**Multiple Functional Base-Induced Highly Ordered Graphene Aerogels**

*Dongfang Dai,*[1] *Yan Zhou,*[1] *Wenqiu Xiao,*[2] *Jie Wang,* [1] *Zhenliang Hao,*[1] *Hui, Zhang,*[1] *Jinming Cai** [1,2]

1 Faculty of Material Science and Engineering, Low Dimensional Nanomaterials Vacuum Laboratory, Kunming University of Science and Technology, Kunming 650093, China.

2 Dongguan D&R Graphene Institute, Dongguan 523170, China.

* Corresponding Author: j.cai@kust.edu.cn (J. Cai)

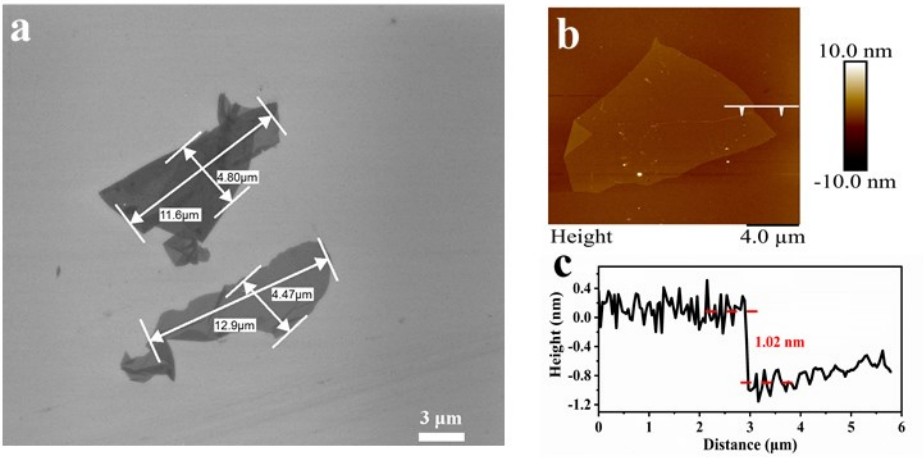


Figure S1. SEM (a) and AFM (b) images of GO sheets. (c) Height profile of a GO sheet.

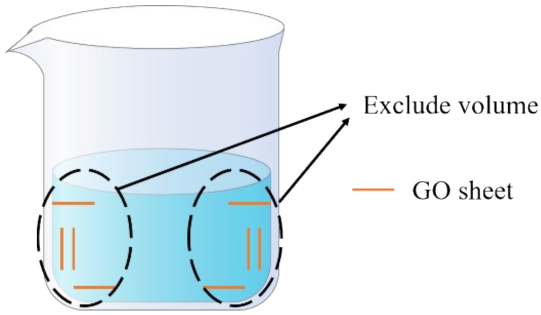


Figure S2. Schematic illustration of surface anchoring effect.

The orientation of GO sheets tends to be parallel to the solid surface, which has been reported in previous literature. It is mainly an entropic effect that parallel arrangement could minimize the excluded volume, thus minimize the orientational free energy per unit area according to the following equation:

$$G = nkTd\sin\theta$$

where G, n, d and θ represent the orientational free energy, number density of GO sheets, lateral dimension of GO sheets, and the angle between GO sheets and surface. GO sheets tend to align parallel to the surface ($\theta = 0°$), resulting in the minimum orientational free energy.

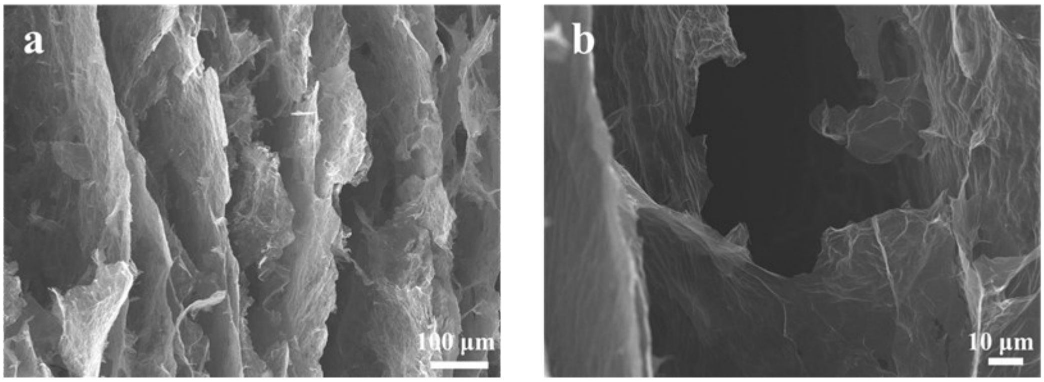


Figure S3. (a) and (b) were the SEM of the longitudinal section of HOGA.

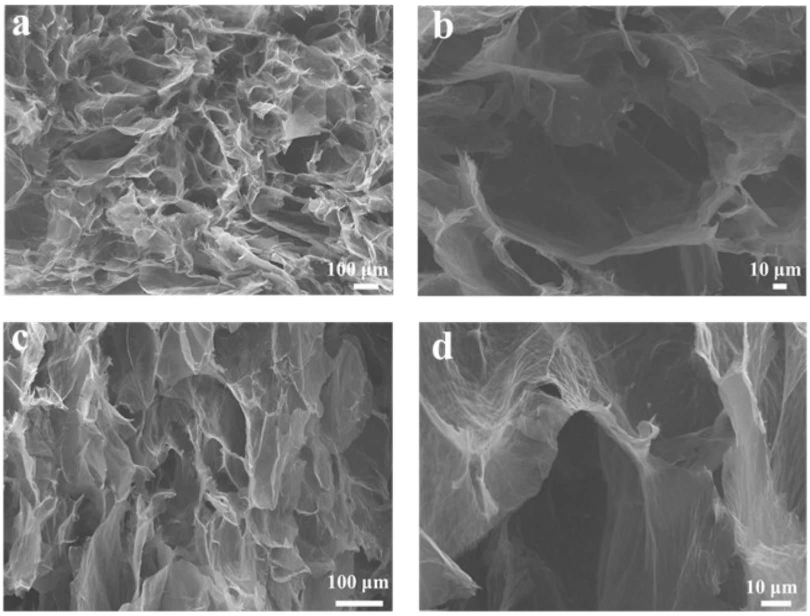


Figure S4. The SEM images of LAAGA. (a) and (b) were the cross section; (c) and (d) were the longitudinal section.

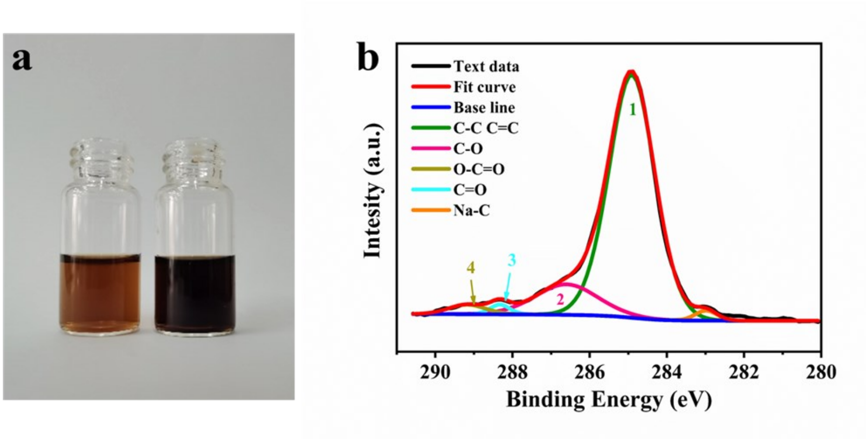


Figure S5. (a) Photographs of GO and GO + NaOH suspensions. The solution turned black quickly after adding NaOH. (b) C 1s XPS spectra of GO + NaOH.

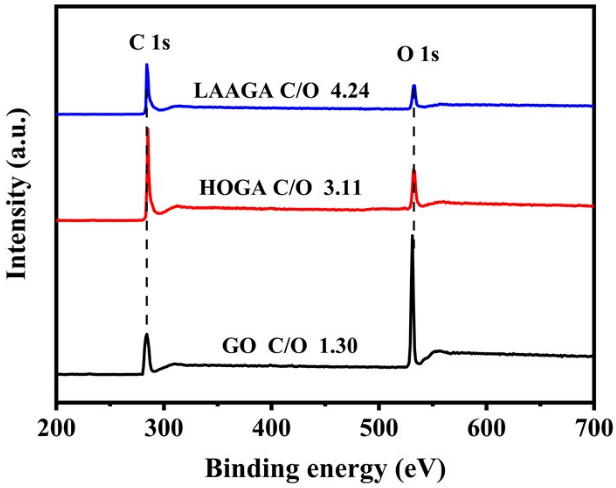


Figure S6. XPS spectrum of GO, HOGA and LAAGA, and the ratios of C/O were 1.30, 3.11 and 4.24 respectively.

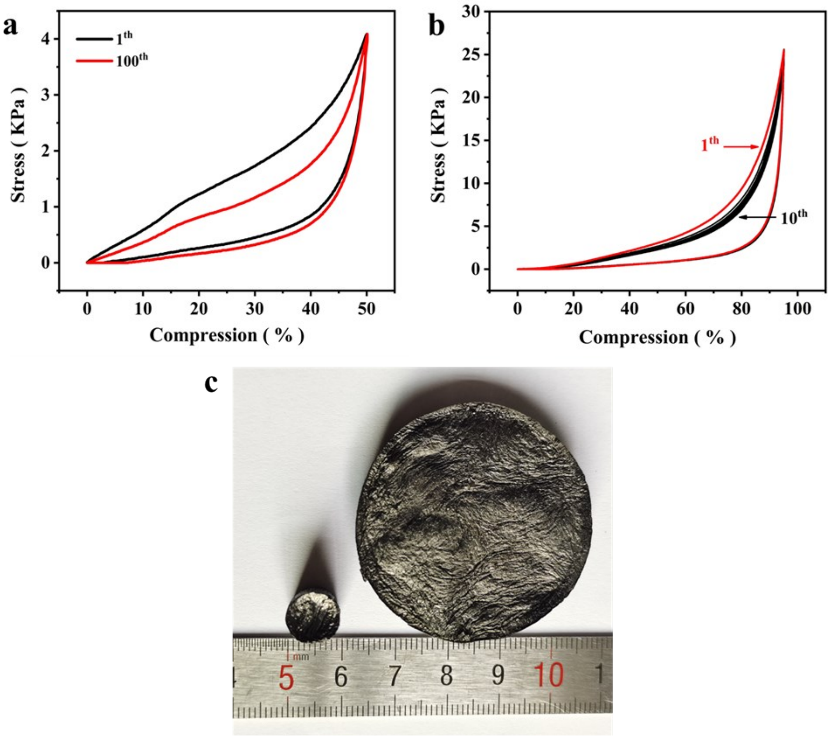


Figure S7. a) The compressive stress–strain curves of large HOGA at 50 % compression 100 cycles remained maximum stress. b) The compressive stress–strain curves of large HOGA at 95 % compression 10 cycles remained maximum stress. c) Photos of HOGA and large HOGA.

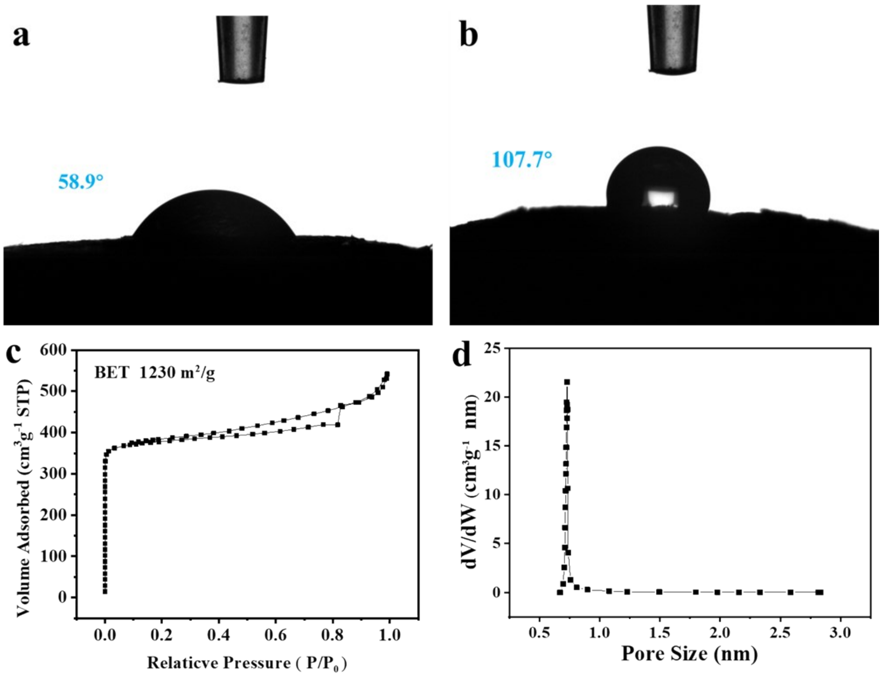


Figure S8. Water contact angle of GO a) and HOGA b). c) BET of the HOGA and d) is the micropore distribution of the HOGA.

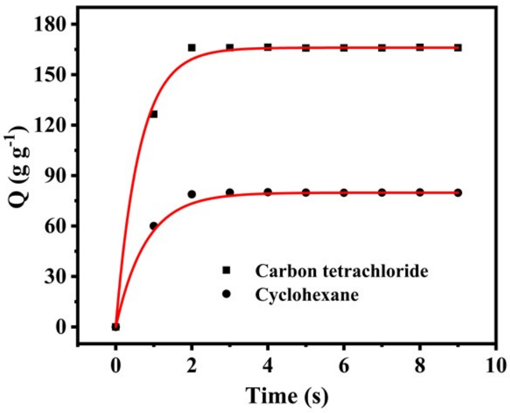

Figure S9. Adsorbtion curves of carbon tetrachloride and cyclohexane by HOGA.

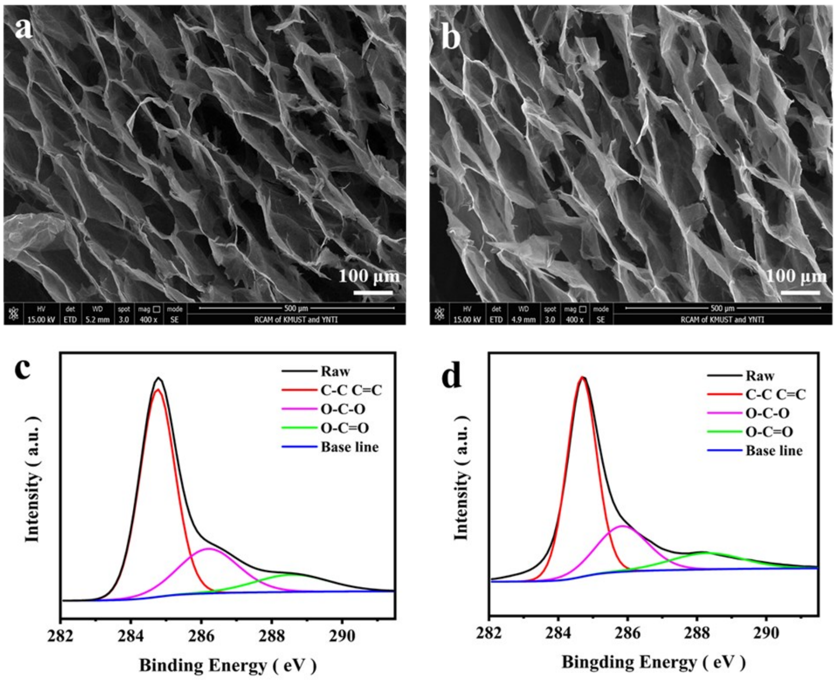


Figure S10. SEM images (a, b) and XPS spectra (c, d) of HOGA after absorption-squeezing and absorption-combustion cycles, respectively.

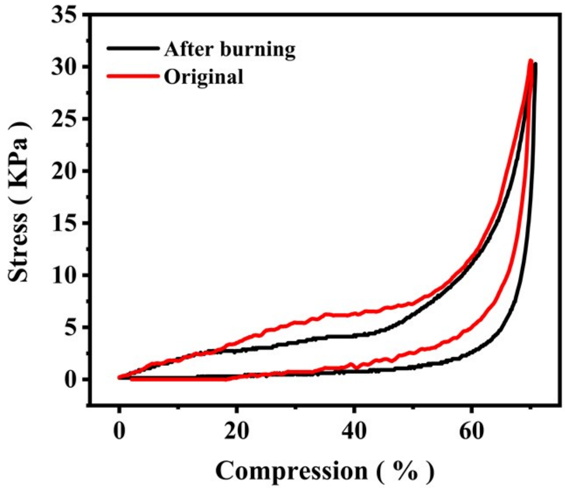

Figure S11. The stress-strain curves of HOGA before and after burning.

Table 1. Adsorption capacities (Q) of the HOGA measured for a range of organic solvents in terms of their densities and sorts.

| Organics | Density (g/mL) | Q (g/g) |
|---|---|---|
| Cyclohexane | 0.78 | 80 |
| Acetone | 0.788 | 82.5 |
| Diesel | 0.84 | 81 |
| Toluene | 0.87 | 83 |
| Ethyl acetate | 0.902 | 82.64 |
| benzene | 0.88 | 86.37 |
| ethanol | 0.789 | 83.64 |
| DMF | 0.945 | 93.2 |
| Anisole | 0.996 | 96.56 |
| NMP | 1.028 | 97.49 |
| Ethylene glycol | 1.116 | 126.73 |
| Dichloromethane | 1.325 | 130.94 |
| Carbon tetrachloride | 1.595 | 165.98 |