Supporting Information for

Introducing catalytic gasification into chemical activation for the conversion of natural coal into hierarchically porous carbons with broadened pore size for enhanced supercapacitive utilization

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Table S1. Proximate and ultimate analyses of Zhundong coal.

| Proximate analysis (wt-%) | Ultimate analysis (wt-%, daf) |
|--------------------------|-------------------------------|
| $M_{ad}$ | $A_{ad}$ | $FC_{ad}$ | $V_{daf}$ | C | H | O* | N | S |
| 11.79 | 3.68 | 56.64 | 32.70 | 73.52 | 6.55 | 18.51 | 0.91 | 0.51 |

$^{ad}$ Air-dried basis, $^{d}$ dry basis, $^{daf}$ dry and ash-free basis, $^{*}$ by difference.

Table S2. Ash composition analyses of Zhundong coal.

| Ash composition analysis (wt-%) |
|-----------------------------|
| SiO$_2$ | Al$_2$O$_3$ | Fe$_2$O$_3$ | TiO$_2$ | CaO | MgO | K$_2$O | Na$_2$O | MnO$_2$ | SO$_3$ | P$_2$O$_5$ |
| 13.33 | 10.71 | 6.19 | 0.47 | 37.75 | 9.98 | 0.62 | 9.78 | 0.16 | 6.52 | 0.19 |

Fig. S1. HRTEM images of Zhundong raw coal (a); MPC-500 (b) and HPC (c). All the HRTEM images demonstrate the mainly amorphous carbon nature.
Fig. S2 (a) XRD patterns of prepared porous carbons; (b) Raman spectra of prepared porous carbons. Both XRD and Raman spectra suggest that the obtained porous carbons are mainly amorphous which is in agreement with the HRTEM results (Fig. S1). However, the differences between prepared porous carbons reveal that MPC-500 prepared under lower temperature (500) shows a relatively higher graphitization degree (higher 002 peak in XRD and G-to-D band ratio in Raman) that those of other samples. This is because high temperature treatments destroy the graphite-like crystal structure of raw coal and lead to the resulting MPC-950, HPC and MF-PC with lower graphitization degree.
Fig. S3 (a) CV curves of MPC-950 at various scan rates; (b) Galvanostatic charge-discharge curves of MPC-950 under different charge-discharge current densities.
Fig. S4 Comparison of MF-PC and MPC-950 in three electrode system using 6 M KOH as electrolyte. Without the mineral component in the coal structure, MF-PC mainly have micropores. Owing to the larger BET surface area, MF-PC perform much better performance compared with MPC-950. However, it provides both poorer gravimetric capacity and rate capacity compared with HPC.
Table S3. Typical results of carbon materials for three-electrode test in literatures with aqueous electrolyte systems

| Sample                                      | Electrolyte | System voltage | Rate performance | Ref. |
|---------------------------------------------|-------------|----------------|------------------|------|
|                                              |             | voltage        | Capacitance (F g⁻¹) | Current (A g⁻¹) |      |
| Nitrogen-doped Interconnected Carbon Nanosheets | 2 M KOH     | 0~1.8 vs. SCE  | 260              | 1     | 20   | S1   |
| Nitrogen-doped Porous Carbon                | 2 M KOH     | 0~1 vs. SCE    | 255              | 1     | 10   | S2   |
| Hierarchically Porous Functional Biomass Carbons | 1 M KOH     | -1~0 vs. SCE   | 281              | 0.2   | 4    | S3   |
| Nitrogen-doped Porous Graphitic Carbon      | 6M KOH      | -1.2~0.2 vs. SCE | 293          | 1     | 30   | S4   |
| Nitrogen-doped porous nanofibers            | 6 M KOH     | -1~0 vs. SCE   | 202              | 1     | 30   | S5   |
| Nitrogen-doped Ordered Mesoporous Carbon    | 6M KOH      | -0.9~0 vs. SCE | 227              | 0.2   | 2.2  | S6   |
|                                              | 1 M H₂SO₄   | 0~0.8 vs. SCE  | 262              | 0.2   | 2.2  |      |
| Human hair-derived carbon flakes            | 6M KOH      | -1~0 vs. SCE   | 128              | 80    |      | S7   |
| Shape-controlled carbon nanosheets          | 1 M H₂SO₄   | 0~1 vs. Ag/AgCl| 145              | 30    |      | S8   |
| Two-dimensional Porous Carbon Nanosheets    | 6M KOH      | -1~0 vs. SCE   | 300              | 0.5   | 100  | S9   |
| Yeast Cells Derived Carbon                  | 6M KOH      | -1.2~0.2 vs. Ag/AgCl | 175    | 100   |      | S10  |
| Hierarchical Porous Carbon Sheets from Coal Tar Pitch | 6M KOH     | -1~0 vs. SCE   | 290              | 1     | 10   | S11  |
| HPC                                         | 6M KOH      | -1~0 vs. SCE   | 308              | 1     | 100  | Our work |
**Table S4.** Typical results of carbon materials as cathode for lithium ion capacitors with organic electrolyte systems

| Sample                        | Electrolyte | **System voltage** | **Rate performance** | **Cycling Stability** | Ref. |
|-------------------------------|-------------|--------------------|----------------------|-----------------------|------|
| Nitrogen-Doped Porous Carbon  | 1 M LiPF$_6$ | 2.5-4.5 V vs. Li/Li$^+$ | 117 F g$^{-1}$(81.5 mAh g$^{-1}$) (0.1 A g$^{-1}$) 60.8 F g$^{-1}$(42.3 mAh g$^{-1}$) (30 A g$^{-1}$) | 86% after 2000 cycles (5 A g$^{-1}$) | S12  |
| Activated Carbons             | 1 M LiPF$_6$ | 3.0-4.6 V vs. Li/Li$^+$ | 159 F g$^{-1}$(110.6 mAh g$^{-1}$) (0.1 A g$^{-1}$) | ~82% after 1000 cycles (0.1 A g$^{-1}$) | S13  |
| 3D Carbon Nanofibers          | 1 M LiPF$_6$ | 2.0-4.5 V vs. Li/Li$^+$ | 162 F g$^{-1}$(113 mAh g$^{-1}$) (0.1 A g$^{-1}$) 90.6 F g$^{-1}$(63 mAh g$^{-1}$) (10 A g$^{-1}$) | 87% after 5000 cycles (2 A g$^{-1}$) | S14  |
| LTO/Graphene hybrid           | 1 M LiPF$_6$ | 1-4 V vs. Li/Li$^+$ | 69 F g$^{-1}$(178 mAh g$^{-1}$) (0.25 A g$^{-1}$) 58 F g$^{-1}$(120 mAh g$^{-1}$) (10 A g$^{-1}$) | 95% after 1000 cycles (5 A g$^{-1}$) | S15  |
| HPC                           | 1 M LiPF$_6$ | 2.0–4.5 V vs. Li/Li$^+$ | 183 F g$^{-1}$(127 mAh g$^{-1}$) (0.25 A g$^{-1}$) 112 F g$^{-1}$(78 mAh g$^{-1}$) (10 A g$^{-1}$) | 88% after 5000 cycles (2 A g$^{-1}$) | **Our work** |
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