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Electronic Supplementary Information

Flyweight and Superelastic Graphene Aerogel as High-capacity Adsorbent and Highly Sensitive Pressure Sensor

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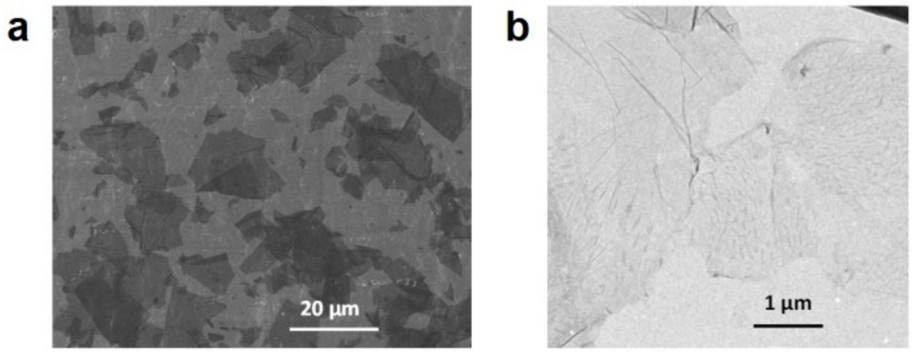

Fig. S1 SEM (a) and TEM (b) images of GO sheets with a lateral size of ~ 20 μm.

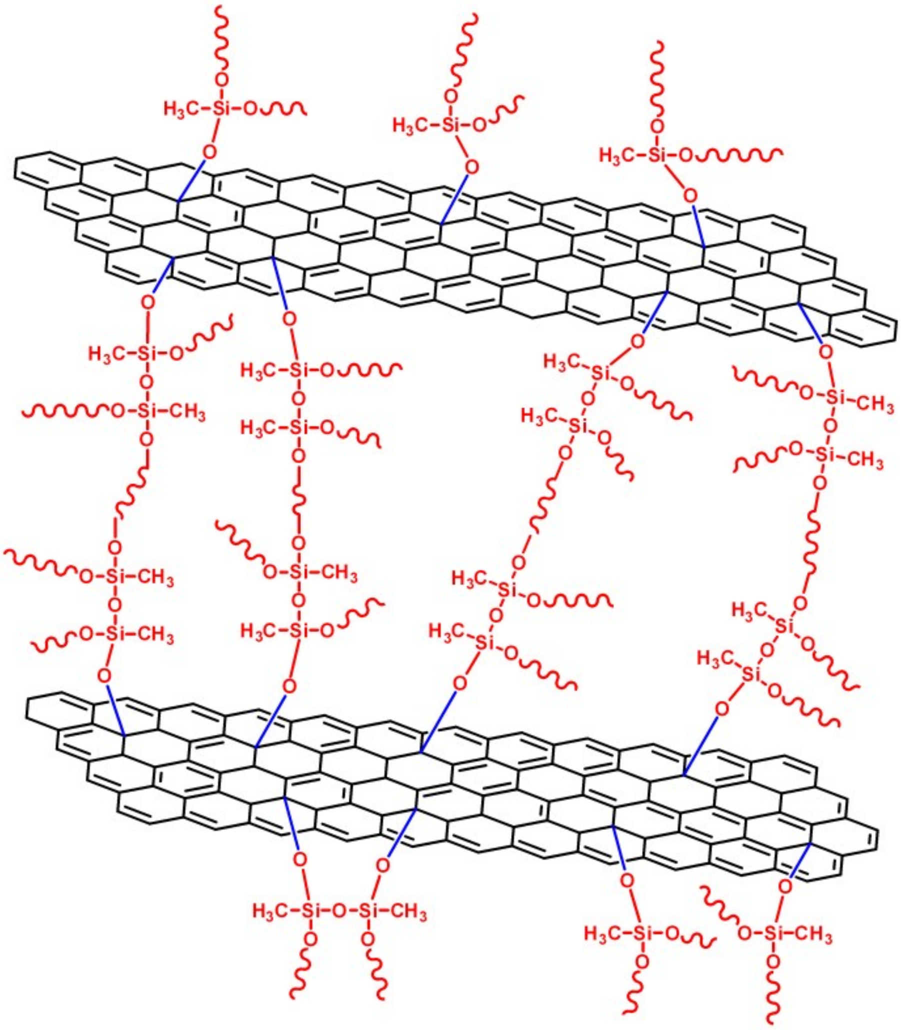

Fig. S2 Schematic of reaction between MTES and GO.

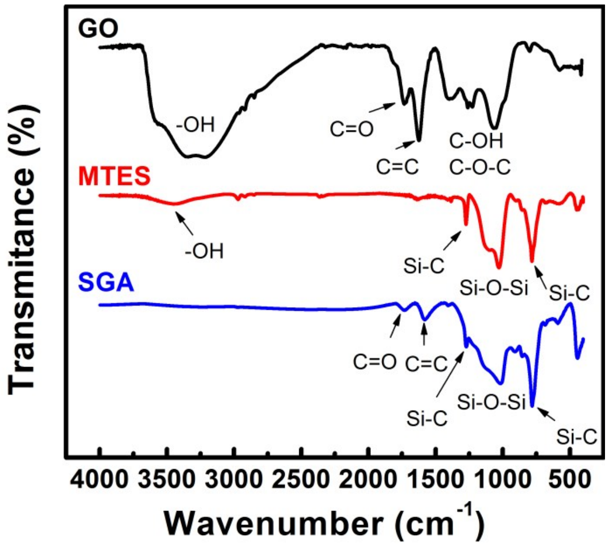

Fig. S3 FTIR spectra of GO, MTES and SGA. The absorption peaks of C=O (carbonyl) at 1732 cm^{-1}, O-H at 3210 cm^{-1} and 1363 cm^{-1}, C-O (epoxy) at 1263 cm^{-1}, and C-O (alkoxy) at 1050 cm^{-1} in GO [1] decrease dramatically in SGA, suggesting that most oxygen groups are reacted or removed. For MTES, the broad band at 3450 cm^{-1} due to O-H stretching of hydrogen bonded H_2O and surface Si-OH [2] almost disappears in SGA, indicating the reaction of Si-OH groups during CVD. Some typical bands like Si-CH_3 (783 cm^{-1} and 1270 cm^{-1}) [2] and Si-O-Si (1000-1150 cm^{-1}) [2] are observed in SGA, confirming the successful modification.

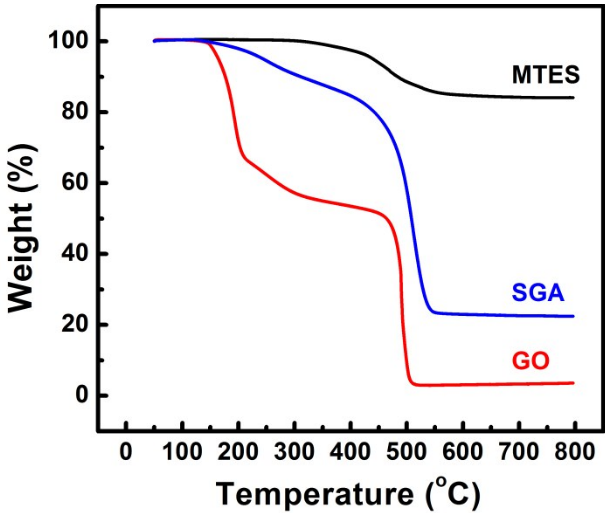

Fig. S4 TGA curves of MTES, GO and SGA in air. MTES shows about 16 % weight loss from 300 °C to 600 °C due to decomposition of pendant groups (–OH and $–CH_3$) while it can retain its siloxane backbone up to 800 °C [3]. GO exhibits about 35 % weight loss at 200 °C due to removal of labile oxygen-containing functional groups [4] and another ~ 20 % weight loss above 450 °C due to decomposition of carbon backbone [5]. SGA decomposes residual functional groups on GO (~10 % weight loss) at 200 °C and $–CH_3$ introduced by MTES and carbon backbone of GO from 400 °C to 550 °C. According to the residue weight (25 %), MTES of~30 % is introduced in SGA.

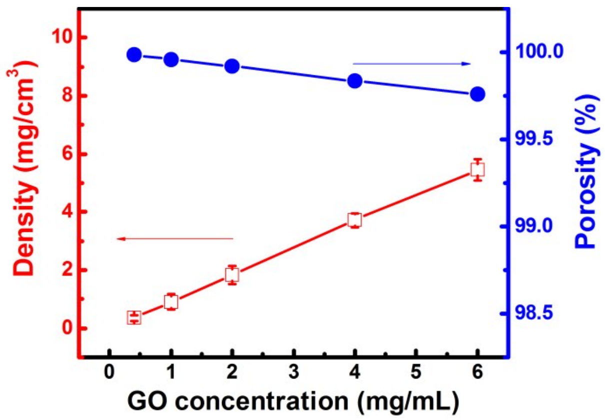

Fig. S5 Density and porosity of SGA as a function of initial concentration of GO in the suspensions.

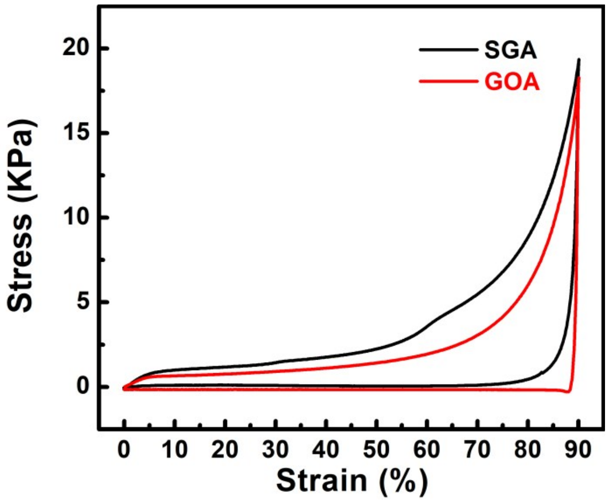

Fig. S6 Compressive stress-strain cycles of SGA (ρ = 3.7 mg/cm^3) and GOA (ρ = 3.8 mg/cm^3) with a maximum strain of 90 %.

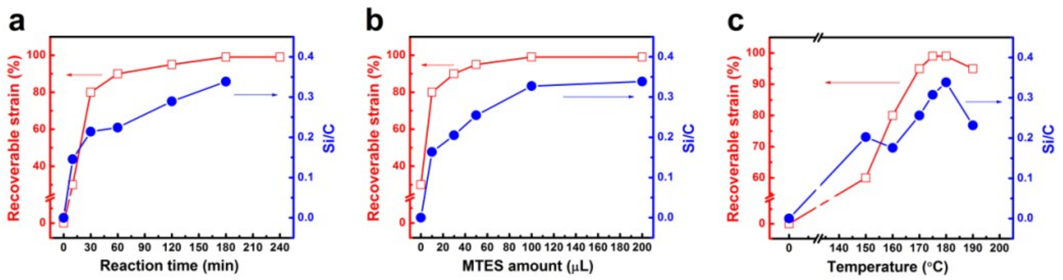

Fig. S7 Recoverable strain and Si/C atom ratio of SGA with various reaction time (a, 100 μL MTES and 180 °C), MTES amount (b, 180 °C and 3 h) and temperature (c, 100 μL MTES and 3 h). It takes 30 min to endow SGA with 80 % recoverable strain; after reaction time of 3 h, over 99 % recoverable strain is obtained. Correspondingly, the Si/C ratio increases to about 0.34. The dependence of elasticity on MTES amount shows a similar trend; the recoverable strain and Si/C ratio stabilize at 99.5 % and 0.34, respectively, at 100 μL MTES. Note that even without MTES, SGA treated at 180 °C for 3 h exhibits 30 % recoverable strain because the thermal reduction enhances the intrinsic elasticity of graphene network. Considering the boiling point of MTES (140 °C), the lowest reaction temperature was set as 150 °C. At reaction temperature 180 °C, both the recoverable strain and Si/C maximize. Higher reaction temperature will remove too much functional groups on GO, which is adverse to the grafting of MTES and lowers Si/C ratio and elasticity.

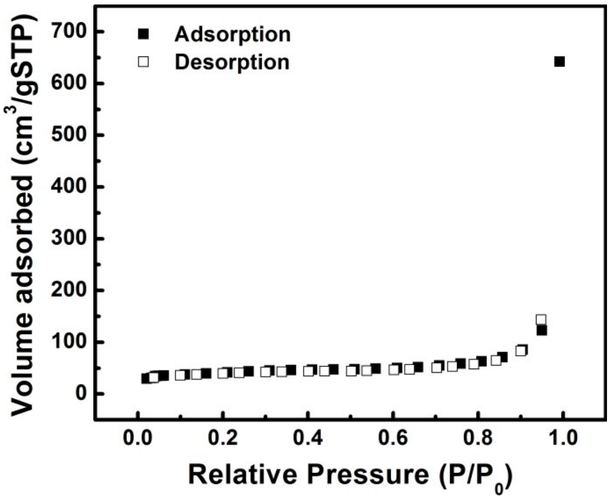

Fig. S8 Nitrogen adsorption/desorption isotherms of SGA.

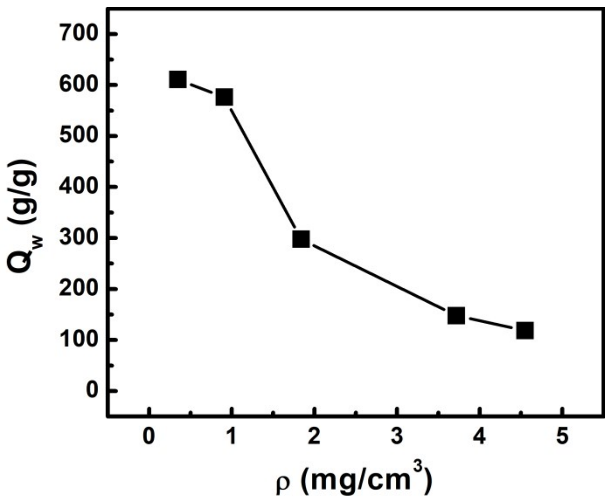

Fig. S9 Q_w of SGA for toluene as a function of ρ.

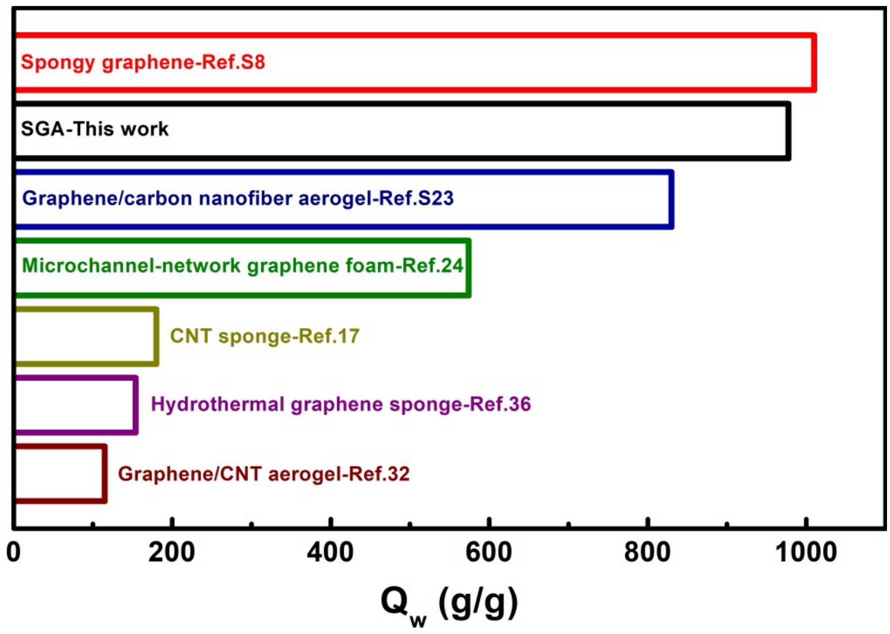

Fig. S10 A diagram of Q_w of various absorbents for chloroform.

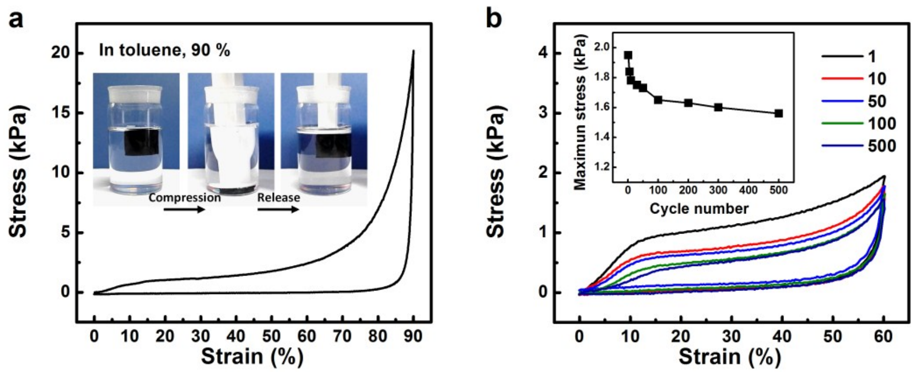

Fig. S11 a) Compressive stress-strain cycles of SGA (ρ = 3.72 mg/cm^3) in toluene at 90 % maximum strain. Insert: compression-recovery process of SGA in toluene. b) Cyclic stress-strain curves of SGA in toluene at 60 % maximum strain. Insert: maximum stress for 500 compression cycles.

Table S1 The maximum recoverable strain of carbon-based 3D materials.

Materials	**Density (mg/cm^3)**	**Maximum strain (%)**	**Ref.**
SGA	0.35-5.45	99.5	This work
Naturally dried graphene aerogels	6.8, 6.9, 7.1, 7.6, 9.3	99	S6
Air-bubbled graphene foam	25.45	99	S7
Spongy graphene	~1.07	98	S8
Tubular graphene	3.3	95	S9
Graphene/iron oxide aerogel	5.8	95	S10
Ultralight graphene aerogel	~3	90	S11
Carbon–graphene monolith	8.5	90	S12
Graphene-coated CNT aerogel	14	90	S13
PDMS/graphene aerogel	~17.6	90	S14
Ultra-flyweight carbon aerogel	1.5	85	S15
Graphene–CNT aerogel	2.52	80	S16
CNT sponge	5-10	80	S17
Carbonaceous nanofibers aerogel	21.3	80	S18
RF–GO–metal aerogel	31.2	80	S19
PVA/graphene aerogel	15	70	S20
N-doped graphene	2.32	50	S21
EDA reduced graphene aerogel	4.4, 6.0, 6.9	50	S22

Table S2 Adsorption capacities of carbon-based 3D materials for oils and organic solvents

Materials	**Q_w (g/g)**	**Ref.**
SGA	407-1035	This work
Spongy graphene	508-1010	S8
Graphene/carbon nanofiber aerogel	393-1002	S23
Ultra-flyweight carbon aerogel	215-913	S15
Microchannel-network graphene foam	137-760	S24
High-temperature reduced graphene sponge	110-616	S25
	240-500	S26
Nitrogen-doped graphene aerogel	40-156	S27
	200-600	S28
Graphene sheet-nanoribbon hybrid aerogel	100-350	S29
Graphene foam from CVD	200-250	S30
	80-130	S31
Graphene/CNT aerogel	110-140	S32
	110-270	S33
Thermally treated reduced graphene aerogel	140-270	S34
Ethylenediamine reduced graphene aerogel	120-250	S22
Ascorbic acid reduced graphene aerogel	120-200	S35
Hydrothermal graphene sponge	70-154	S36
Thermally reduced graphene foam	70-125	S37
Phenolic acids reduced monolithic graphene	25-60	S38
Tannic acid reduced graphene aerogel	15-30	S39
Polymer-grafted graphene foam	40-196	S40
Graphene/poly(vinyl alcohol) aerogel	67-155	S20
Graphene modified melamine foam	60-140	S41
Zirconia-reinforced graphene foam	60-130	S42
Fluorinated graphene aerogel	34-112	S43
Graphene/polypyrrole foams	37-109	S44
Graphene/polyvinylidene fluoride aerogel	20-70	S45
Graphene/Cu aerogel	28-40	S46
Graphene coated polyurethane sponge	35-39	S47
Expanded graphite	60	S48
CNT sponge	80-180	S17
CNT modified polyurethane sponge	22-34.9	S49

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