

**Electronic Supplementary Material (ESI) for**

**Hierarchically aminated graphene honeycombs for  
electrochemical capacitive energy storage**

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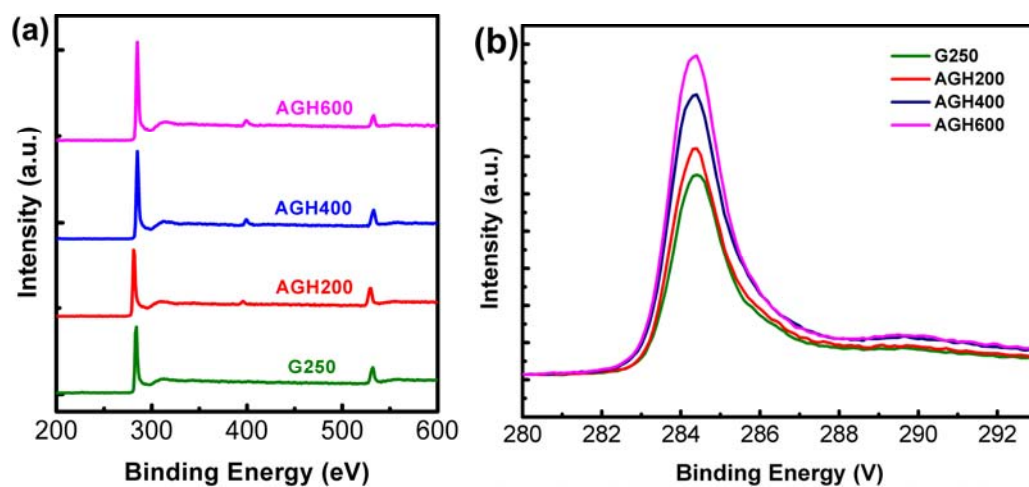
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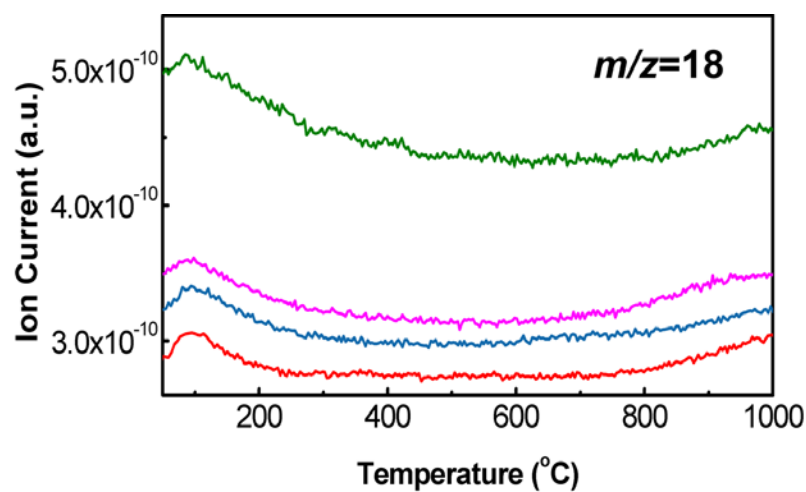
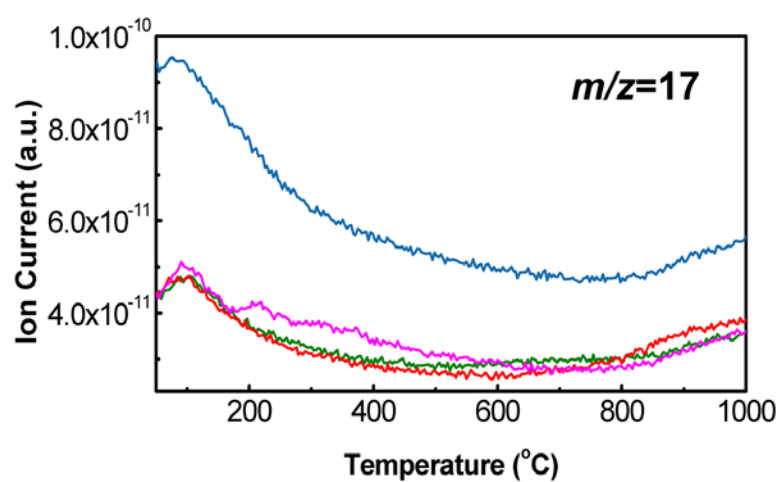
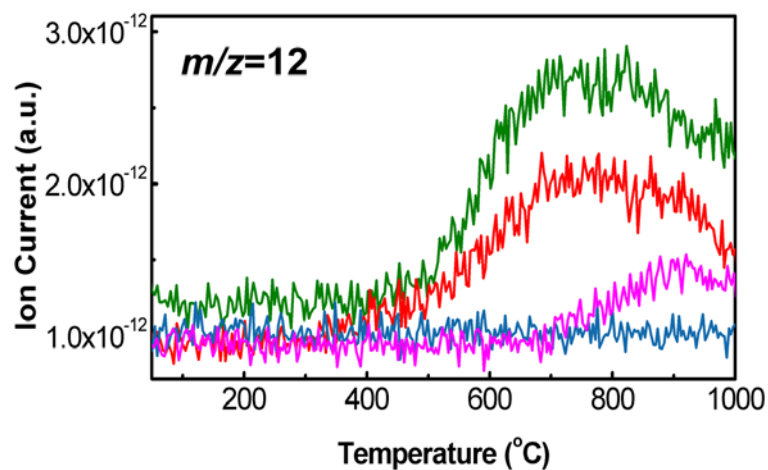
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**Fig. S1** (a) XPS survey spectrum and (b) C1s fine scan spectrum of G250 and AGHs



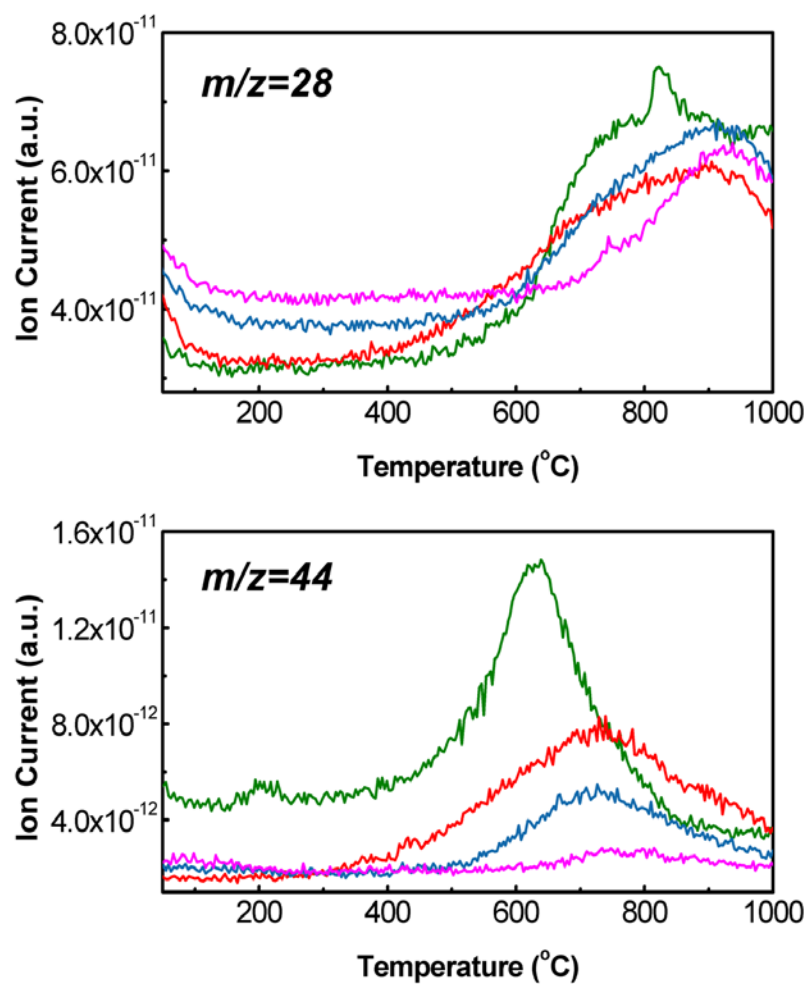
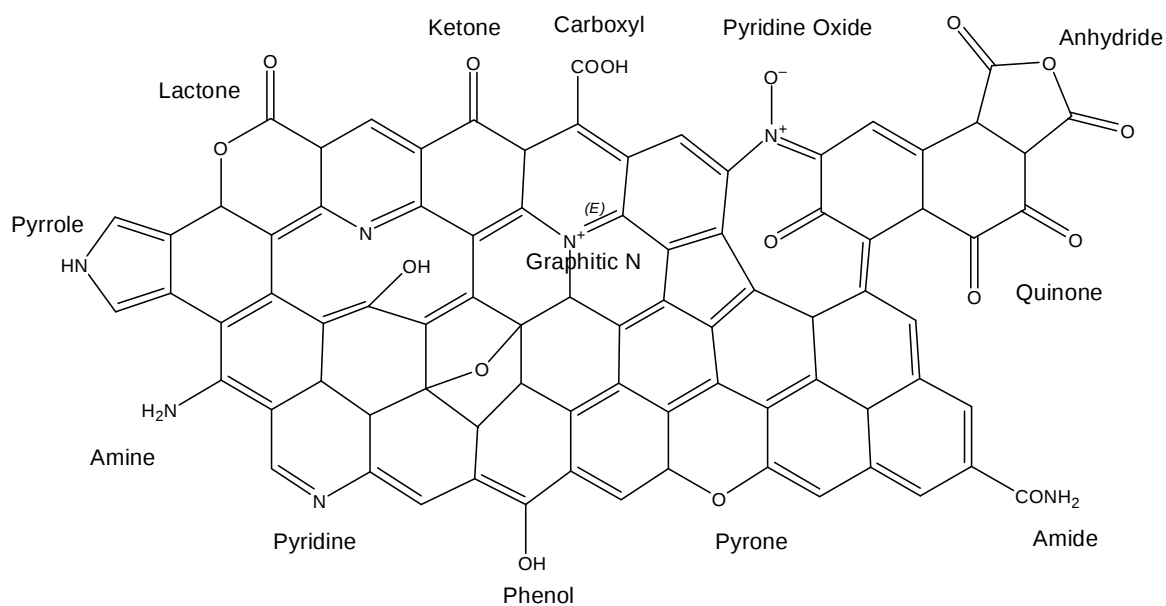


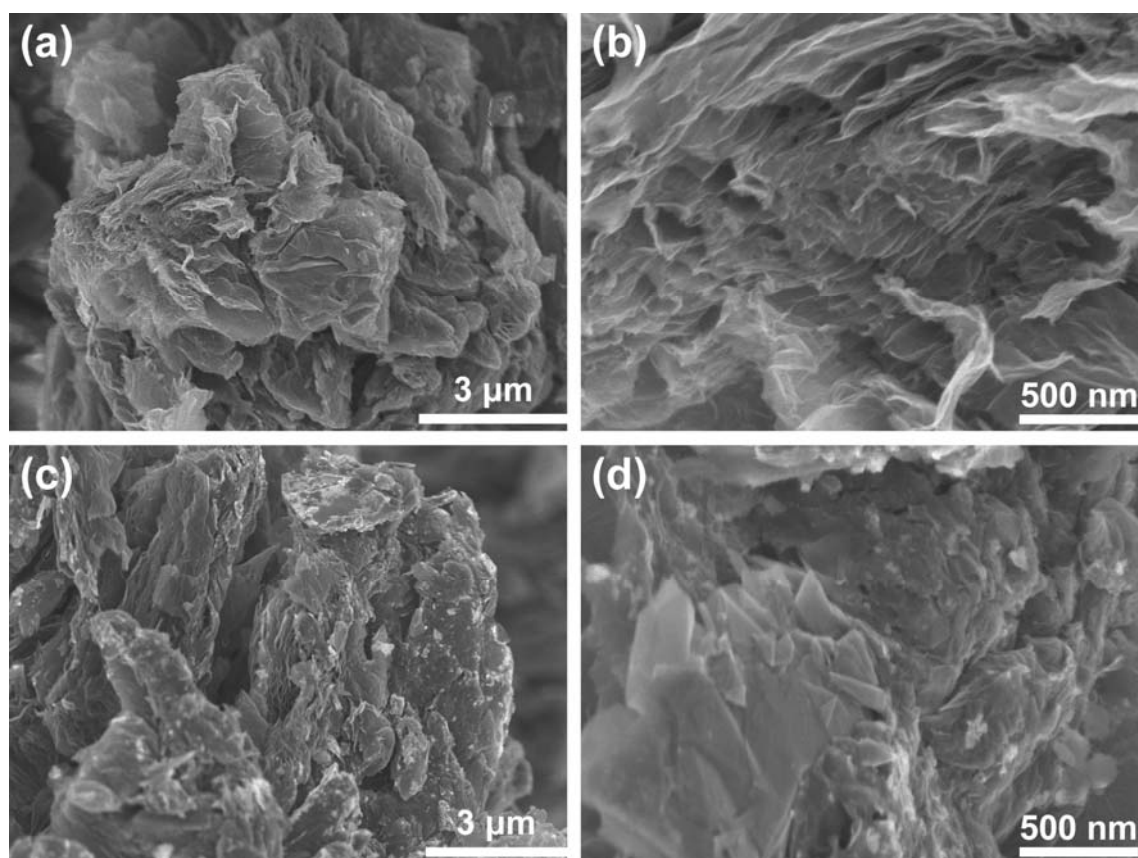
Fig. S2 MS curves of the sweep gases fragments from TG

**Table S1** Assignments of TG-MS peaks

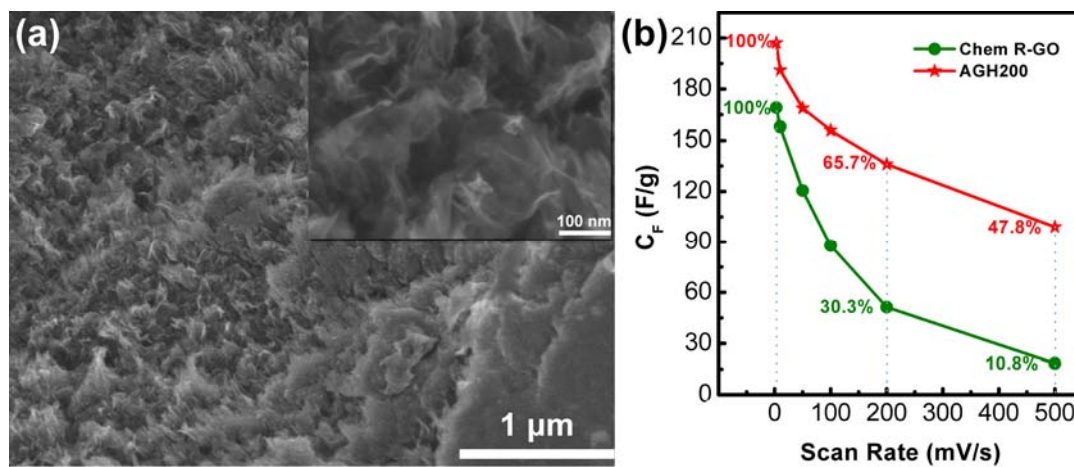
| m/z | Assignments                                                                                                                                                                                           | Evolving Temperature                    |
|-----|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------|
| 12  | Radical C evolved from re-graphitization or crystallization of amorphous sp <sup>3</sup> region in graphene                                                                                           | 550-1000 °C                             |
| 17  | NH <sub>3</sub> from decomposition of nitrogen containing groups (especially for amine/amide species)<br>Radical OH fragments from H <sub>2</sub> O (physically adsorbed water/ chemical dehydration) | 200-500 °C<br>a) ~100 °C;<br>b) >800 °C |
| 18  | H <sub>2</sub> O from evaporation of physically adsorbed water around 100 °C<br>intermolecular dehydration between neighboring -OH, -COOH and -NH <sub>2</sub> pairs                                  | ~100 °C<br>>800 °C                      |
| 28  | CO from decomposition of thermally stable -C=O related carbonyl groups, individual -OH or epoxy sites at relatively higher temperature                                                                | 650-1000 °C                             |
| 44  | CO <sub>2</sub> from desorption of C(O)O related species such as carboxyl, lactone and anhydride                                                                                                      | 200 °C                                  |



**Fig. S3** Different forms of N and O functionalities in aminated carbon materials.



**Fig. S4** Collapse of large pores in AGHs via high temperature amination: SEM images of (a,b) AGH400 and (c,d) AGH600.



**Fig. S5** Chemically reduced graphene oxide (CRG) by  $\text{NH}_2\text{-NH}_2$ : (a) SEM images of compact graphene agglomeration without exterior macropores; (b) Electrochemical performance as supercapacitor electrode.

**Table S2** Summary of supercapacitor performance based on carbon electrodes

| Carbon type             | BET surface area (m <sup>2</sup> /g) | Scanning rate or cycling rate | C <sub>F</sub> (F/g) | C <sub>s</sub> (F/m <sup>2</sup> ) | Reference |
|-------------------------|--------------------------------------|-------------------------------|----------------------|------------------------------------|-----------|
| G250                    | 293                                  | 3 mV/s                        | 174                  | 0.59                               | This work |
| AGH200                  | 247                                  | 3 mV/s                        | 207                  | 0.84                               | This work |
| Activated carbon        | 543                                  | 5 mV/s                        | 184                  | 0.34                               | [1]       |
| LN <sup>a</sup>         | 746                                  | 0.2 A/g                       | 264                  | 0.35                               | [2]       |
| LN+CNT                  | 680                                  | 0.2 A/g                       | 273                  | 0.40                               | [2]       |
| Activated carbon        | 1400                                 | 0.2 A/g                       | 119                  | 0.085                              | [2]       |
| Mesoporous carbons      | 702                                  | 0.2 A/g                       | 112                  | 0.16                               | [3]       |
| B0.7-OMC                | 641                                  | 0.2 A/g                       | 134.6                | 0.21                               | [3]       |
| P0.7-OMC                | 550                                  | 0.2 A/g                       | 154                  | 0.28                               | [3]       |
| N-activated carbon      | 571                                  | 0.1 A/g                       | 220                  | 0.39                               | [4]       |
| Graphene                | 202                                  | 10 mV/s                       | 135                  | 0.67                               | [5]       |
| Graphene-CNT            | 612                                  | 10 mV/s                       | 385                  | 0.63                               | [5]       |
| Graphene                | 705                                  | 20 mV/s                       | 100                  | 0.14                               | [6]       |
| Graphene                | 382                                  | 1 mV/s                        | 279                  | 0.73                               | [7]       |
| N-activated carbon      | 635                                  | 0.1 A/g                       | 210                  | 0.33                               | [8]       |
| Carbide derived carbons | 600-2000                             | -                             | 70-190               | 0.05-0.12                          | [9]       |
| HOPG                    | -                                    | -                             | -                    | 0.5-0.7 <sup>b</sup>               | [10]      |
| Graphite powder         | 4                                    | -                             | 1.4                  | 0.35                               | [10]      |
| Carbon aerogel          | 650                                  | -                             | 149.5                | 0.23                               | [10]      |
| Nanodiamond             | 380                                  | -                             | 15.2                 | 0.04                               | [11]      |
| Carbon nanotube         | 200                                  | -                             | 18                   | 0.09                               | [11]      |
| Activated carbon        | 1150-2300                            | -                             | 27-100               | 0.005-0.041                        | [12]      |
| SWCNTs                  | 357                                  | 1 mA/cm <sup>2</sup>          | 138                  | 0.39                               | [13]      |
| MWCNTs                  | 19.7                                 | 1 mA/cm <sup>2</sup>          | 2                    | 0.10                               | [14]      |
| Activated MWCNTs        | 247                                  | 1 mA/cm <sup>2</sup>          | 14                   | 0.056                              | [14]      |

<sup>a</sup> Activated carbon obtained after pyrolysis at 600 °C of the *Lessonia Nigrescens* seaweed

<sup>b</sup> Measured at the edge plane of highly oriented pyrolytic graphite.

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