

## Supplementary Information

### Rational Design of Flexible Inorganic Composites Membrane with Interconnected Porous Structure as High-Performance Lithium Ion Capacitor Electrodes

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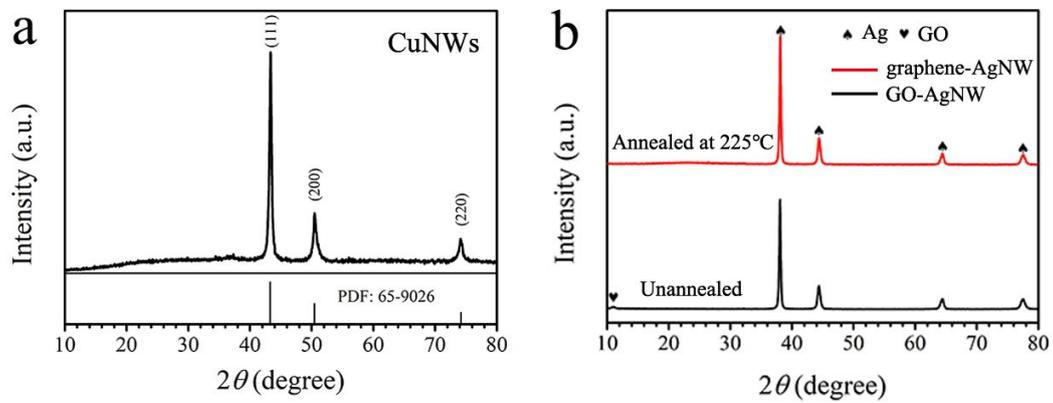
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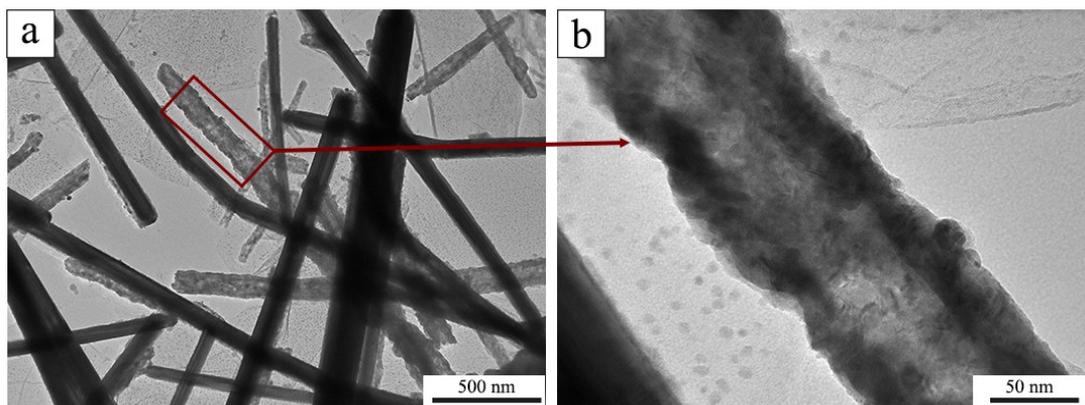
#### Experimental section

The electrochemical measurements of the single membrane electrodes were carried out in a three-electrode system consisted of the as-prepared membrane as working electrode, a Hg/HgO electrode as reference electrode, and a platinum foil as counter electrode in 1.0 mol·L<sup>-1</sup> KOH aqueous solution. The area of all working electrodes was cut into a square of 1\*1cm<sup>2</sup>. The symmetrical solid-state supercapacitor (SSS) was assembled with two identical membranes (1\*1 cm<sup>2</sup>) as negative and positive electrodes, respectively, and PVA/KOH gel as the electrolyte. In addition, the electrode size of the flexible supercapacitor was designed to be 2\*4 cm<sup>2</sup>. In a typical solid electrolyte preparation process, 2 g of PVA was dissolved into 20 mL deionized water under magnetic stirring at 90 °C for 30 min. Then, KOH aqueous solution (10 mL of 1.0 mol·L<sup>-1</sup>) was slowly added to the PVA solution and stirred for 30 min at room temperature to form a uniform gel electrolyte. The supercapacitor preparation was as follows. Firstly, two prepared membrane electrodes were soaked in the gel electrolyte for 10 min, and then cured at room temperature for 30 min. After that, the two electrodes were squeezed face to face to promote close contact between the electrode and the electrolyte. Finally, the assembled SSS was sealed with

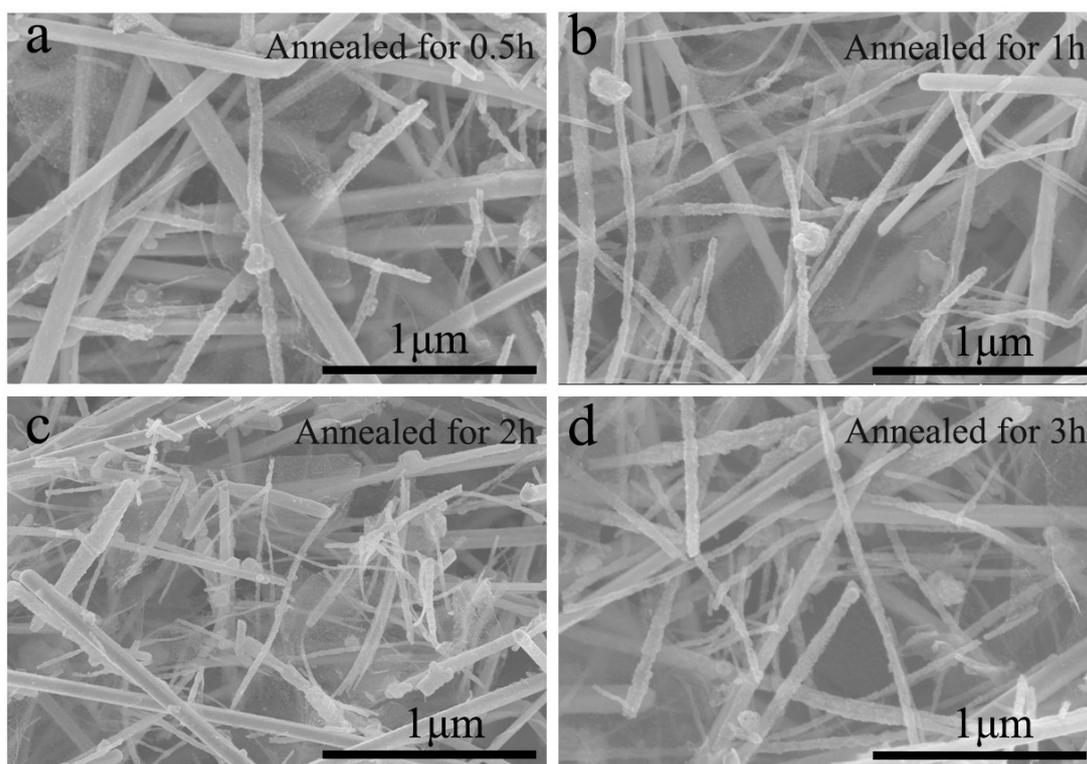
Polyimide (PI) tape.



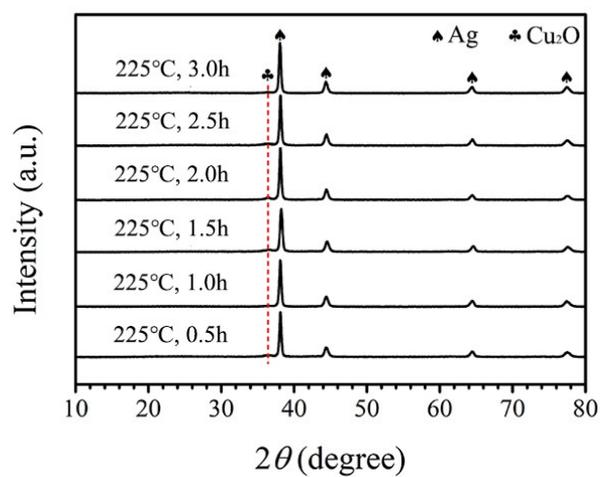
**Figure S1.** XRD patterns of (a) the CuNW membrane and (b) GO/AgNW and graphene/AgNW.



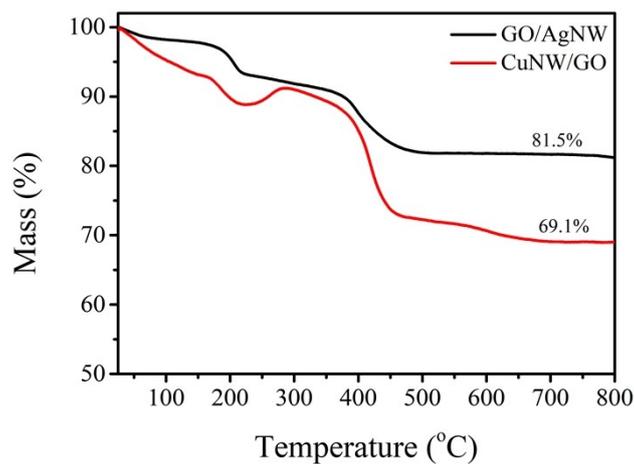
**Figure S2.** TEM images of (a) CGA-225 membrane and (b) Cu<sub>x</sub>ONWs.



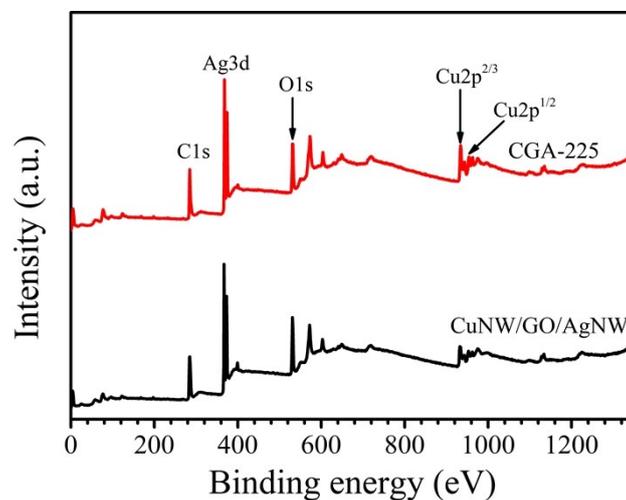
**Figure S3.** SEM images of CGA-225 membrane with different annealing time (a) 0.5 h, (b) 1 h, (c) 2 h, and (d) 3 h.



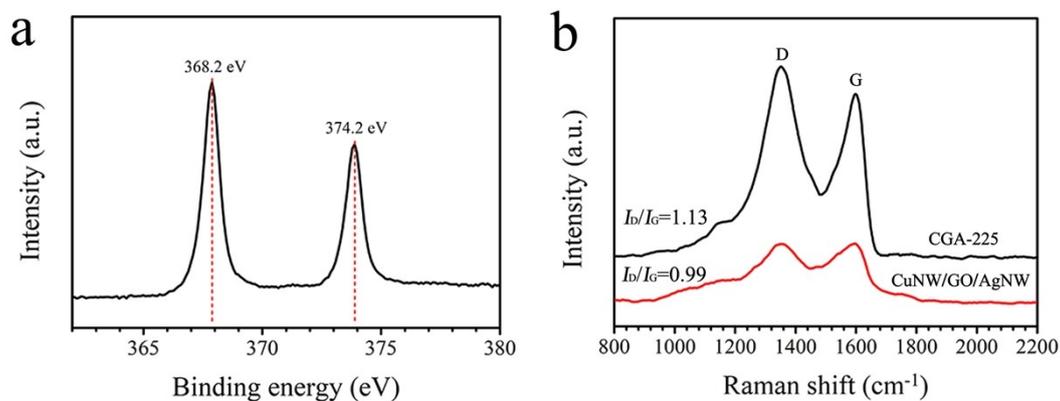
**Figure S4.** XRD patterns of the annealing of CGA membrane at 225 °C for different times.



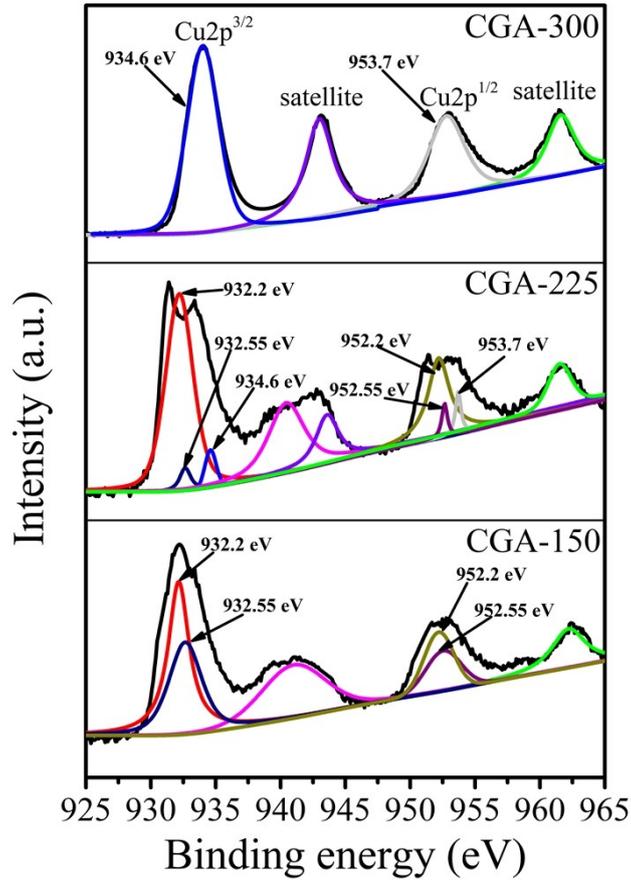
**Figure S5.** Thermogravimetric (TG) curves of GO/AgNW and CuNW/GO.



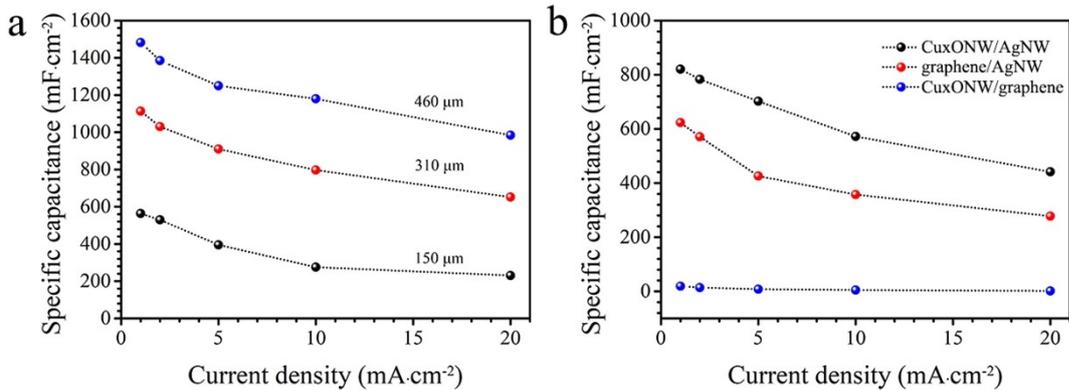
**Figure S6.** XPS spectra of CGA-150 and CGA-225.



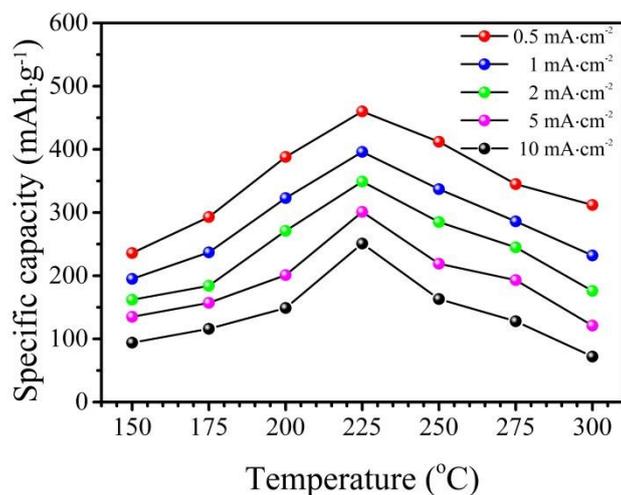
**Figure S7.** (a) XPS spectrum of Ag3d of CGA-225 composite film. (b) Raman spectra of CGA-225 and CuNW/GO/AgNW.



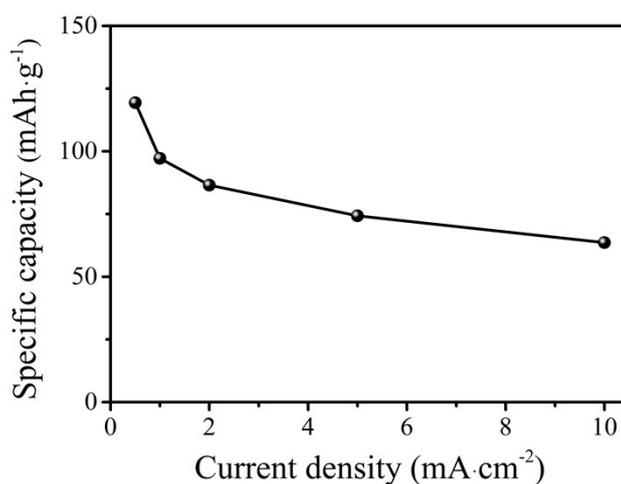
**Figure S8.** XPS spectra of Cu2p for CGA-150, CGA-225, and CGA-300.



**Figure S9.** (a) The areal capacitance of CGA-225 composite membranes with different thickness at different current densities. (b) The areal capacitance of Cu<sub>x</sub>ONW/AgNW, graphene/AgNW, and Cu<sub>x</sub>ONW/graphene composite membrane at different current densities.



**Figure S10.** Specific capacity of CGA at different annealing temperatures for 2h under different current densities.



**Figure S11.** Rate performance curve of AC cathodes at different current densities.

**Table S1.** Atomic percentage ratio (at. %) of Cu<sup>0</sup>, Cu<sup>+</sup>, and Cu<sup>2+</sup> in Cu2p<sup>3/2</sup> XPS spectra of CGA-150, CGA-225, and CGA-300.

Sample	Cu <sup>0</sup> (at. %)	Cu <sup>+</sup> (at. %)	Cu <sup>2+</sup> (at. %)
CGA-150	43.5	56.5	0
CGA-225	3.9	90.4	5.7
CGA-300	0	0	100

**Table S2.** Comparison of the electrochemical performance of graphene-based and copper-based oxide electrodes.

Sample	Current density	Specific capacitance	References
Cu <sub>2</sub> O@Cu nanosheets	2 mA·cm <sup>-2</sup>	390.9 mF·cm <sup>-2</sup>	S1
CuO/Cu foam	1 mA·cm <sup>-2</sup>	641 mF·cm <sup>-2</sup>	S2
NiOOH@CuO/Cu <sub>2</sub> O nanosheet arrays	2 mA·cm <sup>-2</sup>	776 mF·cm <sup>-2</sup>	S3
Cu <sub>2</sub> O/Cu/cellulose	1 mA·cm <sup>-2</sup>	238 mF·cm <sup>-2</sup>	S4
CuO/Cu <sub>2</sub> O@CoO core-shell nanowire	1 mA·cm <sup>-2</sup>	280 mF·cm <sup>-2</sup>	S5
MnO <sub>2</sub> /graphene/Au/Ag	1 mA·cm <sup>-2</sup>	49.1 mF·cm <sup>-2</sup>	S6
graphene/ MnO <sub>2</sub> NWs	1 mA·cm <sup>-2</sup>	767 mF·cm <sup>-2</sup>	S7
Polyaniline/graphene hybrid fibers	0.22 mA·cm <sup>-2</sup>	55.8 mF·cm <sup>-2</sup>	S8
CGA-225	1 mA·cm <sup>-2</sup>	1,113.4 mF·cm <sup>-2</sup>	This work
CGA-225	2 mA·cm <sup>-2</sup>	911.2 mF·cm <sup>-2</sup>	This work

## References

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