Supporting Information For

Functionalization of biomass carbonaceous aerogels and its application as electrode materials for electro-enhanced recovery of metal ions

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CDI decontamination test

The electrosorption capacity of the CDI system was conducted in a continuously recycling system including a home-made single CDI unit, an electrical power supply, a conductivity meter (Type 308A, Leici company), a peristaltic pump (BT100-2J, Baoding LanGe constant Flow Pump Co., Ltd, China) and a water tank. In each experiment, the CuCl₂ solution was continuously pumped from the pump into the cell and the effluent was returned to the water tank. In detail, the flow rate was set constant to 25 mL/min and the total solution volume was 200 mL in the system. The distance of 2 mm between the electrodes and a direct voltage were applied. The CDI electrodes were prepared as following: 80 mg electrode materials with 10 mg carbon black and 10 mg PTFE were dispersed into 5 mL ethanol solution under ultrasonic treatment for 5 min to form dispersion. Then, this dispersion was dropped onto the center of a nickel foam plate. After that, the resultant electrodes were dried at 140 °C overnight. The hole with a diameter of 5 mm was punched in the prepared electrode to allow the water flowing through the CDI device.



Fig. S1 TEM mapping of the CAs/TiO₂.



Fig. S2 FTIR spectra of CAs and CAs/MO hybrids.



Fig. S3 Nitrogen adsorption–desorption isotherms (A); and their corresponding cumulative distribution curve (B) of the CAs/CeO₂ and CAs/Fe₂O₃ hybrids.



Fig. S4 The SAED patterns of the as-obtained CAs/MO hybrids.



Fig. S5 TEM images of the CAs/TiO₂ hybrids with different added amount of Ti^{3+} precursor: 5 mg (A) and 15 mg (B) Ti^{3+} precursor in 15 mL aqueous solution containing 30 mg of CAs.



Fig. S6 CAs/TiO₂ electrodes in different CuCl₂ concentrations at a scan rate of 20 mV/s.



Fig. S7 Water contact angle measurements of the prepared electrodes.



Fig. S8 The actual CDI device.



Fig. S9 Cu(II) removal capacity and pH value of decontamination experiments for 50 mg/L CuCl₂ solution at various applied potentials.



Fig. S10 Cu 2p high resolution XPS spectra of the CAs/TiO₂ electrodes after the

experiments at open circuit and applied voltage of 1.2 V.



Fig. S11 The removal efficiency of these electrodes with different initial concentration of Cu(II).



Fig. S12 Current transient curve for CAs/TiO₂ in a 200 mg/L Cu(II) solution at 1.2 V. The charge efficiency Λ is obtained from the formula:¹

$$\Lambda = \frac{\Gamma \times F}{\Sigma}$$

in which Γ is the deionization capacity (mol/g), F is the Faraday constant (96485 C/mol) and Σ (charge, C/g) is calculated through integrating the current. Base on the formula, the charge efficiency of CAs/TiO₂ was calculated to be 0.44.



Fig. S13 SEM image of the CAs/TiO₂ hybrids after cycling three times.



Fig. S14 Underlying mechanism of the CDI process for Cu(II) removal in the presence of NaCl.

| | C _{s max} (mg/g) | b (L/mg) | R ² |
|--|------------------------------|-------------|-----------------------|
| CAs/TiO ₂ , 0 V | 19.280 | 0.430 | 0.993 |
| CAs, 1.2 V | 30.353 | 0.309 | 966 |
| CAs/Fe ₂ O ₃ , 1.2 V | 41.424 | 0.268 | 0.996 |
| CAs/CeO ₂ , 1.2 V | 49.281 | 0.238 | 0.990 |
| CAs/TiO ₂ , 1.2 V | 57.134 | 0.337 | 0.989 |

Table S1 Parameters for Langmuir isotherm models

Reference

1. X. Xu, Z. Sun, D. H. Chua and L. Pan, Sci. Rep., 2015, 5, 11225.