Electronic Supplementary Information (ESI)

Highly Adsorptive Graphene Aerogel Microsphere with Center-diverging Microchannels Structure

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1. Slush bath[1]

The slush bath was prepared by slowly pouring liquid nitrogen into ethyl acetate with stirring in a Dewar bottle. The slush bath can be kept for a few hours at low temperature（-85 ºC to -45 ºC for 2.5h）.

2. Electrospraying device

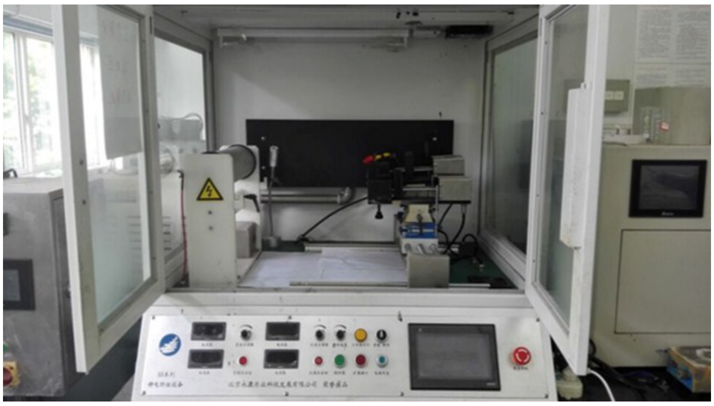

Figure S1. The picture of Electrospraying device (Beijing Yongkang Co. Ltd, China)

3. Optimization of electrspraying parameters

The appropriate voltage and distance between the tip of the needle to the surface of the target liquid in the container are important for fabricating desirable microspheres. Within a constant distance, as low voltage was applied, the liquid dripped from the tip of needle to form huge and nonspherical particles (Figure S2a) because electric force is not enough for the split-up of droplets. In contrast, too strong voltage could cause an unstable "*Taloy cone*" jet[2], resulting in non-uniform particles (Figure S2 c). Only when the moderately high voltage was applied, the stable "*Taloy cone*" can be obtained to prepare the GOAMs with desirable morphology (Figure S2 b-e). On the other hand, the distance between electrodes also plays an important role in controlling the morphology of the microspheres. When the distance is not long enough for the fully splitting of charged droplet, big microspheres along with small microspheres were formed (Figure S2 d) no matter how high the voltage is. When the distance is too long, the droplets have increasing kinetic energy to hit the cold liquid surface, which causes the deterioration of sphericity (Figure S2 f). In our experiments the optimized electrospraying conditions are that the distance is 10 cm with an applied voltage of 8 kV and the voltage should increase slightly as the flow rate increases.

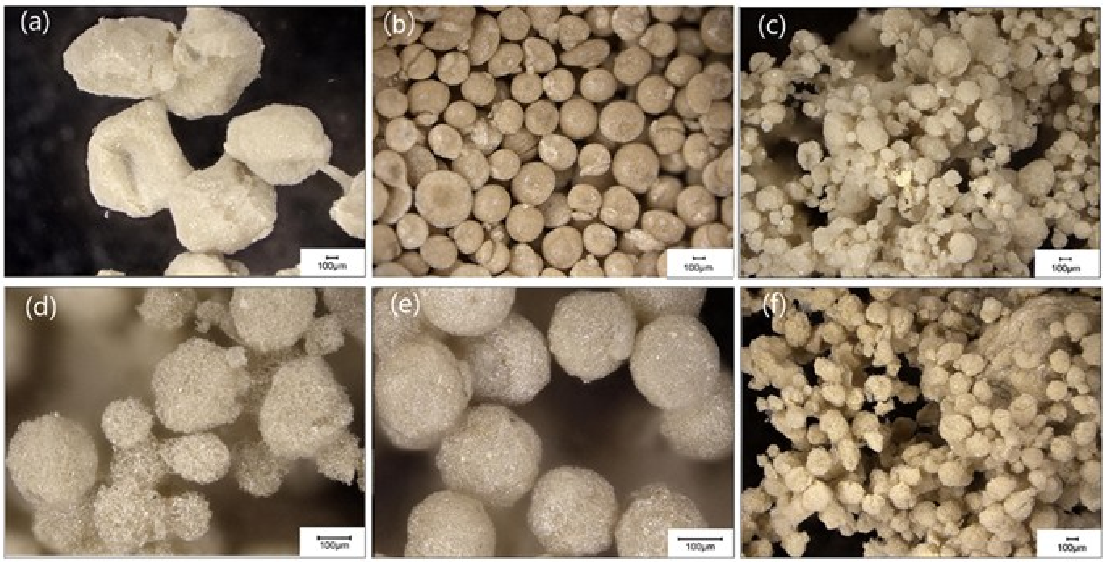

Figure S2. Optical microscope images of GOAMs fabricated in different electrospraying conditions at a flow rate of GO dispersion (7.5 ml/h) and GO concentration (6 mg/ml): (a) Distance: 10 cm, Applied voltage:6.5 kV; (c) Distance:10 cm, Applied voltage:10 kV; (b) and (e) Distance: 10 cm, Applied voltage: 8 kV; (d): Distance: 6 cm, Applied voltage: 8 kV; (f) Distance: 14 cm, Applied voltage: 8 kV.

4. Size-controllable fabrication of GOAMs

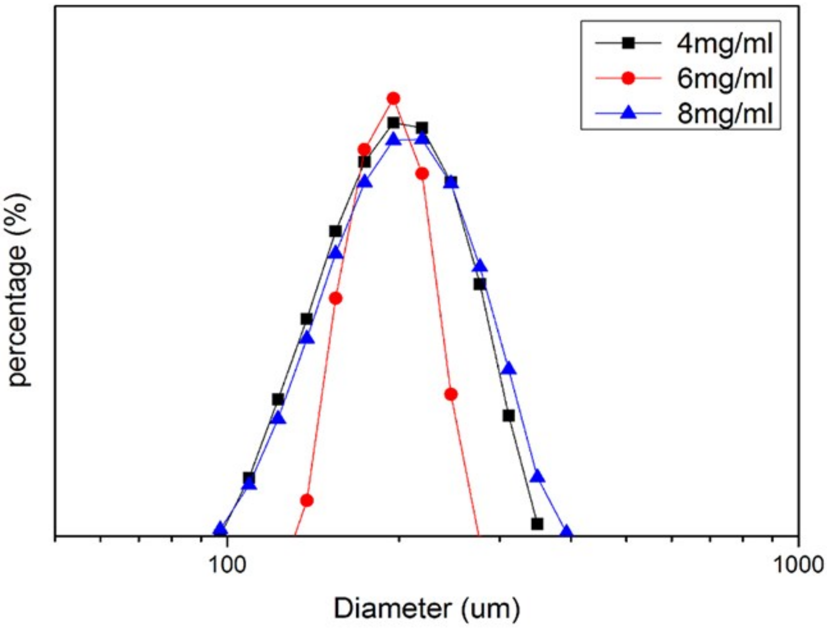

Figure S3. Grading analysis of GOAMs made from various GO concentrations (Distance between electrodes: 10 cm; Applied voltage: 8 kV, Flow rate of GO dispersion: 3.75 ml/h

5. Typical optical pictures of Dandelion-like GOAMs

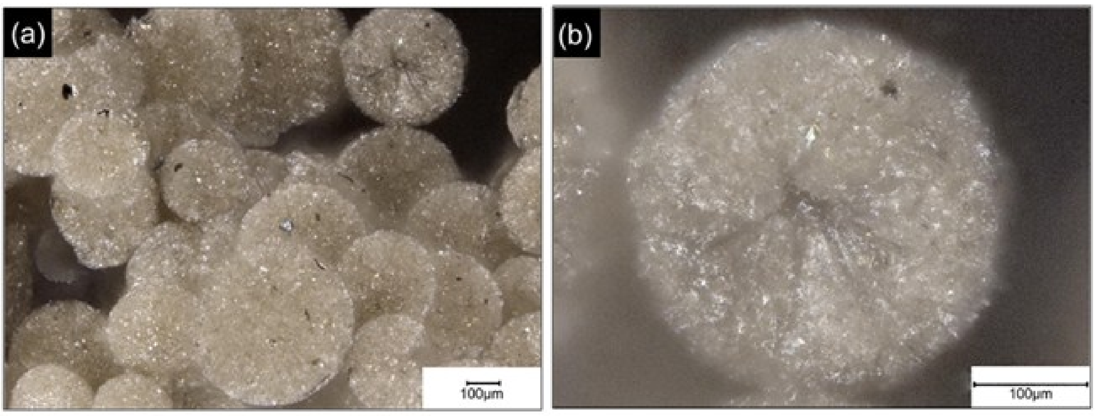

Figure S4. The optical pictures of GOAMs

6. Characterization of rGOAMs

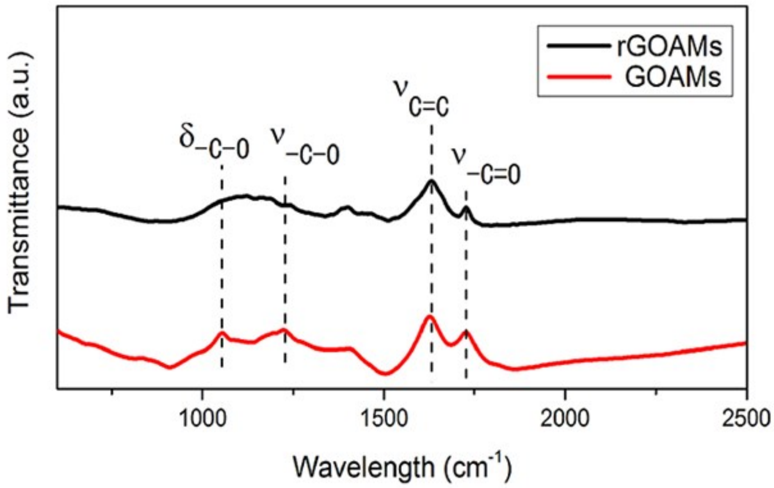

Figure S5. FTIR of GOAMs and rGOAMs

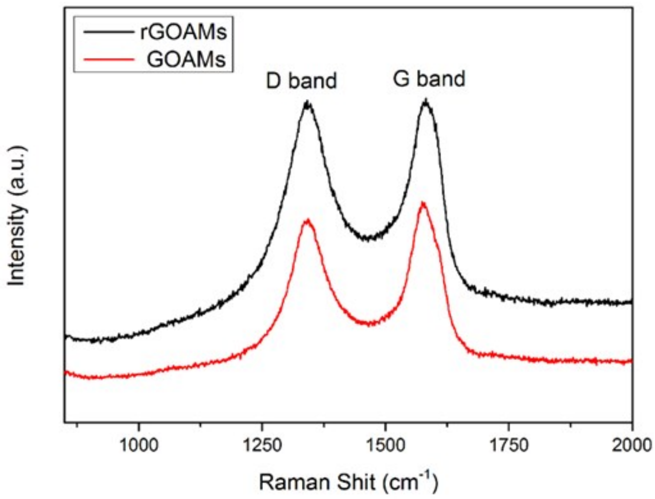

Figure S6. Raman spectra of GOAMs and rGOAMs

7. Absorption capacities as a function of densities (ρ) of organic liquids.

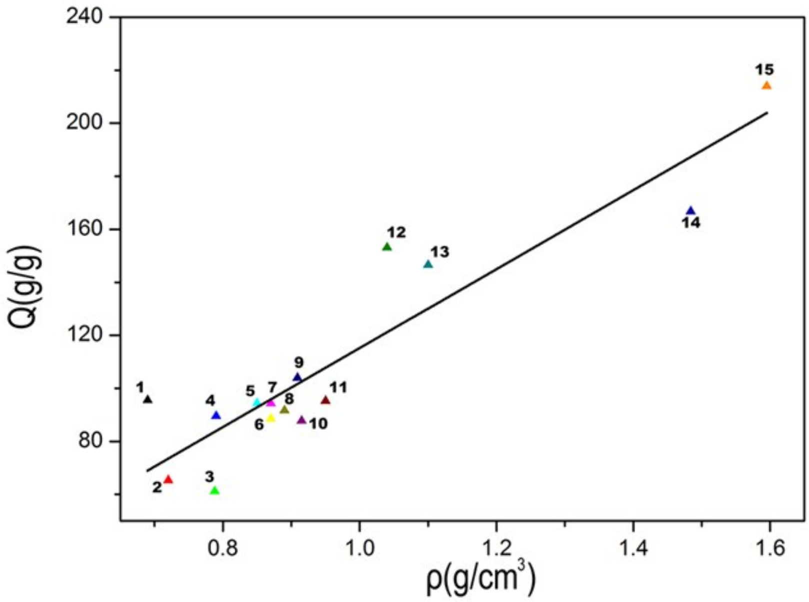

Figure S7. Absorption capacities as a function of densities of organic liquids, The numbers (1-15) represent：n-Hexane, Gasoline, Acetone, Ethanol, Diesel oil, Vegetable oil, Toluene, THF, DMF, 1,4-dioxane, Lubricating oil, Pump oil, DMSO, Chloroform, and Phenixin, respectively.

8. Comparison of various absorbents.

Table S1. Comparison of various absorbents

Absorbents	Absorbates	Adsoption capacity($g \cdot g^{-1}$)	Ref.
1. Graphene coated sponge	Oils and organic solvents	50-160	3
2. Spongy graphene	Oils and organic solvents	20-86	4
3. Graphene sponge	Oils and organic solvents	60-160	5
4. Soot treated spongy graphene	Oils and organic solvents	140-616	6
5. RGO foam	Cyclohexane,chlorobenzene, toluene, petroleum, motor oil	5-40	7
6. RGO foam	Oils and organic solvents	70-125	8
7. Graphene/polypyrrole foam	Diesel, kerosene, etc	36.8-109	9
8. Graphene/CNTs foam	Compressor oil, sesame oil, toluene, DMF chloroform, dichlorobenzene	80-130	10
9. Giant graphene/CNTs aerogel	Oils and organic solvents	215-743	11
10. Graphene/CNTs aerogel	Oils	110-140	12
11. Graphene/α-FeOOH aerogel	Cyclohexane,toluene,gasoline, paraffin oil, vegetable oil, phenoxin	12.5-26	13
12.Graphene/Cu_2O aerogel	Oils	28-40	14
13. Graphene/ PVDF aerogels	Oils and organic solvents	20-70	15
14. Graphene aerogel （ethylenediamine reduction）	Oils and organic solvents	120-250	16
15. Graphene aerogel （L-phenylalanine reduction）	Oils and organic solvents	100-260	17
16. Graphene/resol aerogel	Organic solvents	200-400	18
17. Ni-doped graphene/carbon Cryogel	Motor oil, vegetable oil	22.2-23.2	19
18. N-Doped Graphene Framework	Oils and organic solvents	200-600	20
19. r-EPGM (Reduced expanded porous graphene macroform)	Oils	20-70	21
20. rGOAMs（Reduced graphene oxide aerogel microspheres）	Oils and organic solvents	60-214	Our work

9. Control sample: unidirectional freezing-drying rGO monolith foam

The GO monolith foam was fabricated by unidirectional freeze-drying as described in reference[22] and was reduced to rGO monolith foam by the same thermal process as rGOAMs mentioned above.

Figure S8. Photo of rGO monolith foam

References

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