Electronic Supplementary Information for:

Atomistic Mechanisms underlying Plastic Flow at UltraLow Yield Stress in Ductile Carbon Aerogels

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Description of Material Preparation

Carbon aerogel (density 0.013 g/cm³) was prepared by solvothermal reduction from graphene oxide (GO). GO was prepared based on the method adopted by Wan et al. [4]. To get the graphene aerogel, graphite flakes with an average 500 μm lateral size (Qingdao Jin Ri Lai Graphite Co., Ltd., industrial grade) in batches of 2.5 g were gradually introduced into a mixture of fuming nitric acid and sulfuric acid in a 1:3 volume ratio (100 mL). The mixture was stirred at room temperature for 24 h. Subsequently, the mixture was diluted with water, and the solid materials were separated by filtration. The resulting solid products underwent multiple rinses with water and were then dried at 60°C. Next, the dried powders were subjected to a brief heating process in an oven at 1000°C for 10 sec to render the graphite expandable. Next, 2.5 g of powder, 2.1 g K₂S₂O₈, 150 mL of sulfuric acid, and 3.1 g P₂O₅ were sequentially added to a flask and stirred at 80°C for 5 hours. After cooling to room temperature, deionized water was added to the mixture. The suspension underwent filtration and was washed with water using a 0.22 μm pore polycarbonate membrane, which was further air-dried at room temperature. After that, the solids were immersed in 50 mL concentrated H₂SO₄ at 0 °C, and 7 g KMnO₄ was added very slowly (during about 1 h) accompanied by magnetic stirring. Subsequently, the mixture was heated to 35 °C for 2 h and diluted with 1.5 L water, followed by the addition of 5 mL H₂O₂. The mixture was deposited for 2 days, after which the clear upper liquid was removed, and the precipitates were washed with water and 1 M HCl. Finally, the washed GO solution was calibrated to 2 mg mL⁻¹ for further experiments. For the preparation of graphene aerogel, 80 μL of vitamin C reducing agent with a mass percentage of 12% was gradually added to
20 mL GO solution while stirring magnetically. After 1 h, the mixed solution was transferred into a Teflon-lined stainless-steel autoclave, which was then heated in hydrothermal conditions (160 °C, 1 MPa) for 12 h in an autoclave to exclude the O-bearing groups from the carbon structure and create a graphene hydrogel. The obtained hydrogel was washed with water and subjected to -80°C pre-cooled freeze-drying to produce graphene hydrogel. The resulting hydrogel was dried naturally for four days yielding a cylinder with a 1.2 cm diameter and a 2 cm height. The height was reduced to 1.4 cm using a solid-state UV picosecond laser. The sample was tested in the Byes3000 Universal testing machine as described below. We report in Figure S1 representative SEM images of the aerogel cylinder. More details about the characterization of the material, including oxygen content, surface area, and pore size distribution, will be presented in future reports.

![SEM images of the carbon aerogel](image)

**Figure S1.** SEM images of the carbon aerogel with a mass density of 0.013 g/cm³, at two different magnifications (a,b).

**Description of Indentation Experiments**

The sample was tested in the Byes3000 Universal testing machine with a 20 N force sensor, loading at a 0.1 mm/min rate. Two glass slides were fixed on the upper and lower loading planes to ensure uniform force distribution during the compression test. We report in Figure S2 the compressive test apparatus.
**Figure S2.** The compressive test apparatus.

**Plastic flow behavior in M26 and M34 phases**

**Figure S3.** Shear simulations of the M26 phase. The parabolas of plastic yield extracted from the analysis described in the text.
Figure S4. Plots of parabolas of plastic yield extracted from the analysis of the shear simulations of: (a) the M34 phase, and (b) the M26 phase in the opposite direction of shear.

Isotangent construction

Figure S5. Microscopic energy curves corresponding to an ideal plastic behavior and the corresponding isotangent construction to determine the plastic yield stress.
TEM images

**Figure S6.** Visual comparison of: (a,c) experimental and (b,d) simulated (M34 and M18 phases, respectively) TEM images of amorphous carbonaceous phases. Experimental TEM images in (a) adapted from Figure 1 of ref. 1. Experimental TEM images in (c) adapted from Figure 6 of ref. 2. Simulated TEM images adapted from ref. 3. For reference, the length of the simulated unit cells is: (b) 14.5 nm, and (d) 10 nm, respectively.
Figure S7. Schematic strain/stress diagram obtained via compressive tests on two different samples of our carbon aerogel with a mass density of 0.013 g/cm³.
References


