Crystallization behaviors of silica improved by residual hydrogen bonding interactions under high temperature

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Supporting Information

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Experimental section

Preparation of honeycomb graphene oxide (GO)

GO was prepared by the modification of Hummers's method from flake graphite (average particle diameter of 4 μm, 99.95% purity, Qingdao Tianhe Graphite Co. Ltd., Qingdao, China). 5 g of graphite and 3.75 g of $NaNO_3$ (A.R.) were placed in a flask. Then, 375 mL of H_2SO_4(A.R.) was added with stirring in an ice-water bath, and 22.5 g of $KMnO_4$(A.R.) were slowly added over about 1 h. Stirring was continued for 2h in the ice-water bath. After the mixture was stirred vigorously for 5 days at room temperature, 700 mL of 5 wt % H_2SO_4 aqueous solution was added over 1 h with stirring, and the temperature was kept at 98 °C. The resultant mixture was further stirred for 2h at 98 °C. The temperature was reduced to 60 °C, 15 mL of H_2O_2 (30 wt % aqueous solution) was added, and the mixture was stirred for 2 h at room temperature. To remove the ions of oxidant and other inorganic impurity, the resultant mixture was purified by repeating the following procedure cycle 2 times: centrifugation, removal of the supernatant liquid, addition of 2L of a mixed aqueous solution of 3wt% H_2SO_4/0.5wt% H_2O_2 to the bottom solid, and dispersing the solid using vigorous stirring and bath ultra-sonication for 30 min at a power of 140 W. Then a similar procedure was repeated: two times using 3wt% HCl aqueous solution (2L) and one time

using H_2O (2L). The final resultant water solution was dialyzed for two weeks to further remove the remaining HCl acid and other impurity. After centrifugation, water in the resultant solide was removed by freeze drying for 48 h.

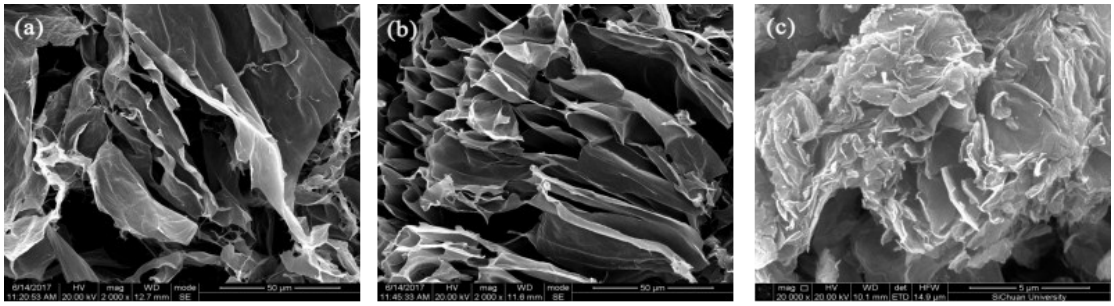

Figure S1. SEM micrographs of graphene and its derivatives: G (a), GO (b) and HFG(c)

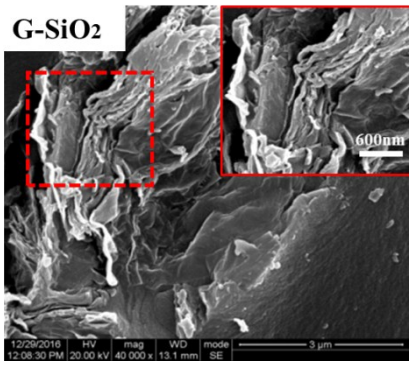

Fig.S2. SEM images of G-SiO_2

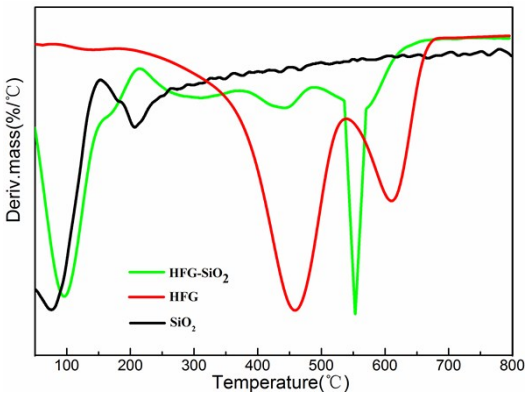

Figure S3. TGA lines of SiO_2, HFG and HFG-SiO_2

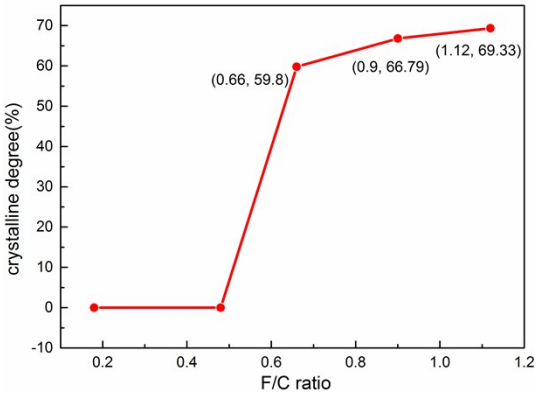

Figure S4. Effect of fluorine content of fluorinated graphene on crystalline morphology of calcined products of composites in fluorinated graphene and silica

Table S1 .Crystallization behaviors of sintered silica

contaminants	Temperature(℃)	products
LiCl	650-700	Quartz
NaCl/NaBr	800	Cristobalite
KCl/KBr	850	Cristobalite
Li_2O	1000	Cristobalite
Na_2O/K_2O	700	Cristobalite
Rb_2O/Cs_2O	900	Cristobalite
Polyimide	800	Cristobalite
polyvinyl alcohol	900	Quartz+Cristobalite
Highly fluorinated graphene	**900**	**Cristobalite**

Table S2. Thermal properties of HFG and crystalline degree of silica-HFG at nitrogen and air atmosphere

atmosphere	Thermal properties of HFG			Crystalline degree of silica-HFG(%)
	$T_{5\%}$	$T_{10\%}$	T_{max}	
nitrogen	238.2	420.5	466.9	78.6
air	227.6	393.7	457	59.8

Table S3. Chemical composition of graphene and its derivatives measured by XPS

samples	C content (%)	F content (%)	O content (%)	The ratio of F/C	The ratio of O/C
G	83.55	0	16.45	0	0.2
LFG	72.38	13.52	14.09	0.18	0.19
MFG	59.04	28.78	12.17	0.48	0.21
HFG	53.74	35.54	10.72	0.66	0.20
HFG1	45.27	40.74	13.99	0.90	031
HFG2	43.89	49.11	7.00	1.12	0.16
GO	75.4	0	24.6	0	0.33