

## Supporting Information

### Large-Scale Synthesis of Uniform Cu<sub>2</sub>O Nanocubes with Tunable Sizes by In-Situ Nucleation

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#### Experimental section

##### Synthesis

A method of controlling the number of Cu<sub>2</sub>O seeds in solution is demonstrated to synthesize Cu<sub>2</sub>O nanocubes with various sizes. A larger quantity of Cu<sub>2</sub>O seeds yields smaller Cu<sub>2</sub>O cubes. The amount of Cu<sub>2</sub>O seeds was tuned by controlling the copper hydroxide (Cu(OH)<sub>2</sub>) concentrations. The formation of Cu(OH)<sub>2</sub> was reduced that a portion of Cu<sup>2+</sup> ions were chelated by trisodium citrate. In this work, all chemicals were purchased from Aldrich Company and directly used without further purification. The Cu<sub>2</sub>O nanocubes were synthesized from copper sulfate (CuSO<sub>4</sub>, 99%, Aldrich), sodium hydroxide (NaOH, 99%, Aldrich), trisodium citrate dihydrate (C<sub>6</sub>H<sub>5</sub>Na<sub>3</sub>O<sub>7</sub> · 2H<sub>2</sub>O, 99%, SHOWA), and ascorbic acid (C<sub>6</sub>H<sub>8</sub>O<sub>6</sub>, 99.7%, J.T.Baker). In a typical procedure, a round-bottom flask was filled with 400 mL of deionized water and various amounts of trisodium citrate and placed in a water bath that was maintained at 20 °C for 20 min with vigorous stirring. Then, 1 mL of 1.2 M CuSO<sub>4</sub> solution was rapidly injected using a pipette. After 5 min, 1 mL of 4.8 M NaOH solution was injected into the solution. The clear blue solution immediately turned turbid blue, indicating the precipitation of Cu(OH)<sub>2</sub>. After another 5 min, 1 mL of 1.2 M ascorbic acid (AA) was injected as a reducer and the solution was maintained in a water bath for another 30 min. The color of the solution rapidly turned from turbid blue to yellowish brown. The concentration of Cu<sup>2+</sup>: citrate: OH<sup>-</sup>: AA in the final solution was 3 mM: x mM: 12 mM: 3 mM (1: X:

4: 1, where  $X = 0.25, 0.50$  and  $0.75$ ). The product was filtered and washed three times with DI water and alcohol. It was then dried for 2 h in an oven that was maintained at  $50\text{ }^{\circ}\text{C}$ . Finally, the sample powder was stored *in vacuo*.

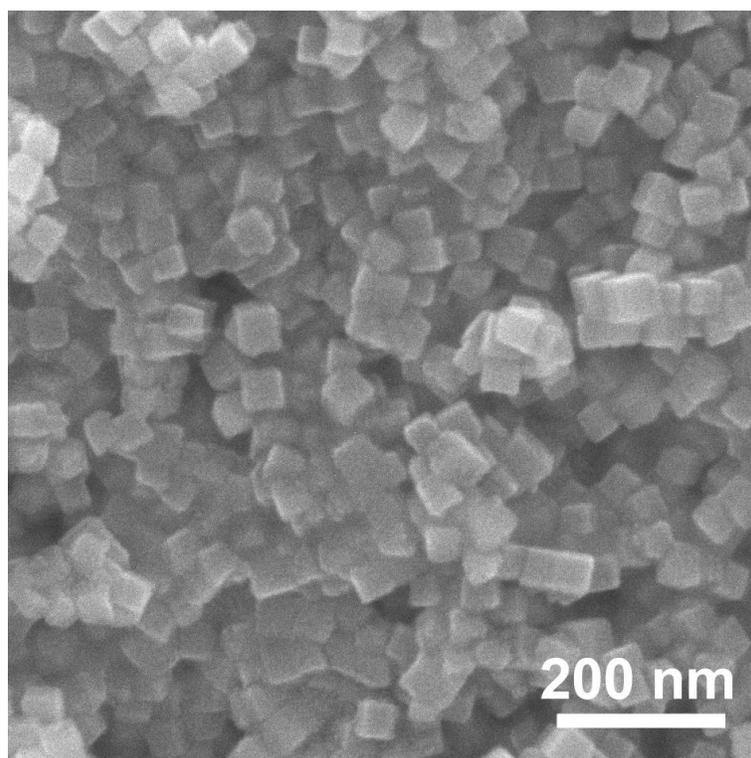
### **Spectroscopic characterizations**

The sample powders and electrodes were characterized using the following instruments; a scanning electron microscope (SEM, JEOL JSM-6500F at 15 kV) equipped with an energy dispersive spectrometer (EDS, Oxford Link Pentafet), a transmission electron microscope (TEM, JEOL JEM-2010F at 200 kV and JEOL JEM-4000EX), and an X-ray diffractometer, (XRD, Bruker AXS D8 Advance).

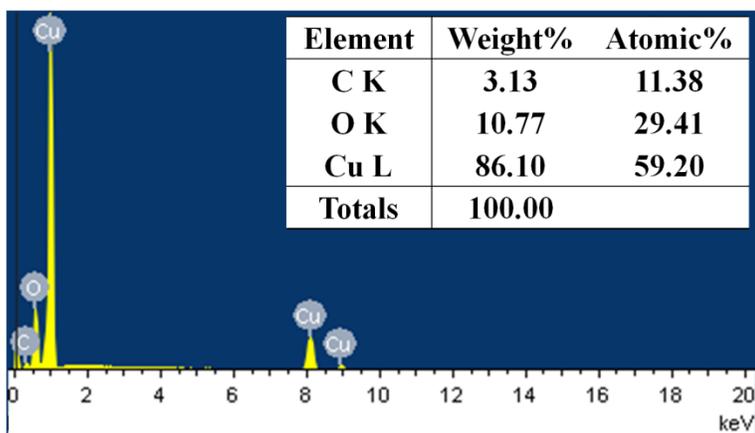
### **Electrochemical evaluation**

To evaluate the efficiency of the electrochemical anode in lithium ion batteries, electrochemical cells with negative electrodes which were made using sample A, sample B, and sample C. Lithium metal was used as the counter and the reference electrodes, and the electrolyte solution was 1.0 M  $\text{LiPF}_6$  in a 1: 1: 1 mixture (by volume) of ethylene carbonate (EC), diethyl carbonate (DEC), and dimethyl carbonate (DMC). The cell separator was a Celgard 2400 membrane. The anode comprised  $\text{Cu}_2\text{O}$  /carbon composite (i.e. sample A, sample B, and sample C) was made by the mixture of 80 wt % active material, 10 wt % carbon black conducting agent (super P) and 10 wt % poly(vinylidene fluoride) (PVDF) as binder. After these substances had been mixed in an N-methyl-2-pyrrolidone (NMP) solution, the prepared slurry was coated on copper foil to a thickness of approximately 50  $\mu\text{m}$ . After coating, the electrodes were dried at  $110\text{ }^{\circ}\text{C}$  for 2 h in a vacuum oven, to ensure that the solvent was evaporated. They were subsequently roll-pressed. The 2032 coin-type cells were assembled in an argon-filled glove box and charge/discharge tests were conducted using an AcuTech System BAT-750B

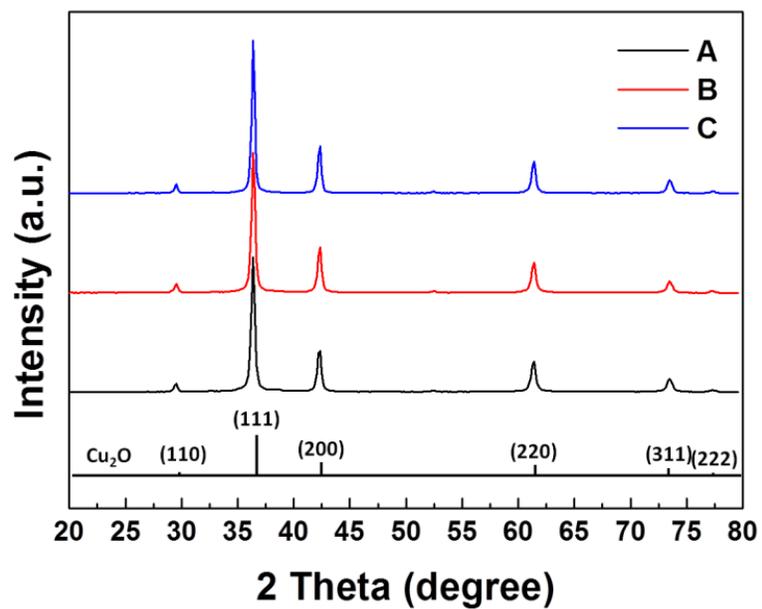
Battery Automatic Tester at a constant current density with a cutoff voltage of 0.02 V to 3.00 V.



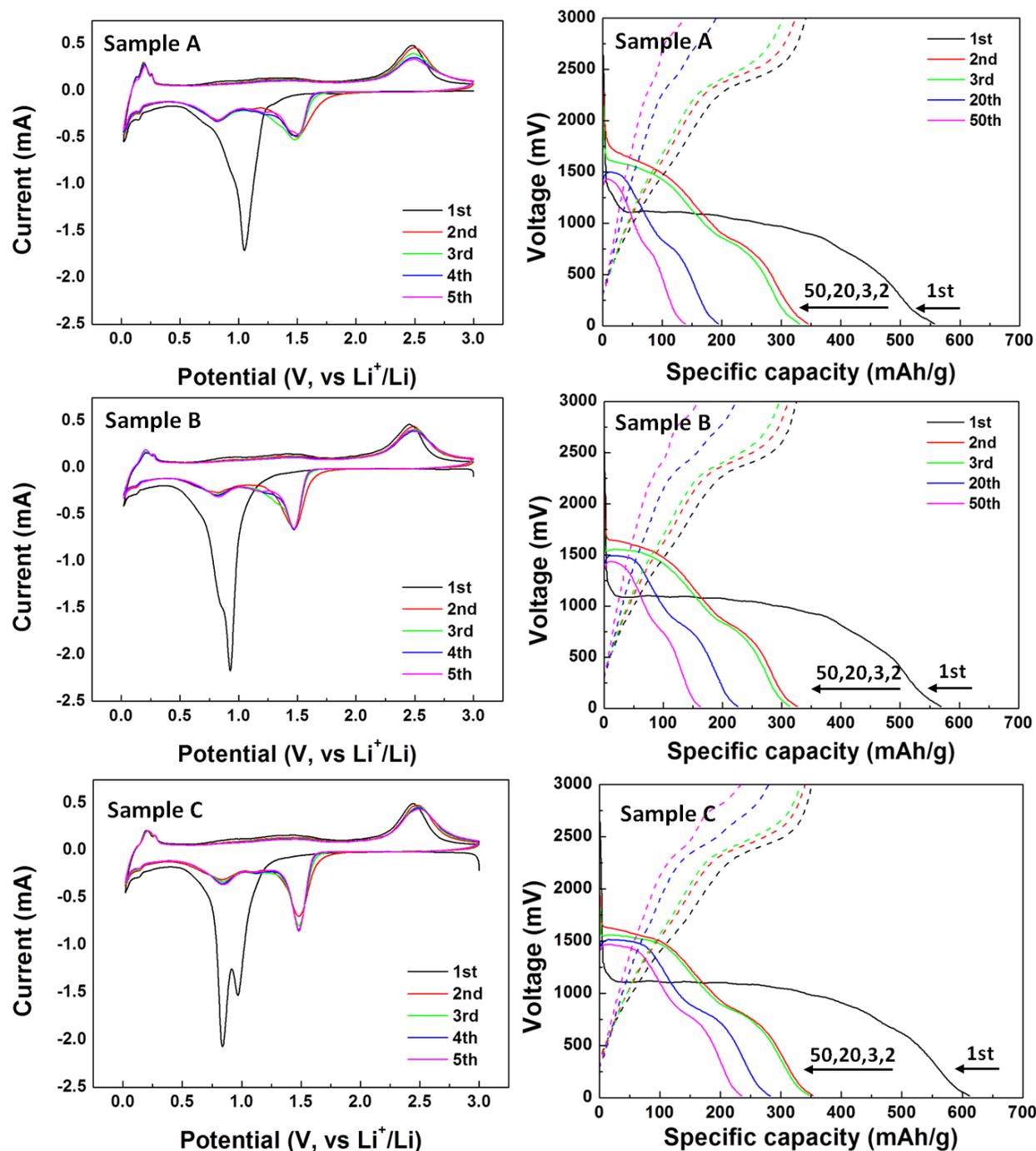
**Figure S1.** SEM images of the sample control 1.



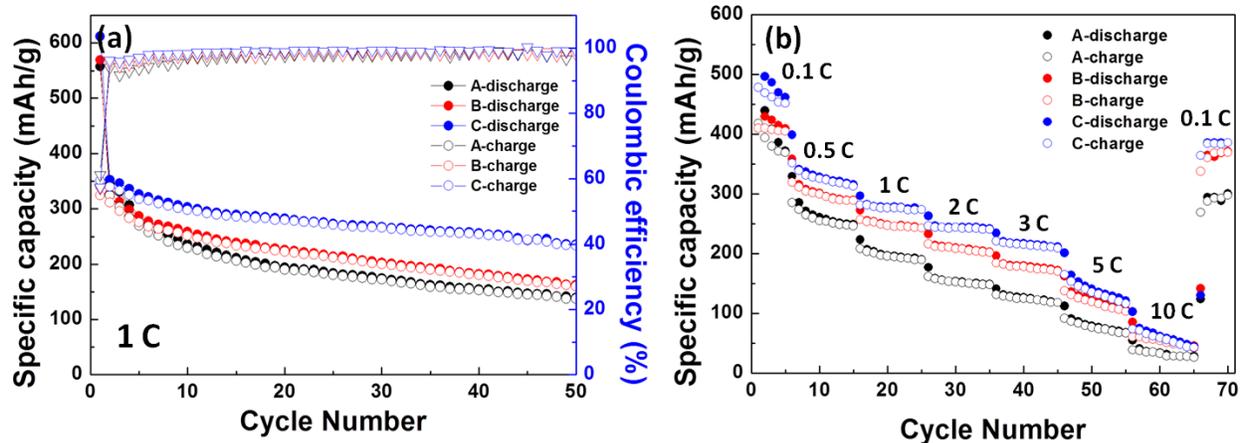
**Figure S2.** Energy dispersive spectrum of nanocubes sample C.



**Figure S3.** X-ray diffraction patterns (XRD) of various sized Cu<sub>2</sub>O nanocubes.



**Figure S4.** Cyclic voltammograms of Sample A, B and C obtained at a scan rate of  $0.2 \text{ mV s}^{-1}$  between 0.02 and 3.00 V (vs. Li/Li<sup>+</sup>); Galvanostatic charge-discharge curves of Sample A, B and C at potential 0.02 – 3.00 V versus Li<sup>+</sup>/Li at a rate of 1 C. Solid and dashed lines represent discharging and charging process and cycle number is indicated by color.



**Figure S5.** (a) Cycling performance of Cu<sub>2</sub>O nanocube electrodes at a rate of 1 C. (b) Specific capacity at various discharging and charging rates.