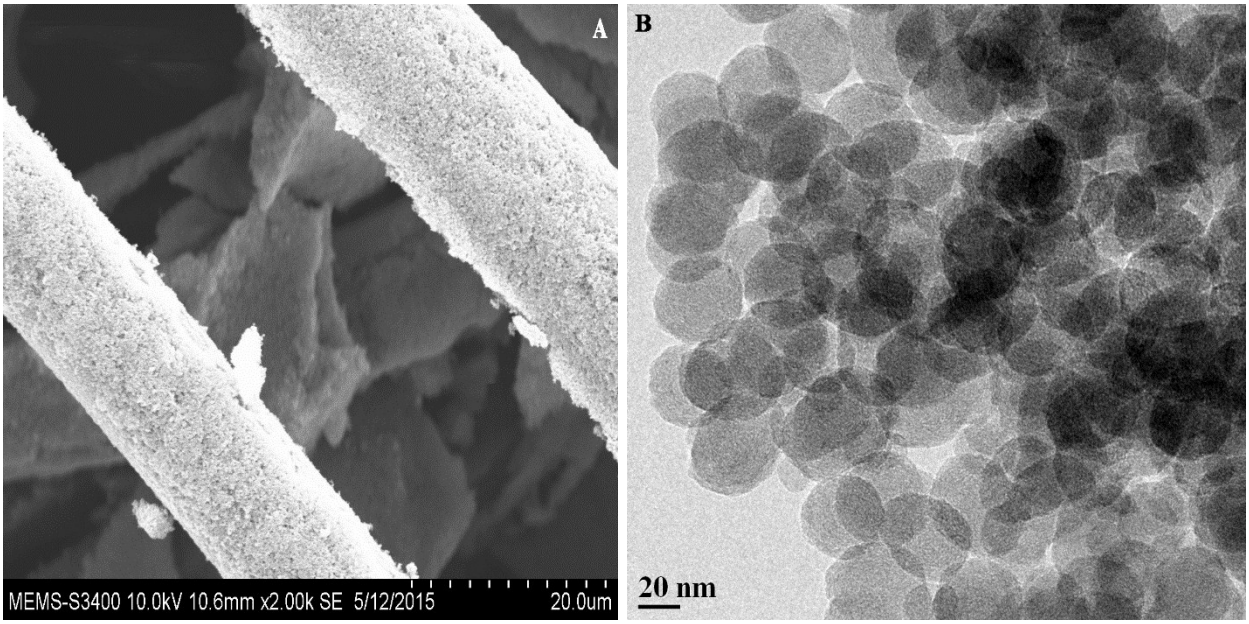

Fig. S2 SEM images showing the cotton wipe (A) with conformal CNO coating and (B) TEM overview of CNOs.

S3 Mechanical conditions of the electrodes after charge-discharge process

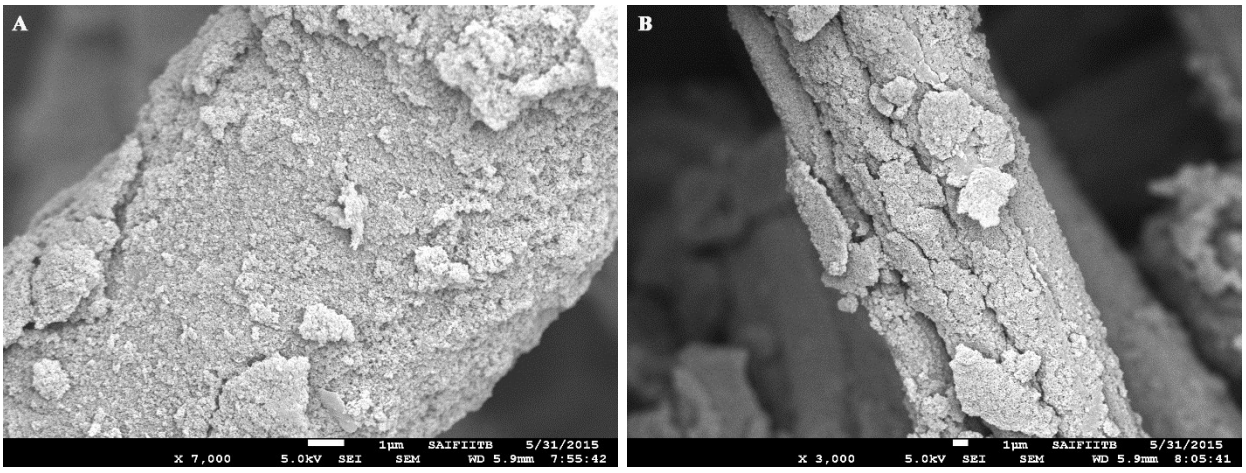

Fig. S3 FESEM images showing the morphology of **(A)** CNO and **(B)** CNO-CuO composite electrodes after 5000 charge-discharge cycles.

S4 N_2 adsorption/desorption isotherms and pore size distribution of CNO and CNO-CuO composite.

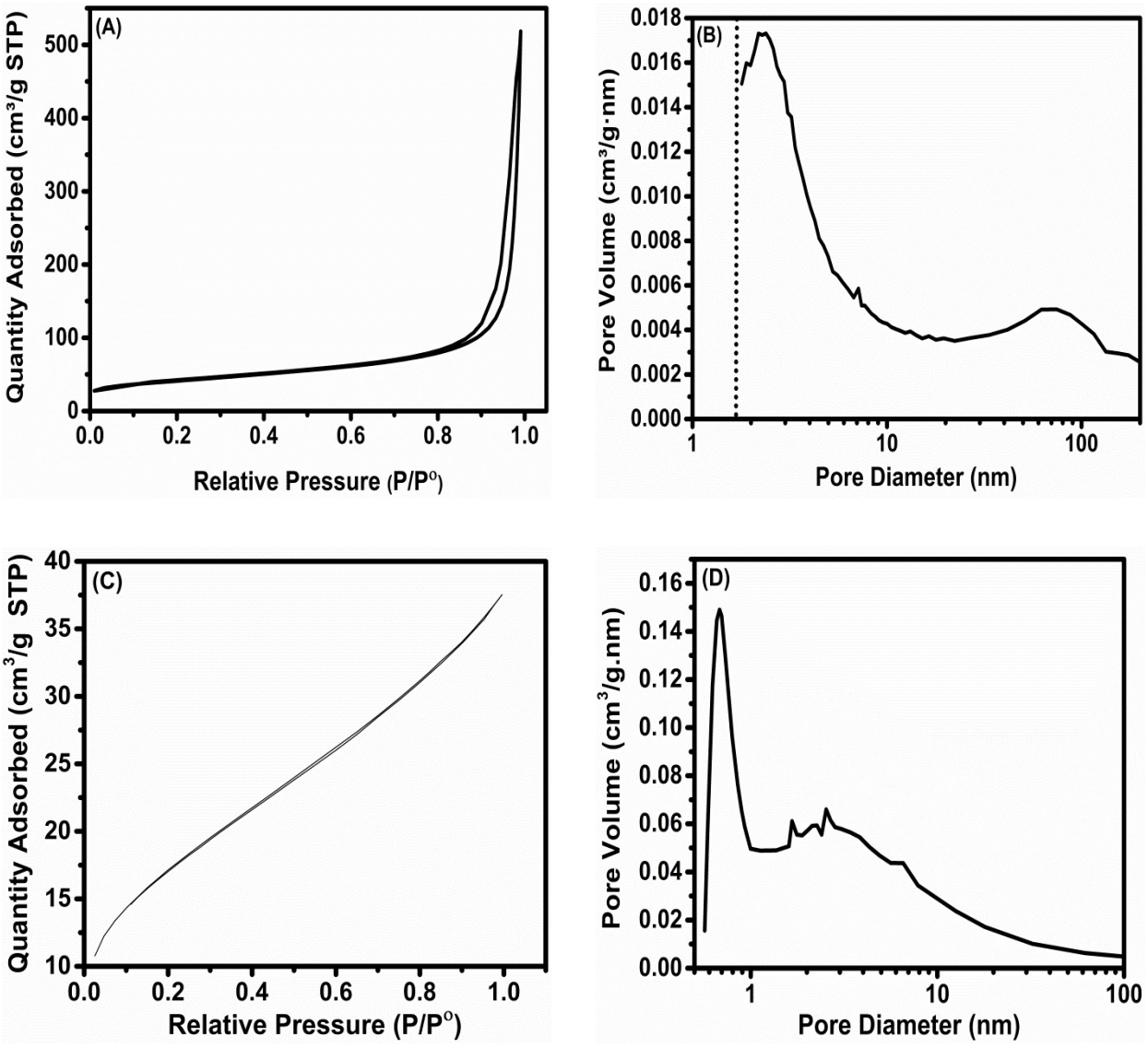

Fig. S4 (A) Nitrogen sorption isotherm of CNO **(B)** Pore size distribution curves of CNO calculated by BJH method **(C)** Nitrogen sorption isotherm of CNO-CuO composite and **(D)** Pore size distribution curves of CNO-CuO composite calculated by BJH method.

S5 Synthesis and morphology of CuO nanoparticles

CuO nanoparticles were prepared by a hydrothermal reaction. 25 mg of copper acetate monohydrate was added to 50 mL of distilled water and sonicated for 1 h. It was then transferred to a Teflon-lined stainless steel autoclave and heated for six hours at 150 °C. After completion of the chemical reaction, the former black suspension was washed and centrifuged several times. After washing, the precipitate was oven dried for 8 h at 70 °C.

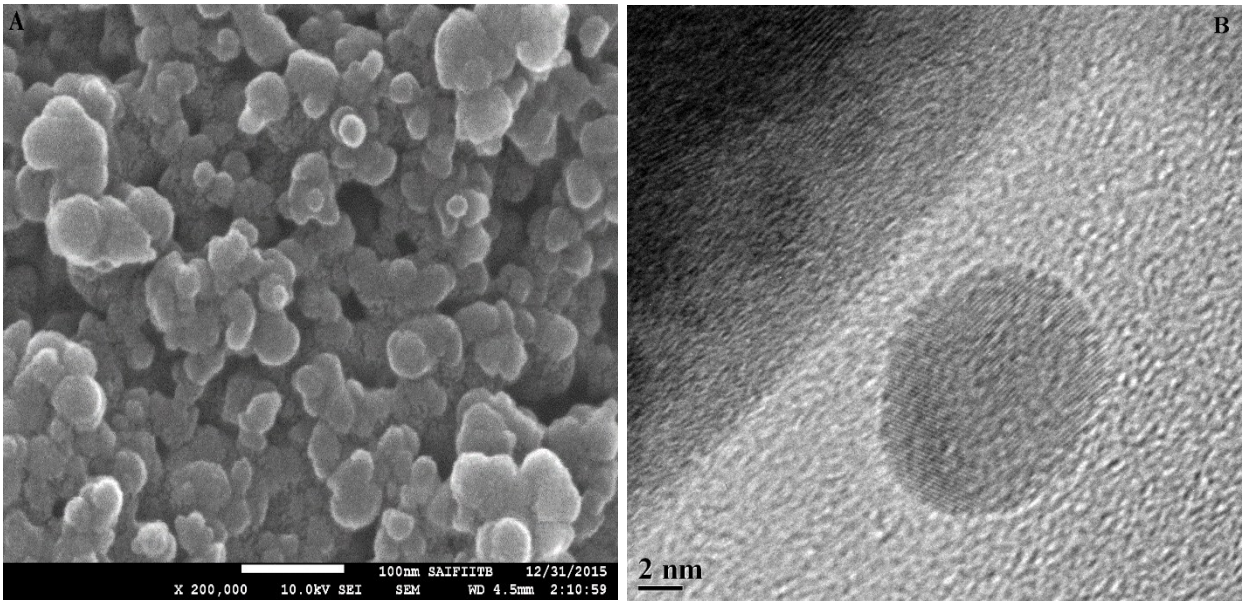

Fig. S5 (A) FESEM image showing the morphology of CuO nanoparticles **(B)** HRTEM image of CuO nanoparticles.

The accurate particle size and morphologies of CuO nanoparticle were confirmed by FESEM and HRTEM images. Fig. S5 (A) and (B) show the morphology of highly crystalline CuO nanoparticles from SEM and TEM analyses, respectively. The SEM image clearly showed agglomeration of CuO nanoparticles with spherical morphology. It showed the presence of agglomerated nanospheres with an average diameter of 10–20 nm. Therefore, from this observation only the rough morphology was found. Nevertheless, the accurate sizes and

morphology of the nanoparticles can be estimated from the TEM analysis. HRTEM images of CuO nanoparticles presented in Fig. S5B revealed spherical morphology of the synthesized nanoparticles with the crystallite size of around 10 nm.