Supporting Information

Experimental Section

Materials

Natural flake graphite (99.95%, 325 mesh) and sodium ascorbate ($C_6H_7O_6Na$, 99%) were purchased from Aladdin. Melamine ($C_3H_6N_6$, 99.5%) was purchased from the Tianjin Institute of Fine Chemicals. Sodium ascorbate ($C_6H_7O_6Na$, 99%) was purchased from Aladdin. Hydrogen peroxide (H_2O_2, 30%) was purchased from Tianjin Zhengcheng Chemical Products Co., Ltd. Deionized water was homemade in the laboratory. All chemicals were of analytical grade and used as received without further purification.

Characterization

The morphology and microstructure of the CGAs and NCGAs. were characterized with scanning electron microscopy (SEM, Hitachi S-4700). High-resolution X-ray photoelectron spectroscopy (XPS, ESCALAB 250) was performed to analyse the surface chemical composition and the atomic ratio of elements in CGAs and NCGAs. An X-ray diffraction (XRD, Ultima IV) system was applied to measure the XRD patterns with Cu Kα radiation and a scanning rate of 5°/min. The nanoscale pore size distribution and specific surface area of the NCGAs were measured via adsorption and desorption of N_2 (BET, ASAP-2460-4N) based on Brunauer–Emmett–Teller (BET) theory and the Barrett–Joyner–Halenda (BJH) method. The contact angles of CGAs and NCGAs were measured with 3 μL droplets of water using the contact angle measuring system (dataphysics TBU 90E) at room temperature

Density

The densities of CGAs and NCGAs were calculated by measuring the volume using a digital Vernier calliper, and their masses were measured using a balance with 0.1 mg accuracy, where the density of the air occupied in the pores was inevitably included. The density (ρ) of the graphene aerogels was calculated by Eq. (1):

$$\rho = \frac{m}{v} \tag{1}$$

Absorption capacity and reusability

CGAs or NCGAs were weighed at room temperature before the liquid was absorbed and then immersed in a beaker containing oil or organic solvents. The static adsorption process was maintained until adsorption equilibrium was reached. Then, the saturated CGAs or NCGA was separated from the adsorbate and weighed. The absorption capacity (Q) was calculated using Eq. (2):

$$Q\ (g/g)\ =\ (m - m_0)/m_0 \quad (2)$$

where Q is the absorption capacity of CGAs or NCGAs and m_0 and m are the weights of CGAs and NCGAs before and after absorption, respectively. All absorption data were obtained by repeating the absorption test three times and taking the average value.

Preparation of NCGAs

GO prepared by Hummers' method was formulated into a 2 mg/ml graphene solution, stirred, and sonicated. Graphene, sodium ascorbate and melamine were mixed into a homogeneous solution in the ratio of 1:0.5:1 by mass, and finally 80ul of 30% hydrogen peroxide was added. The mixture was sealed in a certain number of ampoules and heated in an oil bath pot at 100 °C for 2 h. After the ampoules were cooled to room temperature, the ampoule was destroyed and the obtained hydrogel was immersed in deionized water for 1 h. The washed aerogel was then freeze-dried at -80°C for 24 h to produce melamine-modified nitrogen-doped graphene aerogel. Under 99.9% purity nitrogen protection, the freeze-drying aerogels were transferred to a quartz tube furnace for carbonization treatment. Heat the quartz tube furnace to 800 °C at a heating rate of 5 °C/min and hold it at 800 °C for 120 min, waiting for natural cooling to room temperature to obtain nitrogen-doped carbon graphene aerogels.

Preparation of CGAs

The synthesis process is the same as that of NCGAs, except that melamine is not introduced.

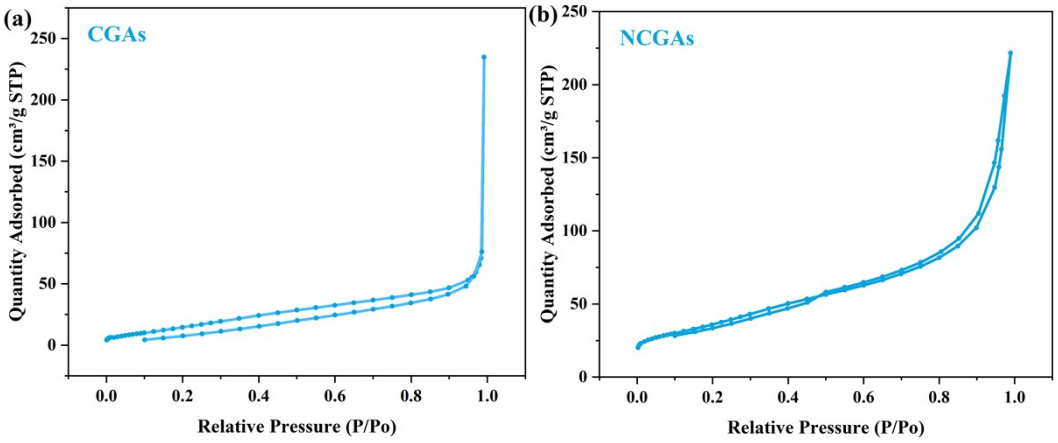

Fig. S1 (a), (b) N2 sorption isotherm of CGAs and NCGAs

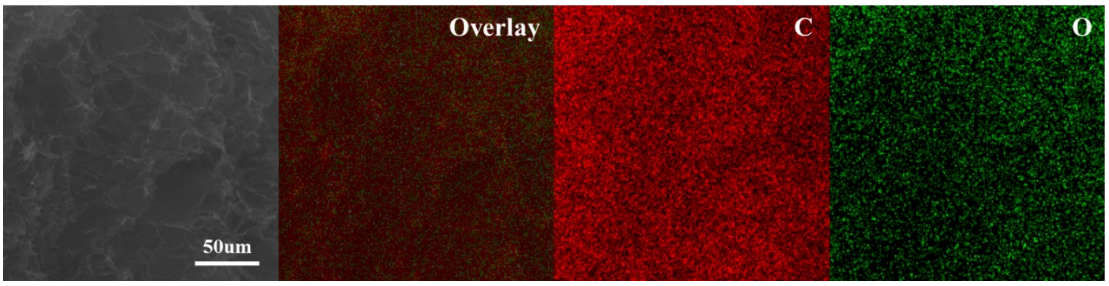

Fig. S2 SEM image and the corresponding elemental mapping images of overlay, C and O for CGAs.

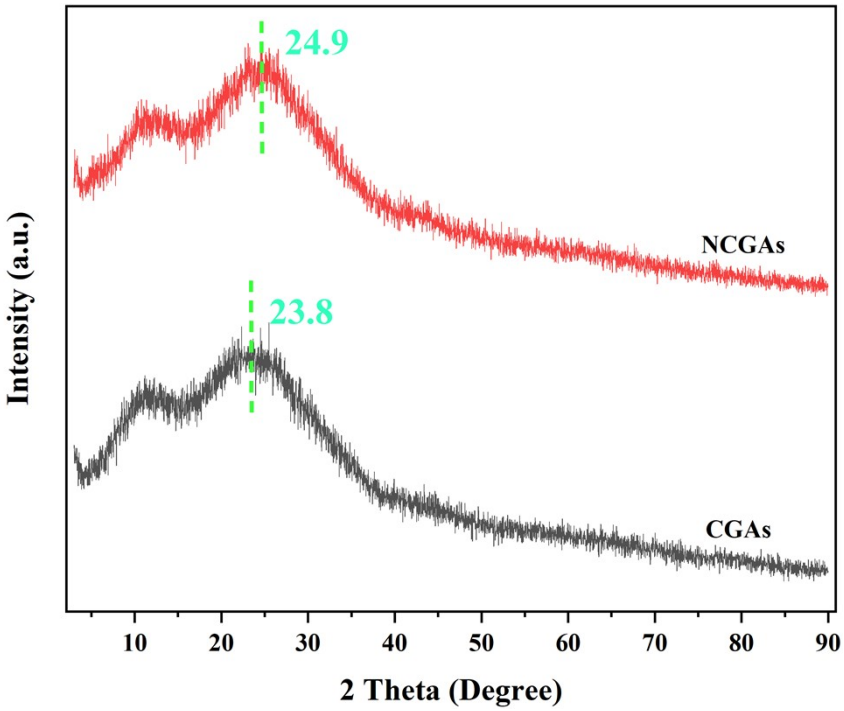

Fig. S3 XRD spectra of CGAs and NCGAs

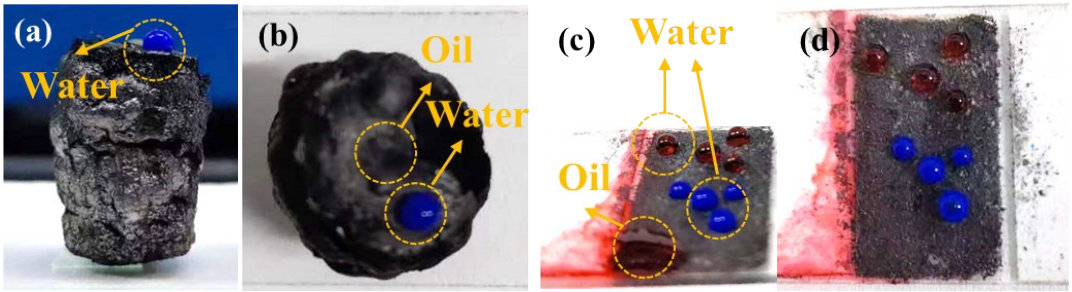

Fig. S4 (a) Water droplets remain on the NCGAs surface (Water is dyed with blue ink). (b) Water droplets remain on the NCGAs surface and oil droplets are quickly absorbed. (c) Water drops and oil on an adhesive surface covered with aerogel powder (Water is dyed with blue ink or Congo red, oil is dyed red by Sudan III)

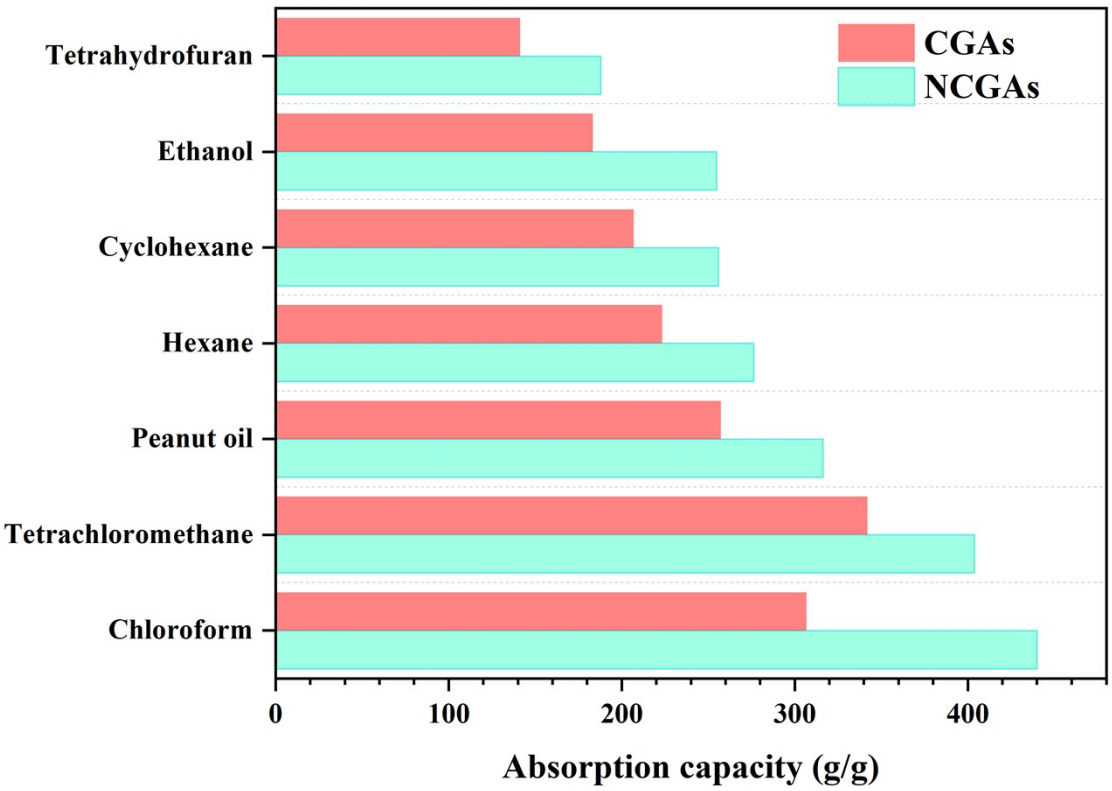

Fig. S5 The absorption capacity of CGAs and NCGAs for various oils and solvents.

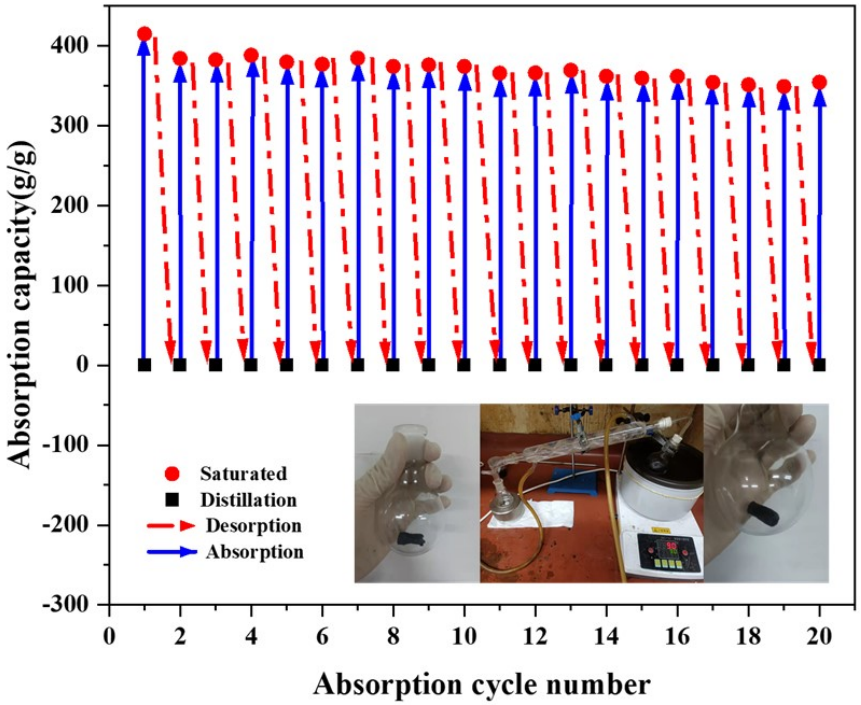

Fig. S6 Chloroform absorption- Distillation and recoverability testing of NCGAs

Table S1 . Comparison of Various Graphene Aerogels

Absorbent material	Maximum absorption capacity (g/g)	Lowest density (mg/cm^3)	Ref.
Graphene/cysteamine aerogel	310	4.2	[1]
Graphene/cellulose aerogel	197	5.9	[2]
Anisotropic graphene aerogels	200	4	[3]
Spongy Graphene	86	12	[4]
Carbon aerogels derived from sisal fibers	188		[5]
Graphene/nanofibrillated cellulose aerogel	265	5.6	[6]
Graphene/polyvinyl alcohol/cellulose nanofiber aerogel	287	6.17	[7]
Carbon nanotubes/graphene hybrid aerogel	83	6.2	[8]
Graphene Aerogel Millispheres	195	5.0	[9]
Graphene/ethylenediamine aerogel	250	4.4	[10]
This work	440	3.5	

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