

Electronic supplementary information for:

**Cascaded Utilization of Magnetite Nanoparticles@Onion-Like Carbons from Wastewater Purification to Supercapacitive Energy Storage**

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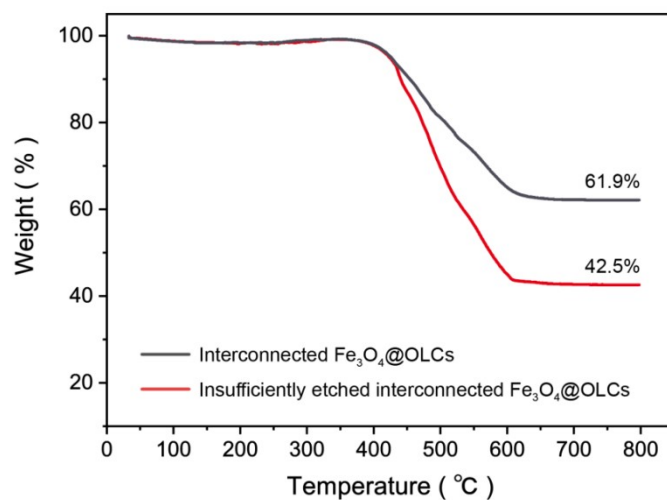
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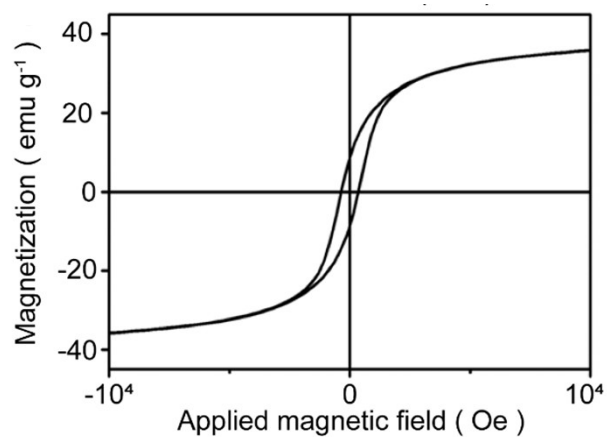
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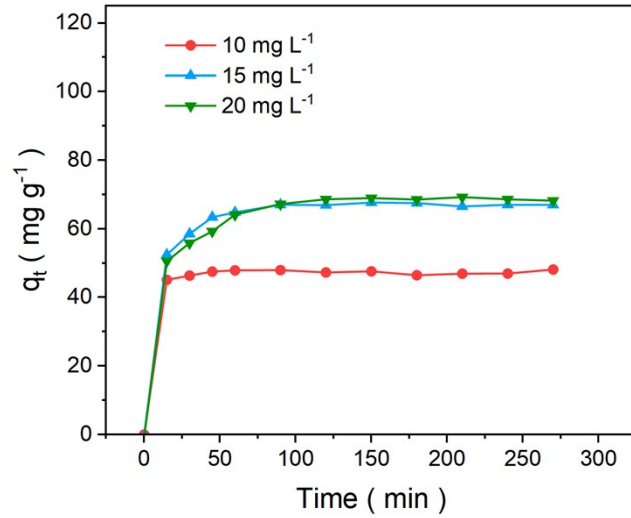
## Supplementary figures



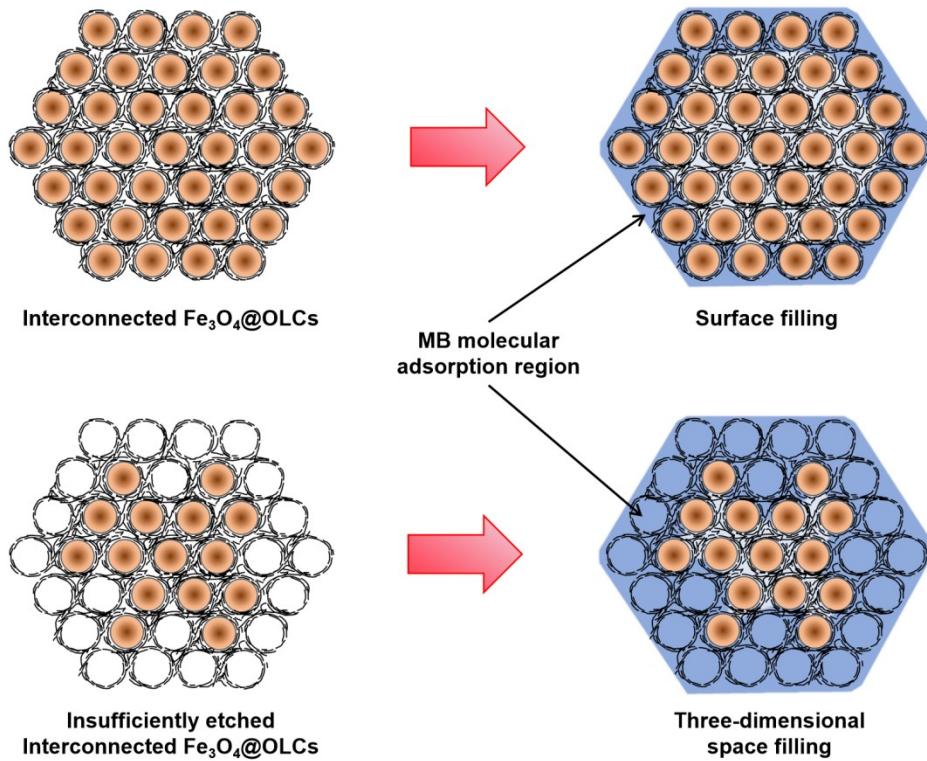
**Fig. S1.** TG curves of interconnected Fe<sub>3</sub>O<sub>4</sub>@OLCs and insufficiently etched interconnected Fe<sub>3</sub>O<sub>4</sub>@OLCs at a heating rate of 10 °C min<sup>-1</sup> from 30 to 800 °C in air.



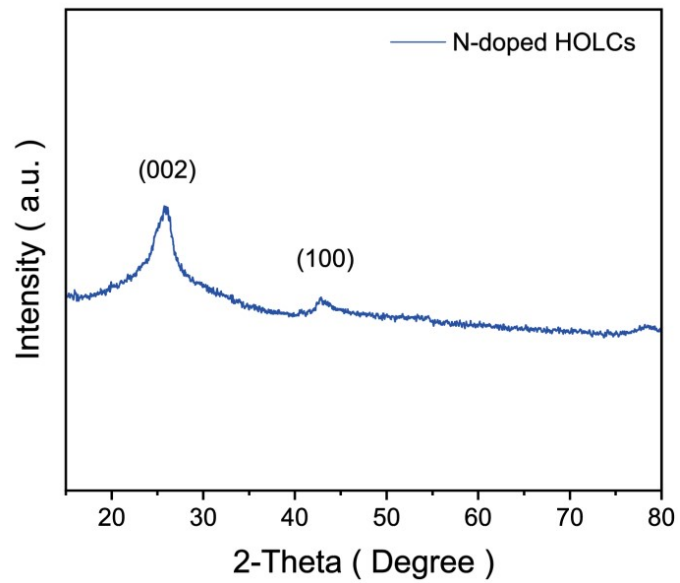
**Fig. S2.** Magnetic hysteresis loop of insufficiently etched interconnected Fe<sub>3</sub>O<sub>4</sub>@OLCs.



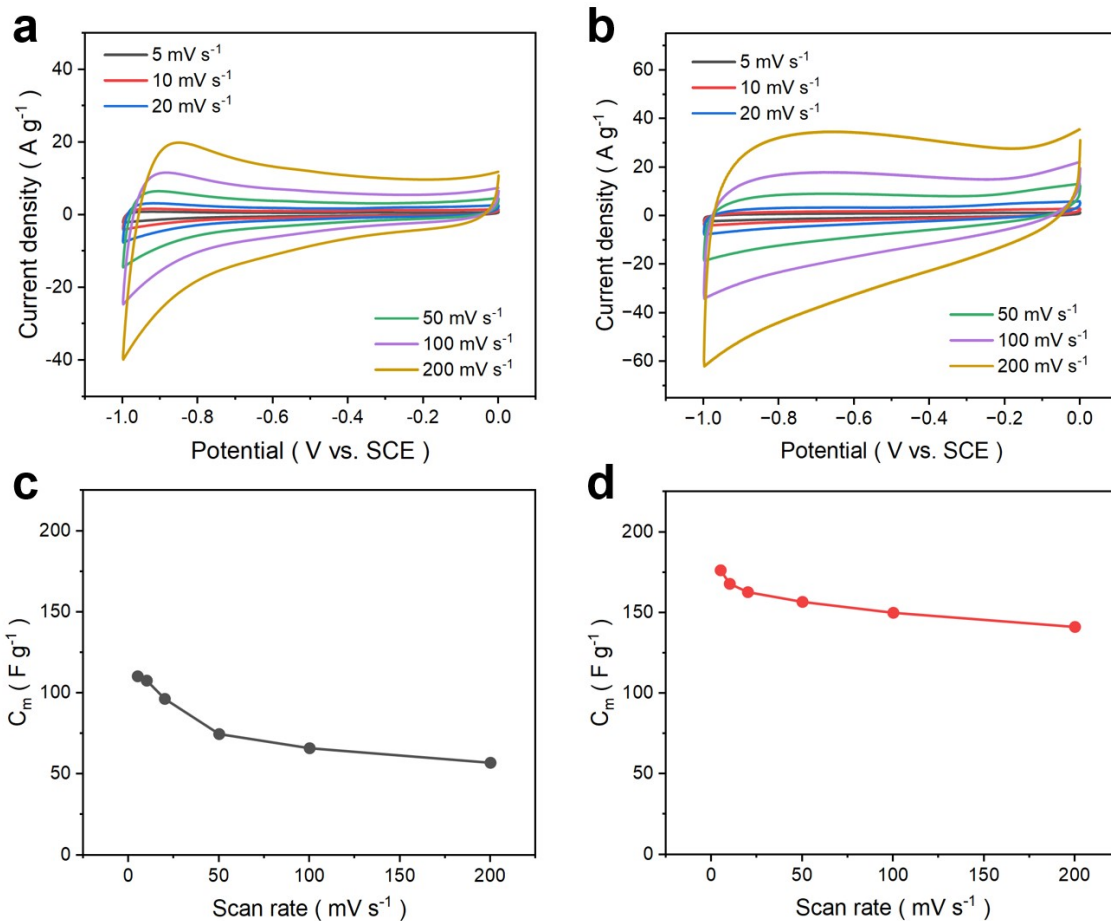
**Fig. S3.** Variation of the adsorption capacities of interconnected  $\text{Fe}_3\text{O}_4@\text{OLCs}$  with treated period.



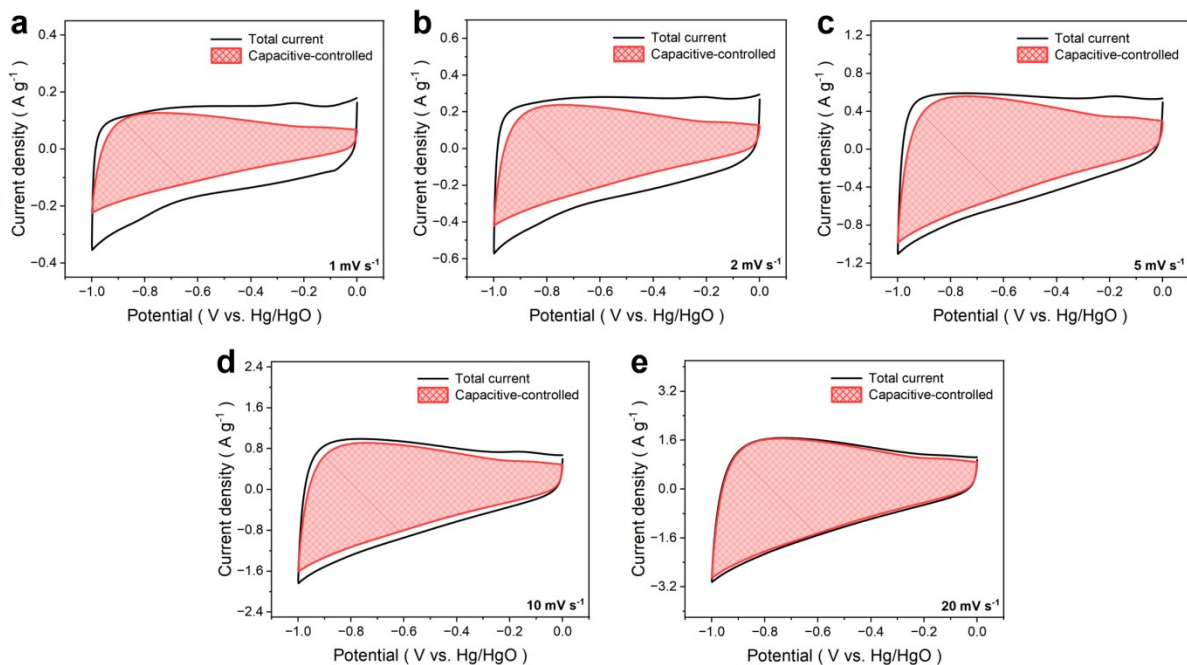
**Fig. S4.** Schematic illustration of the difference in MB adsorption mechanisms between interconnected  $\text{Fe}_3\text{O}_4@\text{OLCs}$  and insufficiently etched interconnected  $\text{Fe}_3\text{O}_4@\text{OLCs}$ .



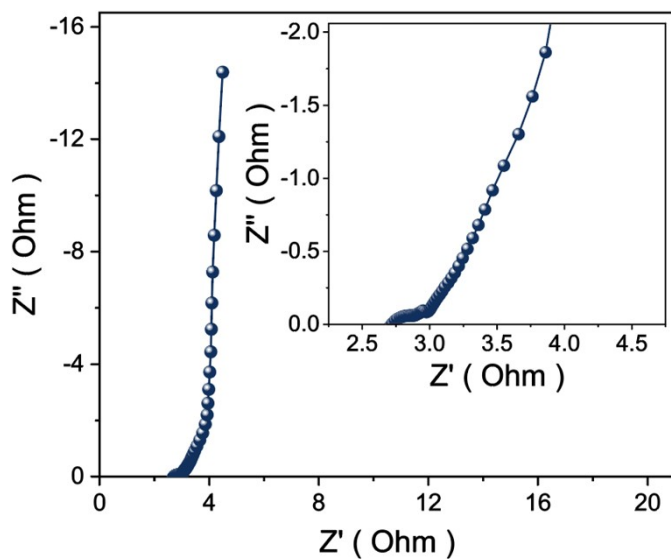
**Fig. S5.** XRD profile of the N-doped HOLCs.



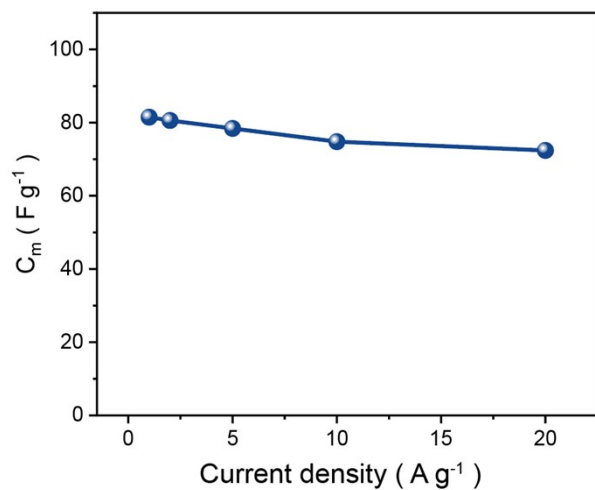
**Fig. S6.** CV curves of (a) insufficiently etched interconnected Fe<sub>3</sub>O<sub>4</sub>@OLCs and (b) N-doped porous carbon structure at different scan rates from 5 to 200 mV s<sup>-1</sup> in three-electrode testing system. Specific gravimetric capacitances (C<sub>m</sub>) calculation results of (c) Fe<sub>3</sub>O<sub>4</sub>@OLCs and (d) N-doped HOLCs at different scan rates from 5 to 200 mV s<sup>-1</sup>.



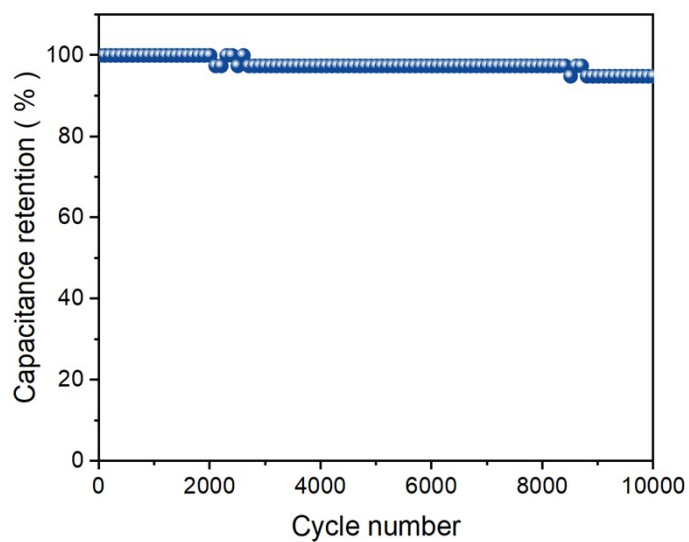
**Fig. S7.** Electrochemical kinetic analysis of N-doped HOLCs. Percentage of capacitive-controlled charge contribution from EDLC based on CV measurements at scan rates of (a) 1, (b) 2, (c) 5, (d) 10 and (e) 20  $\text{mV s}^{-1}$ .



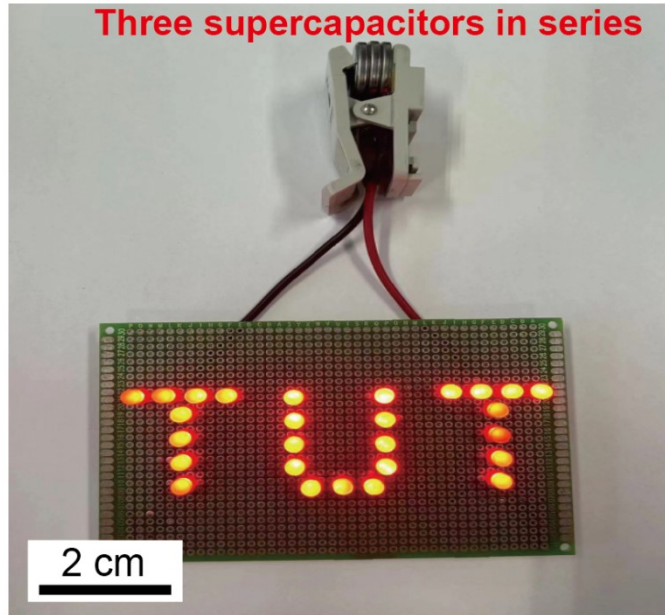
**Fig. S8.** Nyquist plot of supercapacitor.



**Fig. S9.** Specific gravimetric capacitances ( $C_m$ ) versus current densities of the supercapacitor device.



**Fig. S10.** Cycling stability of supercapacitor measured at a charging-discharging current density of  $10 A g^{-1}$ .



**Fig. S11.** 27 red LEDs powered by three supercapacitors connected in series.



**Table S1.** Comparisons of energy storage performances of supercapacitor based on N-doped HOLCs with other carbon-based supercapacitors.

| Materials                           | Electrolyte                         | Energy density (Wh kg <sup>-1</sup> ) | Power density (kW kg <sup>-1</sup> ) | Cycle stability         | Ref.      |
|-------------------------------------|-------------------------------------|---------------------------------------|--------------------------------------|-------------------------|-----------|
| Sulfur-doped carbon nano-onions     | 1M Na <sub>2</sub> SO <sub>4</sub>  | 10.6                                  | 0.1                                  | 95% after 10000 cycle   | S1        |
| Quinone-decorated onion-like carbon | 1M H <sub>2</sub> SO <sub>4</sub>   | 6.4                                   | 19.2                                 | 90% after 10000 cycle   | S2        |
| Nori:ZnCl <sub>2</sub> -2:1         | 6M KOH                              | 6.1                                   | 0.05                                 | 96% after 5000 cycle    | S3        |
| ZIF-8 derived carbon                | 6M KOH                              | 4.66                                  | 0.21                                 | 96% after 10000 cycle   | S4        |
| HPCF                                | 6M KOH                              | 9.1                                   | 3.5                                  | 95% after 10000 cycle   | S5        |
| PANI-carbon nanotube composites     | H <sub>2</sub> SO <sub>4</sub> /PVA | 5.8                                   | 1.1                                  | 83.2% after 2000 cycle  | S6        |
| TPI-P-700                           | 1M H <sub>2</sub> SO <sub>4</sub>   | 10.5                                  | 0.5                                  | 100% after 10000 cycle  | S7        |
| Phosphorus-doped porous carbon      | 1M H <sub>2</sub> SO <sub>4</sub>   | 10.6                                  | 0.22                                 | 98.3% after 5000 cycle  | S8        |
| HPCSLs-700                          | 7M KOH                              | 8.6                                   | 0.014                                | 92% after 10000 cycle   | S9        |
| N-C-HPCS                            | 6M KOH                              | 6.1                                   | 0.25                                 | 108% after 10000 cycle  | S10       |
| N-doped HOLCs                       | 3M KOH                              | 11.3<br>7.9                           | 0.51<br>8.9                          | 94.6% after 10000 cycle | This Work |

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