

## Electronic Supplementary Information

### High-efficiency transformation of amorphous carbon into graphite nanoflakes for stable aluminum-ion battery cathodes

Jiguo Tu,<sup>a</sup> Junxiang Wang,<sup>a</sup> Shijie Li,<sup>a</sup> Wei-Li Song,<sup>\*,b</sup> Mingyong Wang,<sup>\*,a</sup> Shuqiang Jiao<sup>\*,a</sup> and Hongmin Zhu<sup>a,c</sup>

<sup>a</sup> State Key Laboratory of Advanced Metallurgy, University of Science and Technology Beijing, Beijing, 100083, PR China

<sup>b</sup> Institute of Advanced Structure Technology, Beijing Institute of Technology, Beijing 100081, PR China

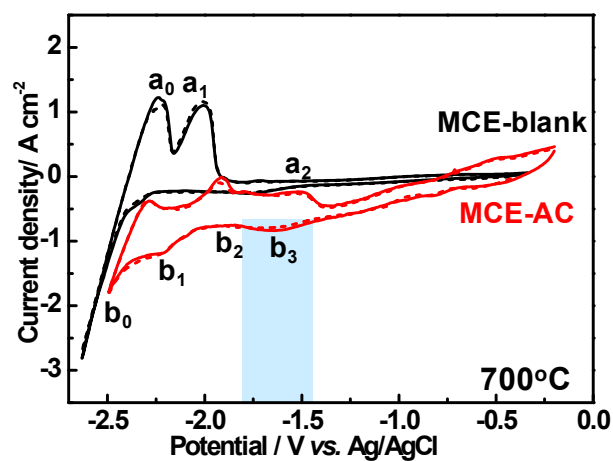
<sup>c</sup> Department of Metallurgy, Graduate School of Engineering, Tohoku University, Sendai, Miyagi 9808579, Japan

#### \*Corresponding Author

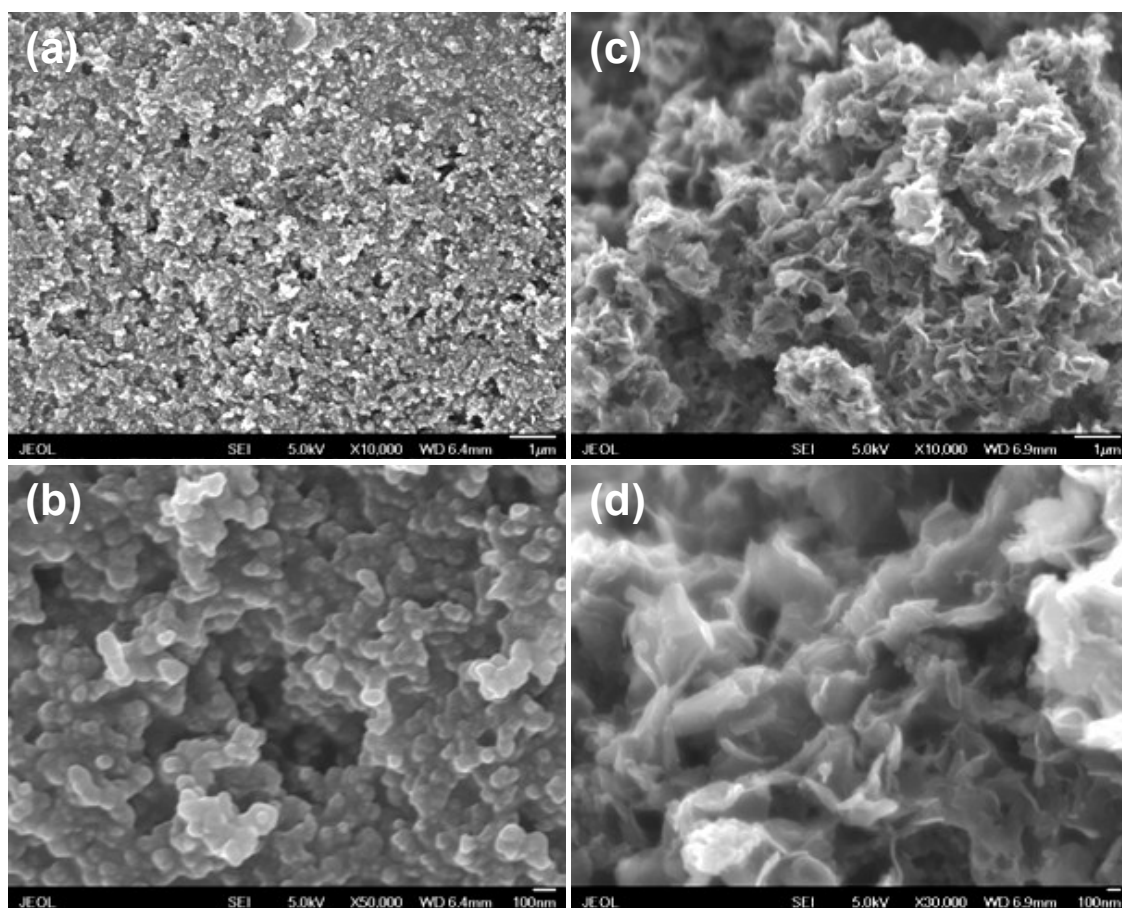
E-mail: weilis@bit.edu.cn, Tel: +86-10-68913302, Fax: +86-10-68913302

E-mail: mywang@ustb.edu.cn, Tel: +86-10-62333617, Fax: +86-10-62333617

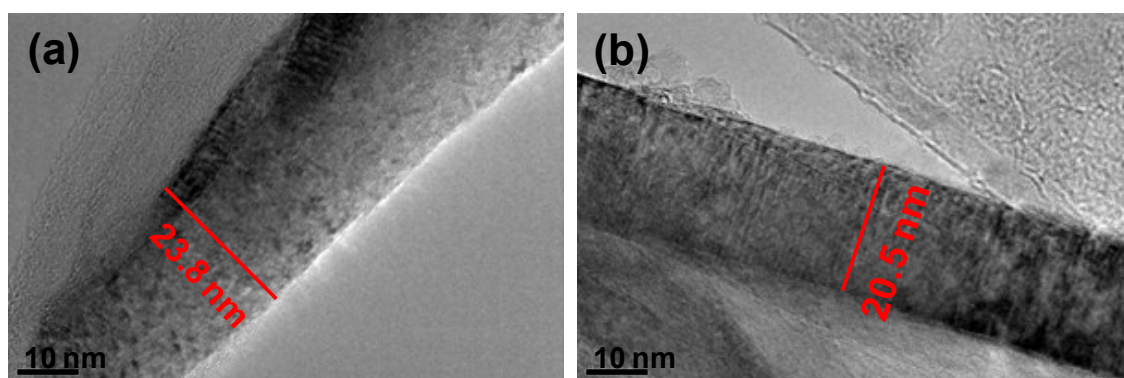
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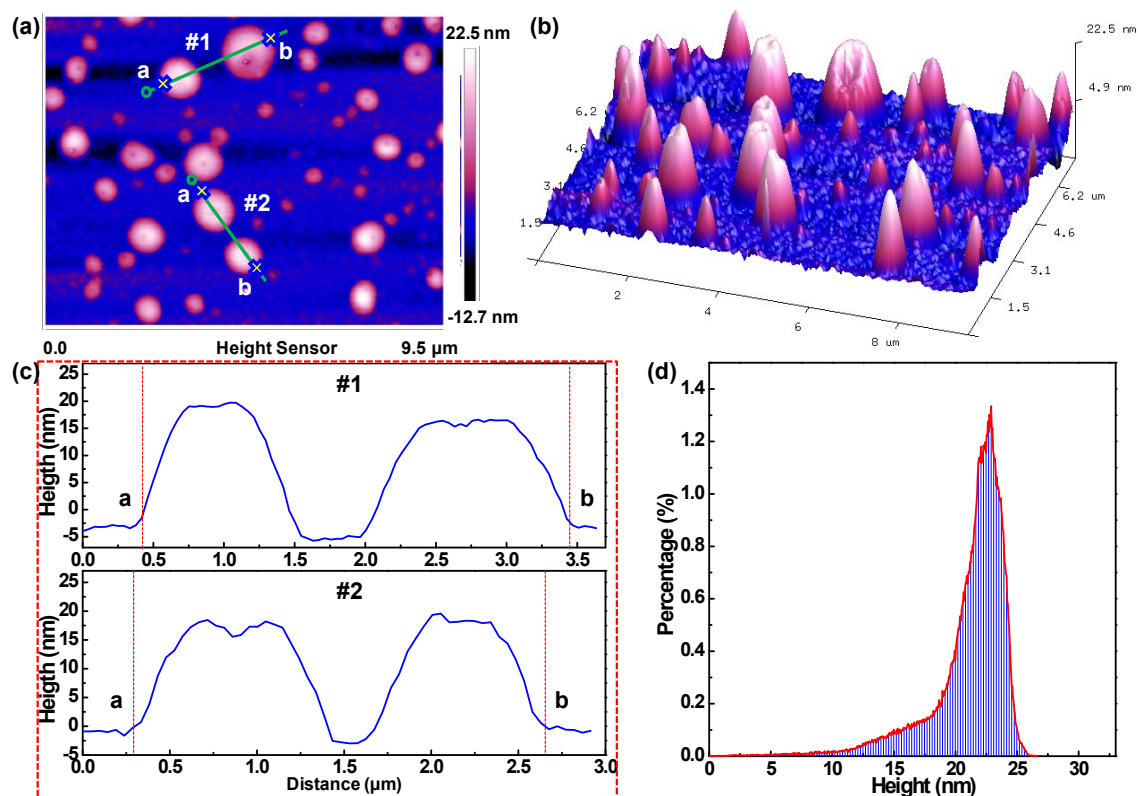
**Fig. S1.** CV curves of the blank MCE and MCE loaded with amorphous carbon in  $\text{CaCl}_2\text{-LiCl}$  melts at 700 °C at a scan rate of 50 mV s<sup>-1</sup>.



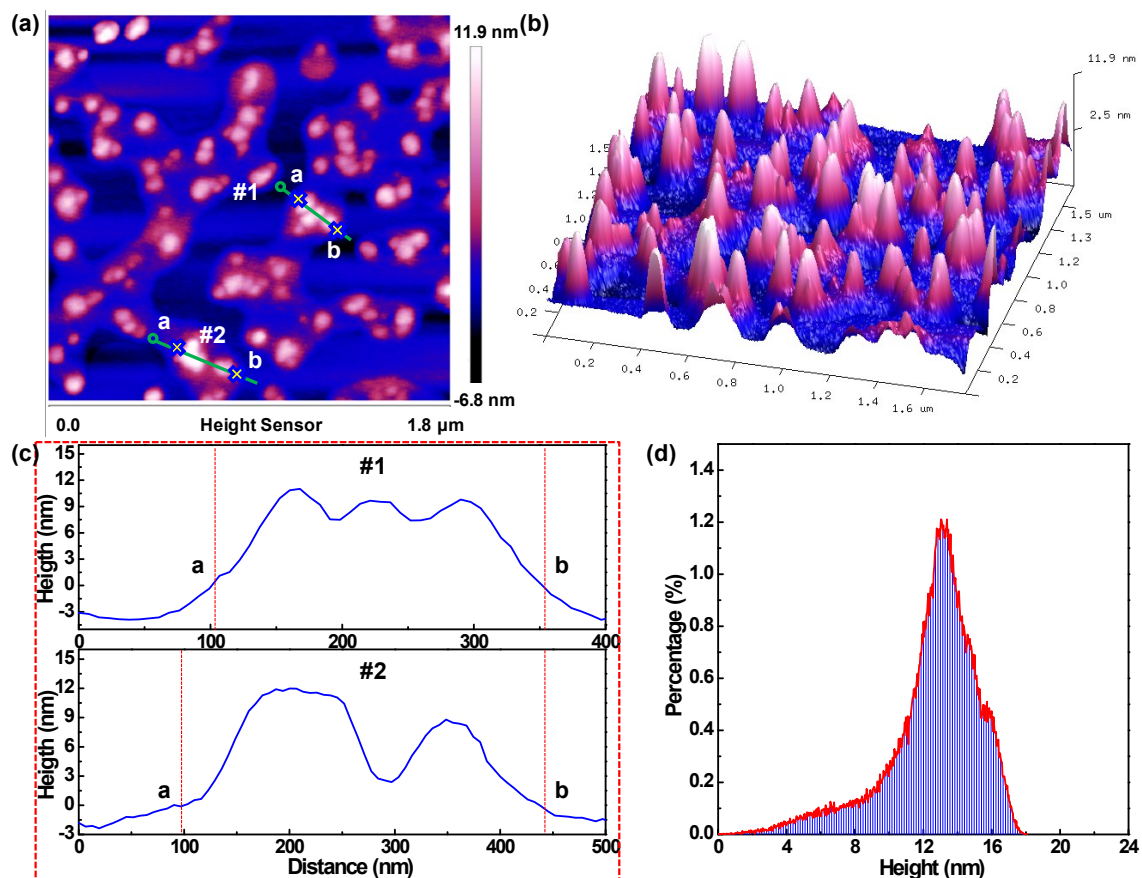
**Fig. S2.** SEM images of the as-prepared samples through constant cell voltage electrolysis under 3.0 V at 700 °C for 2 h (a,b) and 5 h (c,d).



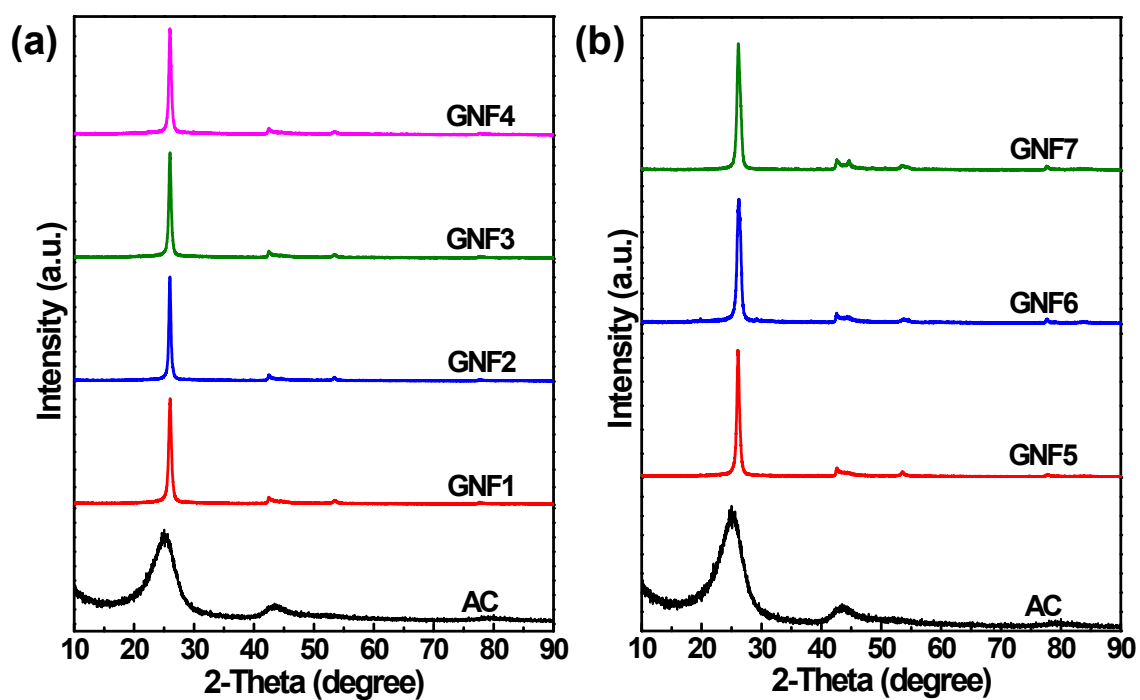
**Fig. S3.** HRTEM images of amorphous carbon after cathodic electrolysis under different constant cell voltages of 2.8 V (a) and 3.0 V (b) at 800 °C for 2 h.



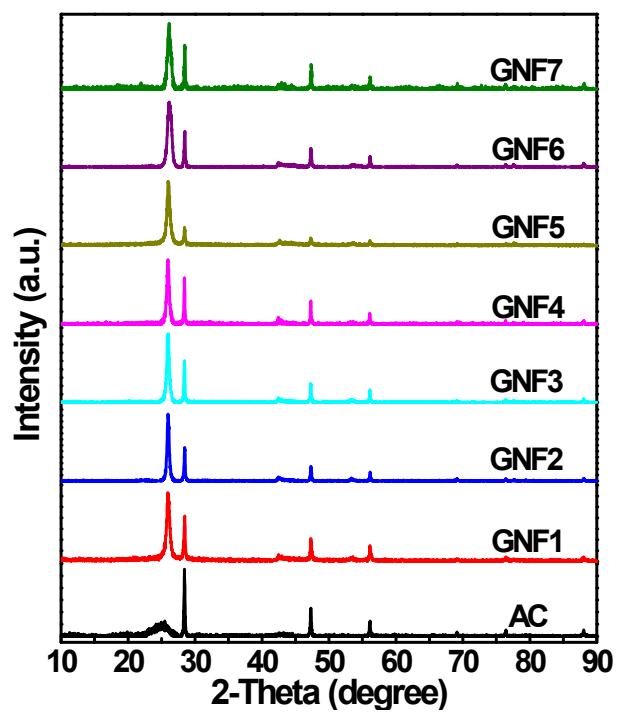
**Fig. S4.** Typical AFM and 3D AFM images of the electrolyzed sample under 3.0 V at 800 °C for 2 h (a,b). The thinness at different locations (c). Percentage of average thickness distribution of the electrolyzed sample (d).



**Fig. S5.** Typical AFM and 3D AFM images of the electrolyzed sample under 2.8 V at 850 °C for 2 h (a,b). The thinness at different locations (c). Percentage of average thickness distribution of the electrolyzed sample (d).

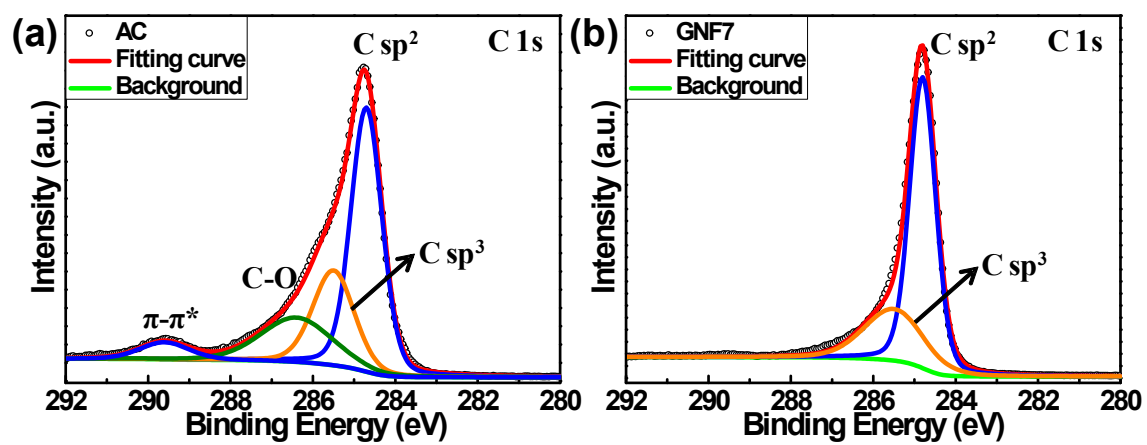


**Fig. S6.** XRD patterns of amorphous carbon before and after cathodic electrolysis under different constant cell voltages at 800 °C for 2h (a). XRD patterns of amorphous carbon before and after cathodic electrolysis under 2.8 V at 850 °C for different times (b).

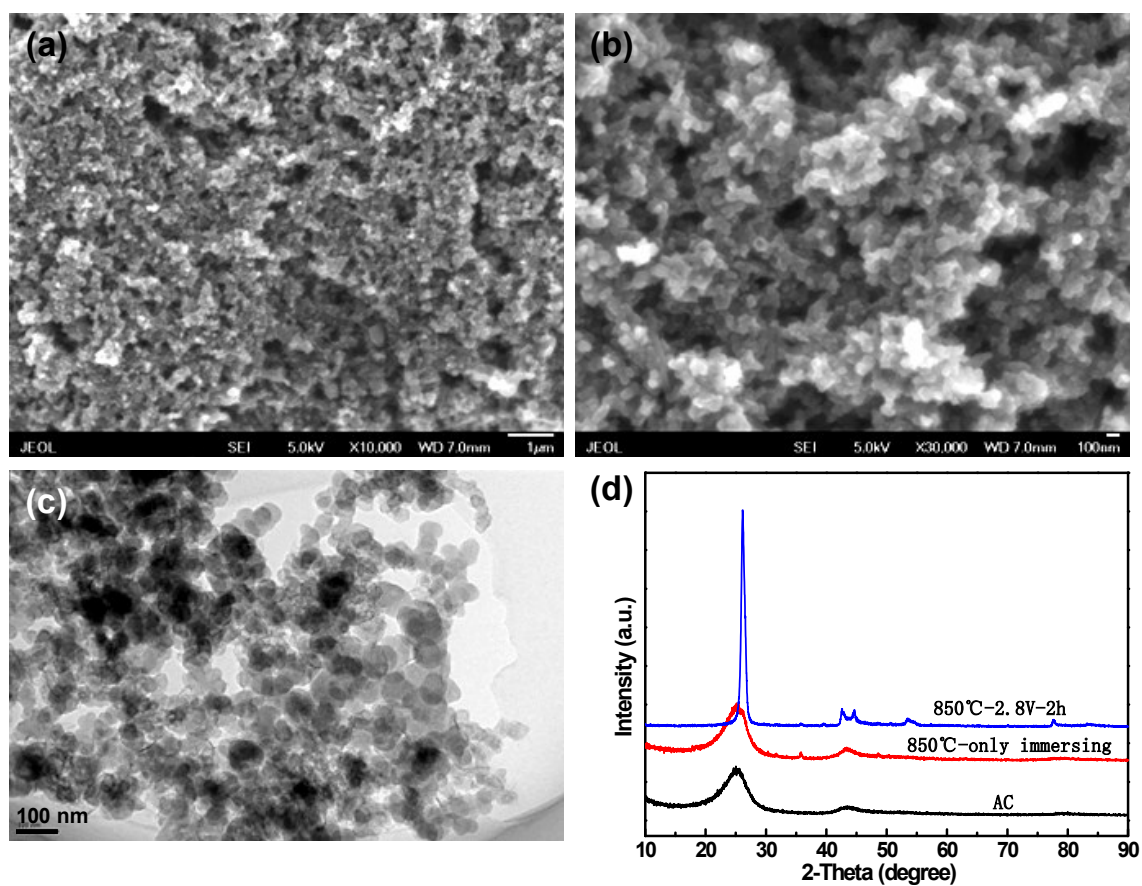


**Fig. S7.** XRD patterns of amorphous carbon before and after cathodic electrolysis at the indicated conditions with a small number of Si standard powders.

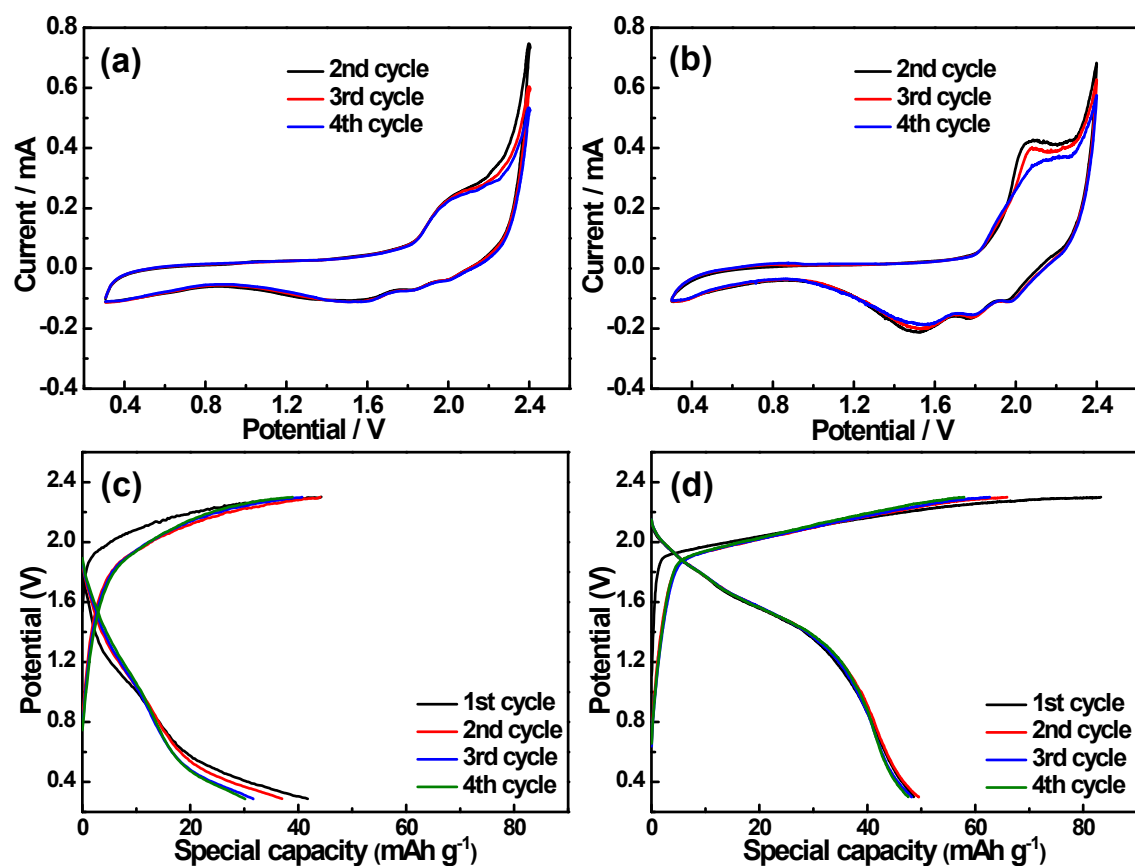




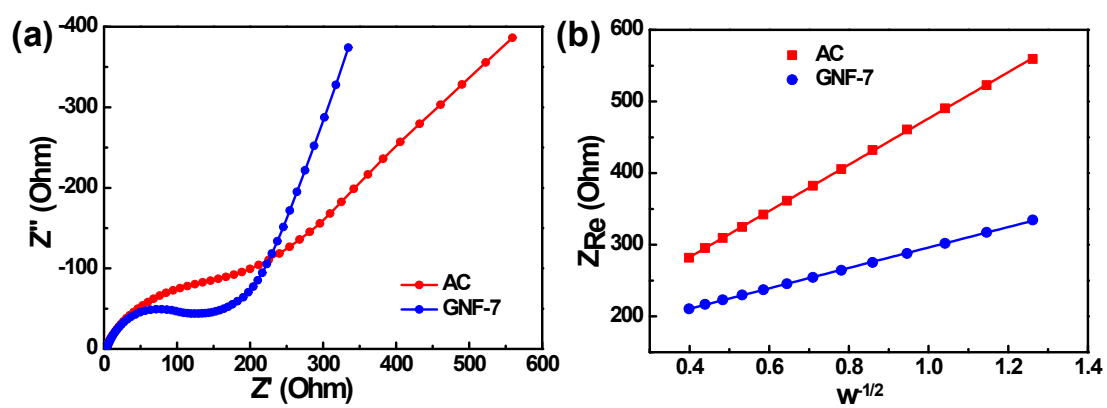
**Fig. S8.** High-resolution XPS spectra of C1s for AC (a) and GNF7 (b).



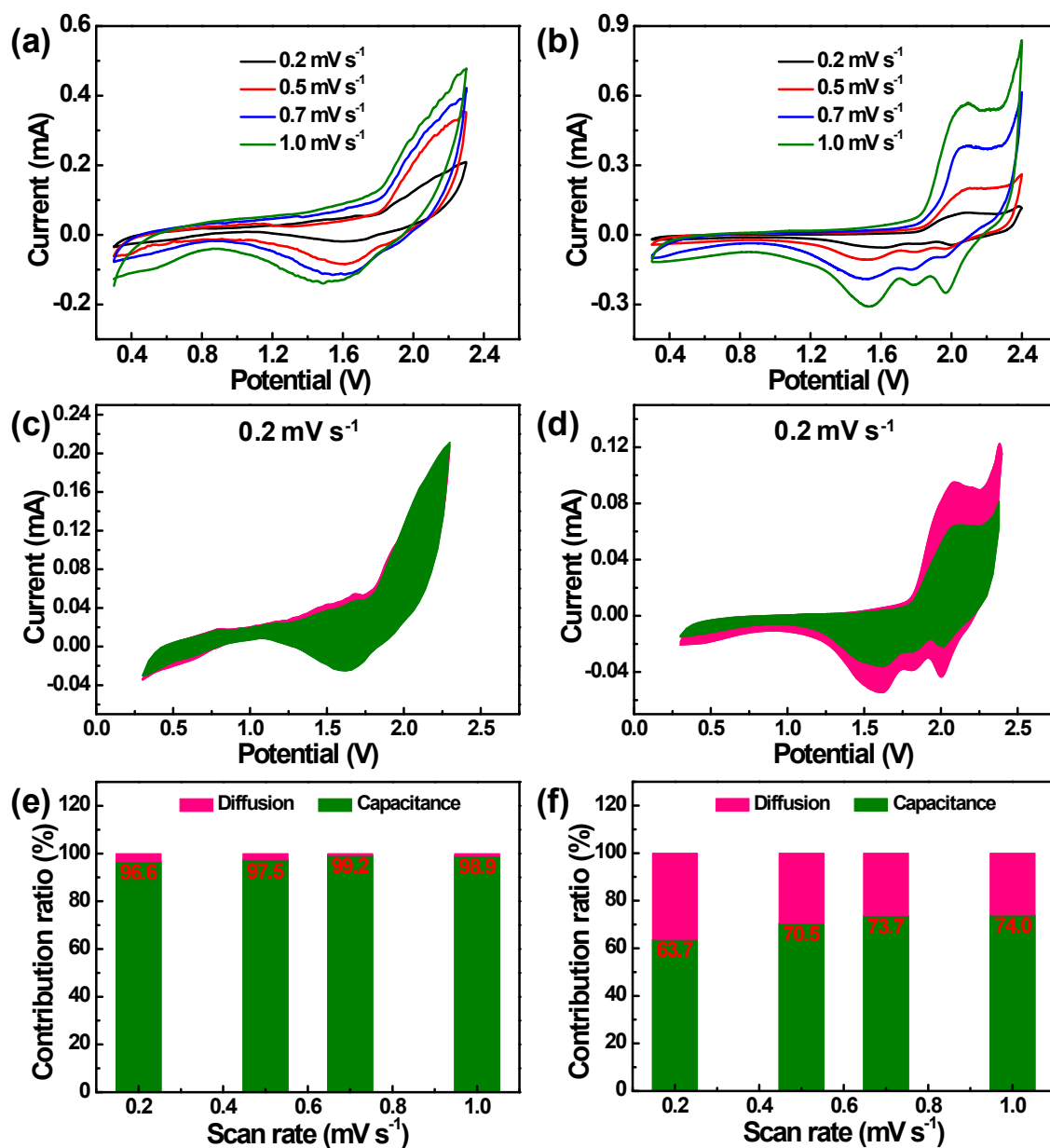
**Fig. S9.** SEM images (a,b), TEM image (c) and Raman spectra (d) of the as-prepared sample by only immersing amorphous carbon into  $\text{CaCl}_2\text{-LiCl}$  salt at 850 °C for 3 h.



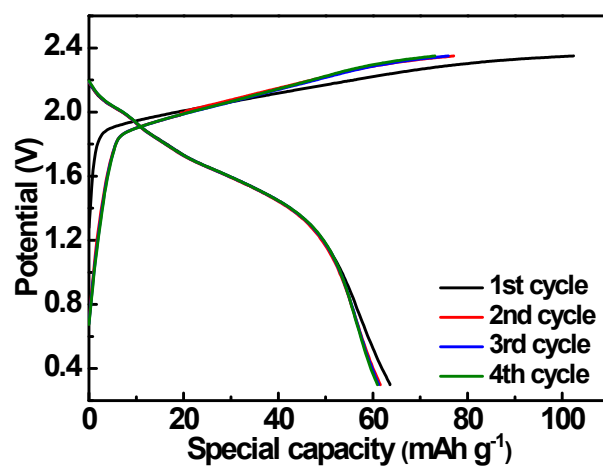
**Fig. S10.** CV curves of AC (a) and GNF7 (b) from 2<sup>nd</sup> to 4<sup>th</sup> cycle at a scan rate of 0.5 mV s<sup>-1</sup>. The charge-discharge curves for the first four cycles of AC (c) and GNF7 (d) at a current density of 100 mA g<sup>-1</sup> under the potentials ranging from 0.3 to 2.3 V.



**Fig. S11.** Nyquist plots (a) and the relationship between  $Z_{Re}$  and  $\omega^{-1/2}$  (b) of AC and GNF-7 electrodes over 10 cycles.



**Fig. S12.** Kinetic analysis of the AC (a,c,e) and GNF7 (b,d,f) cathodes. CV curves at various scan rates (a,b). Separation of the capacitive and diffusion-controlled contributions at a scan rate of 0.2  $\text{mV s}^{-1}$  (c,d). Contribution ratio of the capacitive and diffusion-controlled at different scan rates (e,f).



**Fig. S13.** The charge-discharge curves for the first four cycles of GNF7 cathode at a current density of 200 mA g<sup>-1</sup>.

**Table S1.** The calculated  $I_D/I_G$  of amorphous carbon before and after cathodic electrolysis under different conditions.

| Samples No. | Conditions       | D peak position<br>( $\text{cm}^{-1}$ ) | G peak position<br>( $\text{cm}^{-1}$ ) | $I_D/I_G$ |
|-------------|------------------|-----------------------------------------|-----------------------------------------|-----------|
| AC          | Amorphous Carbon | 1309                                    | 1571                                    | 1.054     |
| GNF1        | 800 °C-2.6V-2h   | 1345                                    | 1575                                    | 0.324     |
| GNF2        | 800 °C-2.7V-2h   | 1345                                    | 1579                                    | 0.236     |
| GNF3        | 800 °C-2.8V-2h   | 1351                                    | 1581                                    | 0.223     |
| GNF4        | 800 °C-3.0V-2h   | 1352                                    | 1582                                    | 0.201     |
| GNF5        | 850 °C-2.8V-0.5h | 1349                                    | 1571                                    | 0.118     |
| GNF6        | 850 °C-2.8V-1h   | 1344                                    | 1572                                    | 0.117     |
| GNF7        | 850 °C-2.8V-2h   | 1356                                    | 1583                                    | 0.112     |
| GNF8        | 700 °C-2.7V-2h   | 1330                                    | 1575                                    | 0.969     |
| GNF9        | 700 °C-3.0V-2h   | 1343                                    | 1571                                    | 0.579     |
| GNF10       | 750 °C-3.0V-2h   | 1337                                    | 1573                                    | 0.340     |

**Table S2.** The calculated graphitization degree of amorphous carbon before and after cathodic electrolysis under different conditions.

| Samples No. | Conditions       | 2 $\theta$ (°) | d <sub>002</sub> (Å) | g (%)    |
|-------------|------------------|----------------|----------------------|----------|
| AC          | Amorphous Carbon | 25.219         | 3.529271             | -103.804 |
| GNF1        | 800 °C-2.6V-2h   | 25.986         | 3.426098             | 16.1651  |
| GNF2        | 800 °C-2.7V-2h   | 25.992         | 3.425321             | 17.0689  |
| GNF3        | 800 °C-2.8V-2h   | 26.008         | 3.42325              | 19.477   |
| GNF4        | 800 °C-3.0V-2h   | 26.013         | 3.422603             | 20.229   |
| GNF5        | 850 °C-2.8V-0.5h | 26.032         | 3.420148             | 23.0836  |
| GNF6        | 850 °C-2.8V-1h   | 26.112         | 3.409851             | 35.0571  |
| GNF7        | 850 °C-2.8V-2h   | 26.186         | 3.40038              | 46.0669  |
| GNF9        | 700 °C-3.0V-2h   | 25.706         | 3.462778             | -26.486  |
| GNF10       | 750 °C-3.0V-2h   | 25.78          | 3.453006             | -15.123  |
| GNF11       | 700 °C-3.0V-5h   | 26.004         | 3.423767             | 18.8753  |



**Table S3.** Comparison between this work and the previous reported graphite materials by different synthetic approaches.

| Synthetic approaches of graphite | Carbon sources                                                                                                                                           | Assisted by catalysts | Product purity | Temperature (°C) | Energy consumption (kWh kg <sup>-1</sup> ) | BET surface area (m <sup>2</sup> g <sup>-1</sup> ) | Pore volume (cm <sup>3</sup> g <sup>-1</sup> ) | I <sub>D</sub> /I <sub>G</sub> | d <sub>002</sub> (Å) | Ref.      |
|----------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------|----------------|------------------|--------------------------------------------|----------------------------------------------------|------------------------------------------------|--------------------------------|----------------------|-----------|
| Thermal annealing                | Only a few graphitizable precursors                                                                                                                      | None catalysts        | High           | 2200             | 32.1                                       | ---                                                | ---                                            | ---                            | 3.39                 | 1         |
|                                  |                                                                                                                                                          |                       |                | >2500            | ---                                        | ---                                                | ---                                            | 0.30                           | 3.39                 | 2         |
|                                  |                                                                                                                                                          |                       |                | 2300             | ---                                        | 260                                                | 0.34                                           | ---                            | 3.42                 | 3         |
| Microwave heating                | Most amorphous carbon precursors                                                                                                                         | Ni, Fe, Co catalysts  | Medium         | 1170             | ---                                        | ---                                                | ---                                            | ---                            | 3.43                 | 4         |
|                                  |                                                                                                                                                          |                       |                | 1418             | 3.0                                        | ---                                                | ---                                            | 0.30                           | 3.42                 | 5         |
| Chemical vapor deposition        | CH <sub>4</sub> , C <sub>2</sub> H <sub>2</sub> , C <sub>2</sub> H <sub>4</sub> , C <sub>6</sub> H <sub>6</sub> , CH <sub>4</sub> +H <sub>2</sub> , etc. | Ni, Fe, Co catalysts  | Medium         | 950              | ---                                        | ---                                                | ---                                            | 0.66                           | ---                  | 6         |
|                                  |                                                                                                                                                          |                       |                | >1000            | ---                                        | ---                                                | ---                                            | ---                            | 3.35                 | 7         |
|                                  |                                                                                                                                                          |                       |                | 700              | 5.5                                        | 109                                                | ---                                            | 1.50                           | 3.41                 | 8         |
|                                  |                                                                                                                                                          |                       |                | 850              | ---                                        | 870                                                | 0.80                                           | 0.62                           | 3.45                 | 9         |
| Catalytic graphitization         | Most amorphous carbon precursors                                                                                                                         | Ni, Fe, Co catalysts  | Low            | 650              | ---                                        | 760                                                | 0.67                                           | ---                            | 3.40                 | 10        |
|                                  |                                                                                                                                                          |                       |                | 900              | ---                                        | 275                                                | 0.22                                           | ---                            | 3.35                 | 11        |
|                                  |                                                                                                                                                          |                       |                | 1000             | 12.8                                       | 907                                                | 0.64                                           | 0.40                           | ---                  | 12        |
| Molten-salt electrolysis         | All amorphous carbon precursors                                                                                                                          | None catalysts        | High           | 820              | 5.5                                        | ---                                                | ---                                            | 0.11                           | ---                  | 13        |
|                                  |                                                                                                                                                          |                       |                | >700             | 4.5                                        | 165.7                                              | 0.83                                           | 0.11                           | 3.40                 | This work |

**Table S4.** The physical properties of amorphous carbon before and after cathodic electrolysis under different conditions.

| Samples No. | Conditions       | Specific surface area (m <sup>2</sup> g <sup>-1</sup> ) | Pore volume (mm <sup>3</sup> g <sup>-1</sup> ) | Pore diameter (Mode Dv(d)) (nm) | Average pore diameter (nm) |
|-------------|------------------|---------------------------------------------------------|------------------------------------------------|---------------------------------|----------------------------|
| AC          | Amorphous Carbon | 108.00                                                  | 349.40                                         | 2.45                            | 17.00                      |
| GNF7        | 850 °C-2.8V-2h   | 165.70                                                  | 825.80                                         | 2.46                            | 39.17                      |

## References

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