Supporting Information

A Facile Method to Prepare Highly Compressible Three-Dimensional Graphene-only Sponge

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1. AFM images of the GO sheets.

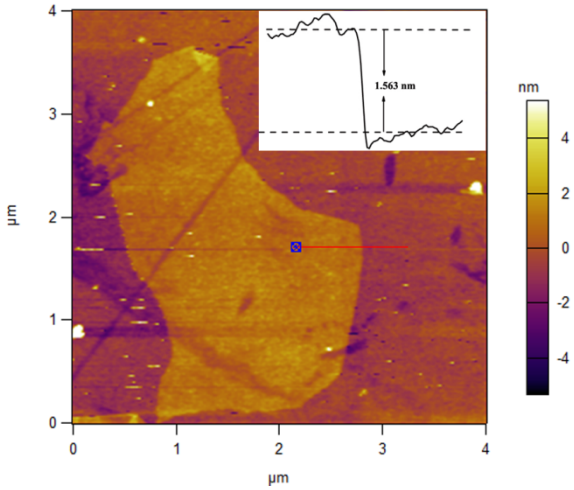

Figure S1 AFM image of the GO sheets used in this work.

2. The reducibility of ammonium sulfide

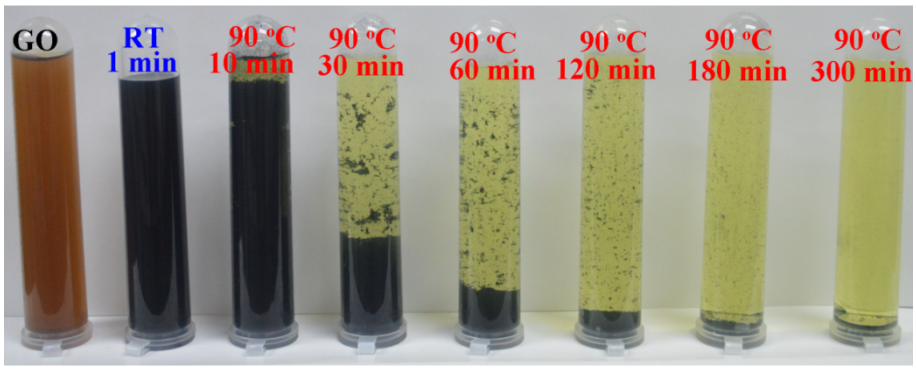

Figure S2. Digital images of the reduction of GO by ammonium sulfide at different time.

The reduction of GO by ammonium sulfide was investigated by the color and the dispersion changes in the water. From Figure S2 we can see that the GO solution was yellow and after the addition of ammonium sulfide the color change quickly to black in just 1 minute. When the reduction happened at 90 °C, the dispersion is deterioration, and the resultant rGO aggregated together and dropped down to the bottom which means the organic groups of GO was reduced to yield graphene. The result was agreed with the conclusion from FTIR spectra.

3. The formation of graphene hydrogels at room temperature

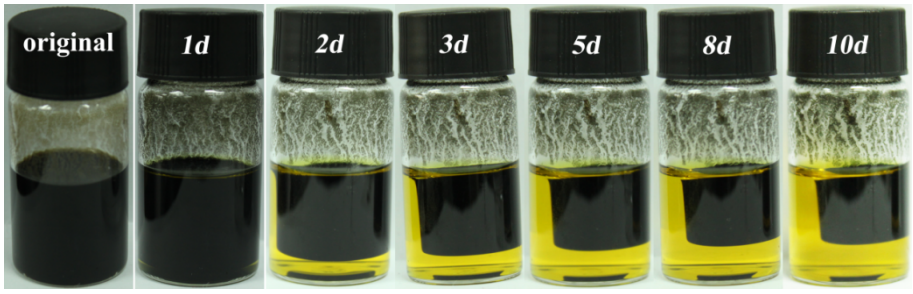

Figure S3. The formation of graphene hydrogel at room temperature.

In order to study the reduction and self-assembly process at room temperature, 1mL of ammonium sulfide solution was added into 10 mL (2 mg mL^{-1}) GO solution (the mass ratio of GO to ammonium sulfide was 1: 10) and kept still in the following few days. The graphene hydrogel was formed obviously after one day at room temperature and as the time went on the hydrogel became more and more smaller. On the eighth day of the self-assembly process the hydrogel could hardly change any more and the volume was almost the same with the hydrogel formed at 90 °C for 3 h (Figure S7). Thus we believe the same amount ammonium sulfide solution could form the same graphene hydrogel at room temperature.

4. Compressive properties of GS and CGS

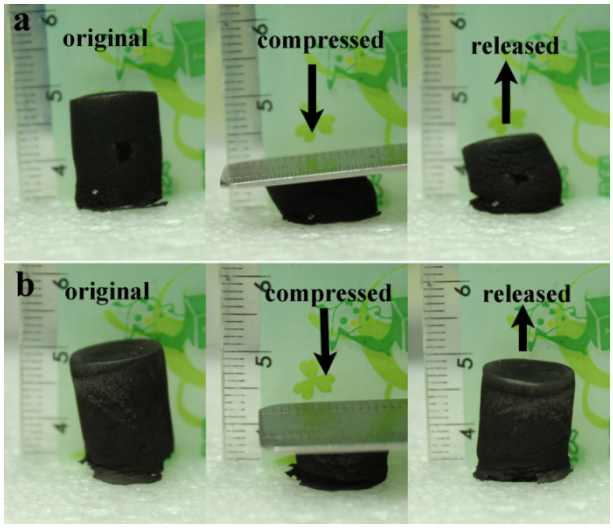

Figure S4. Compress properties of (a) GS, (b) CGS.

The graphene sponge prepared by the graphene hydrogel could hold the 3D structure but it wasn't compressible as shown in Figure S4a. While after treated by ammonium solution the graphene sponge become compressible as shown in Figure S4b.

5. Stress-Strain curves of as-prepared graphene-only sponge at different mass ratio of GO to ammonium sulfide.

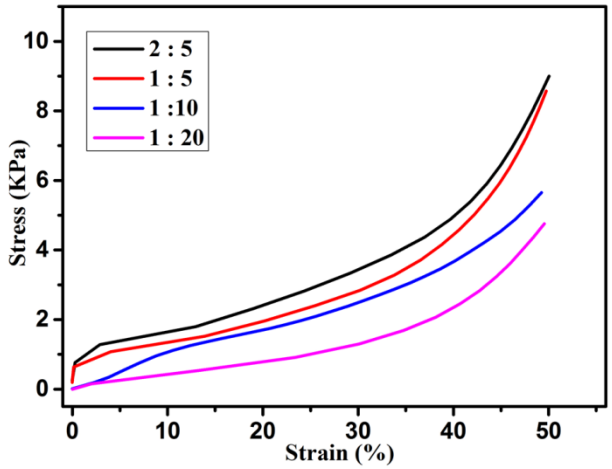

Figure S5 Stress-Strain curves of as-prepared graphene-only sponge at different mass ratio of GO to ammonium sulfide from 2:5 to 1:20.

6. TGA of graphite, GO and CGS

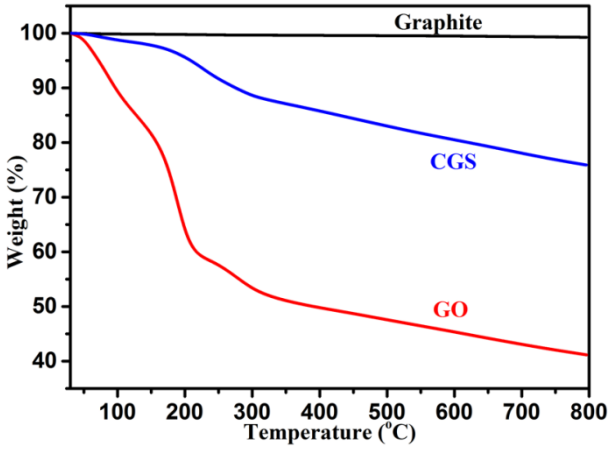

Figure S6. The TGA curves of graphite, GO and CGS (ρ=4.9 mg·cm^{-3}).

Firstly, graphite was very stable during the heating process, suggesting not any organic groups in it. Compared with that of GO, the TGA curve of CGS displays a slowly downward sloping line, which means its enhanced thermal stability due to the removal of oxygen-containing groups.

7. The influence of GO concentration on the volume of hydrogel.

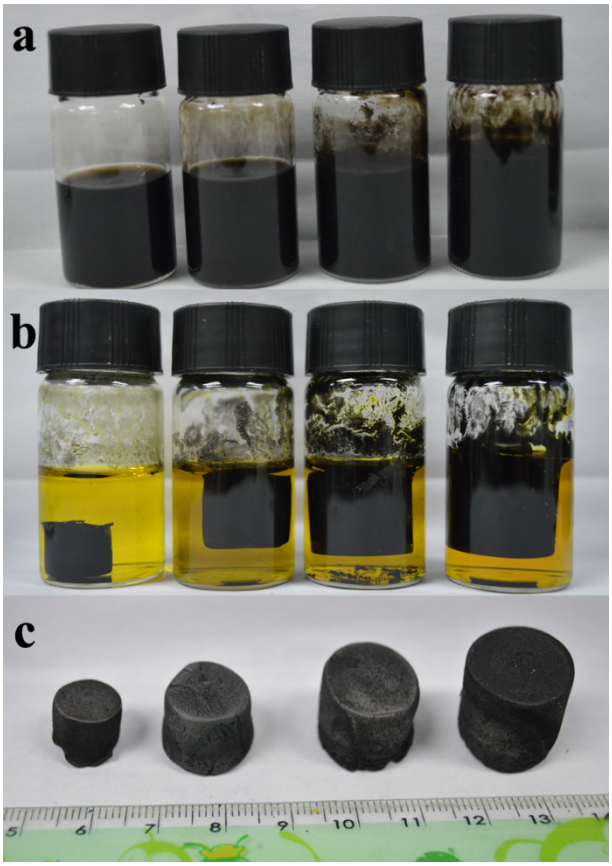

Figure S7. The influence of GO concentrations on the volume of the resultant hydrogels and sponges. From left to right the concentration of GO was 1 mg mL^{-1}, 2 mg mL^{-1}, 3 mg mL^{-1} and 5 mg mL^{-1}. The mass ratio of GO to ammonium sulfide was 1: 10 for all the reactions.

GO solutions with different concentrations including 1 mg mL^{-1}, 2 mg mL^{-1}, 3 mg mL^{-1} and 5 mg mL^{-1} were chosen. 10 mL of GO solution with different concentration was added as well as the addition of ammonium sulfide solution was 0.5 mL, 1 mL, 1.5 mL and 2.5 mL respectively (the mass ratio of GO to ammonium sulfide was 1: 10 for all the reactions). As shown in Figure S7, the mixed solution volume grows slightly for the increasing of ammonium sulfide solution. After the reduction and self-assembly process, the graphene hydrogels formed and the volume increased with the increase of concentration of GO solutions. And the resultant sponges give the same information.

8. The internal microstructure of CGSs

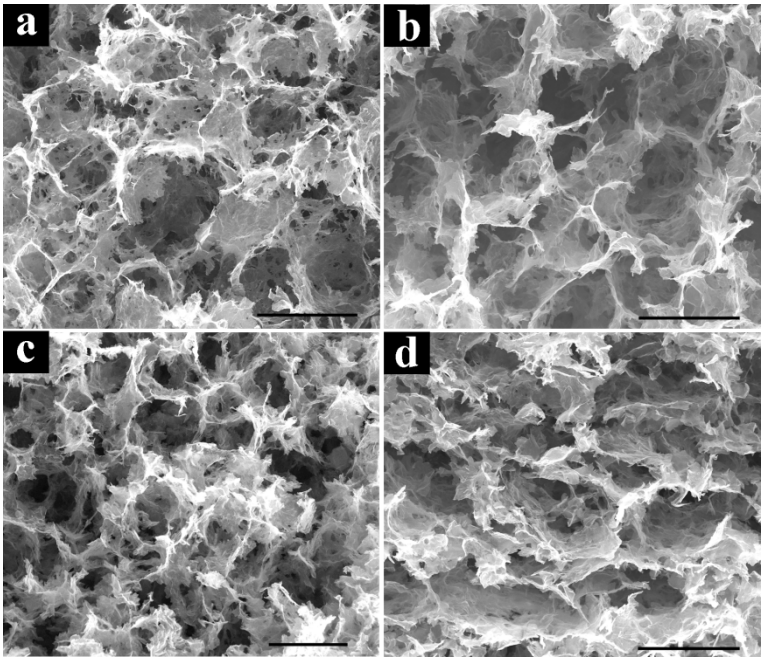

Figure S8 The internal microstructure of as-prepared graphene-only sponge. (a) CGS-1. (b) CGS-2. (c) CGS-3. (d) CGS-5. The scale bar was 100 um for all the SEM pictures.

9. Repeated compression rebound test with the strain of 50% of CGSs

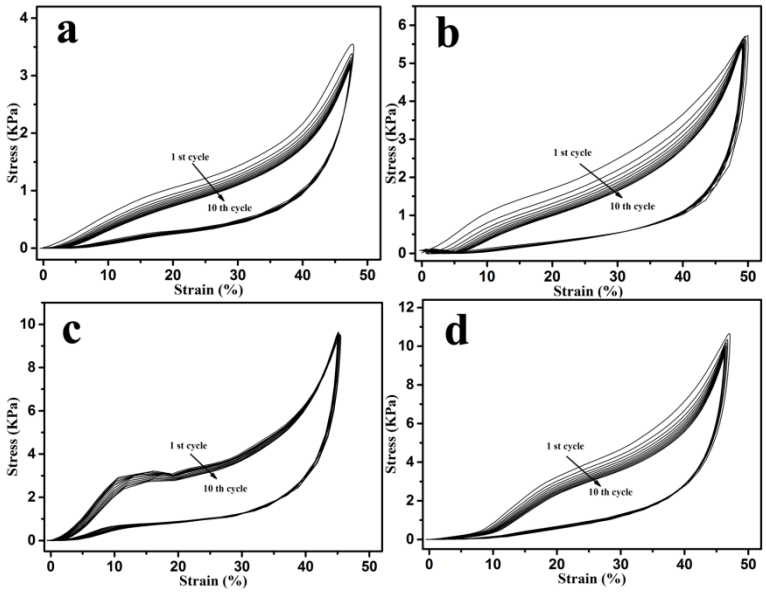

Figure S9 The stress-strain curves of CGSs with the strain of 50% for 10 cycles. (a) CGS-1. (b) CGS-2. (c) CGS-3. (d) CGS-5.

10. Repeated compression rebound test with the strain of 50% for CGSs-3.

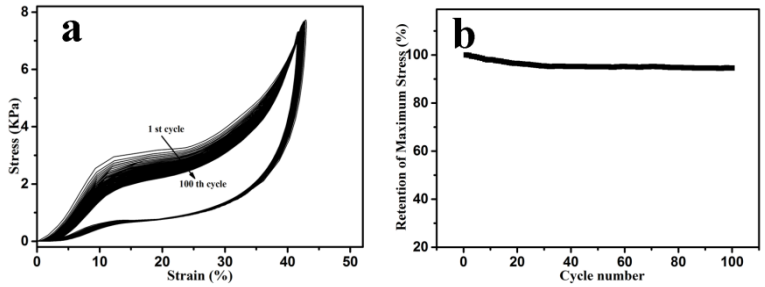

Figure S10 (a) The stress-strain curves of CGS-3 with the strain of 50% for 100 cycles. (b) Retention of maximum stress at 50% strain during the 100 compression cycles (calculated from Figure S10a).

In order to test the compression rebound more harshly, CGS-3 was compressed for 100 times. From Figure S10 we can see that there is hardly any loss for the maximum stress of the CGS-3 and the retention of maximum stress of CGS-3 was as much as 94.5% after 100 cycles. The results mean that the mechanical property is quite stabile during the compression rebound.

11. SEM pictures of CGS during the compression.

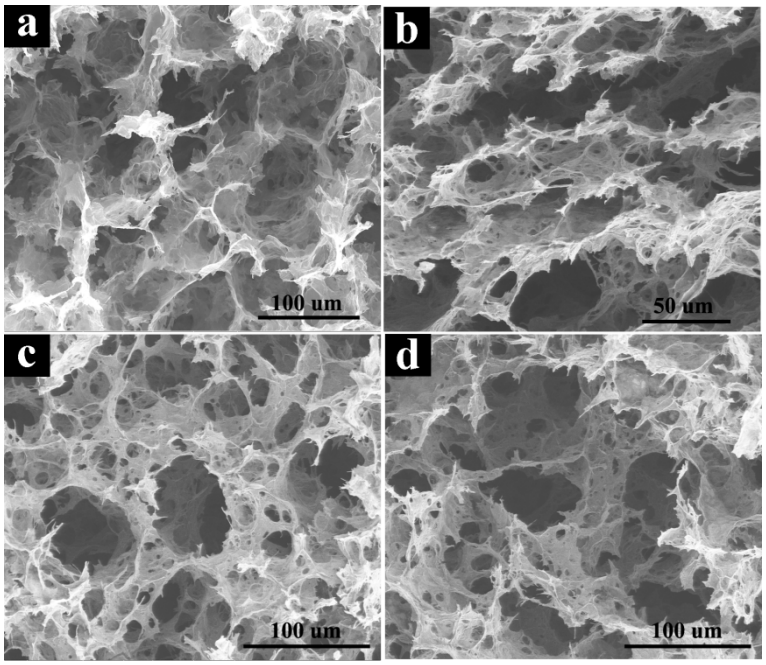

Figure S11 SEM pictures of CGS. The original CGS before compression (a), the CGS during the compression (b), CGS after 1 cycle of compression (c) and CGS-3 after 100 cycles of compression (d).

The inner structure was observed by SEM during the compression. It can be seen that before compression CGS showed round shape cells of the sponge (Figure S11a) and during the compression the round cell of the original sponge had been pressed into narrow shape while the three-dimensional structure was still remained (Figure S11b). The cells were undamaged during the compression. And after the compression the round cell structure of CGS recovered (Figure S11c). Furthermore, Figure S11d showed the inner structure of CGS-3 after 100 cycles and it can be seen that the continuous three-dimensional porous structure was still remained. This phenomenon provides that CGS possesses great stability during the cycles of compression.