Supplementary Information

Controlled Assembly of SiO_x Nanoparticles in Graphene

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Synthesis of SiO_x nanoparticles. The growth of SiO_x nanoparticles is precisely modulated by tune of the temperature and time. In our experiments, after the etching process, the temperature was shifted to 1200 ^{o}C for the following SiO_x growth. During the formation process of SiO_x structures, the hydrogen gas wat shut off while still keeping the flow of argon gas. The evolution of the SiO_x structures has been detected for different time, such as 30 min, 60 min and 90 min and so on. The detailed synthesis conditions are listed in Table S1.

Table S1. CVD condition of the whole process for the assembly of SiO_x patterns in graphene.

Type	Stage	Temperature (^{o}C)	CH_4 (sccm)	Ar (sccm)	H_2 (sccm)	Time (min)
Fractal	Graphene Growth	1150	0.7	1500	20	80
	Graphene Etch	1150	0	1500	20	30
	SiO_x Deposition	1200	0	1500	0	60
Petal-like	Graphene Growth	1150	0.7	1500	20	80
	Graphene Etch	1150	0	1500	40	30
	SiO_x Deposition	1200	0	1500	0	60
Hexagonal	Graphene Growth	1150	0.7	1500	20	80
	Graphene Etch	1150	0	1500	60	30
	SiO_x Deposition	1200	0	1500	0	60

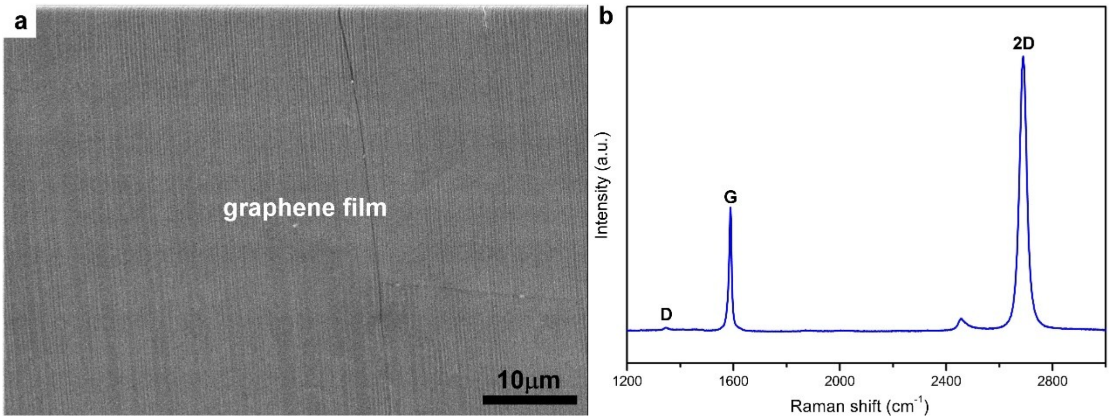

Figure S1. (a) SEM images of as-grown uniform graphene film on the liquid Cu surface. (b) The typical Raman spectrum of the graphene film, suggesting the single-layer features.

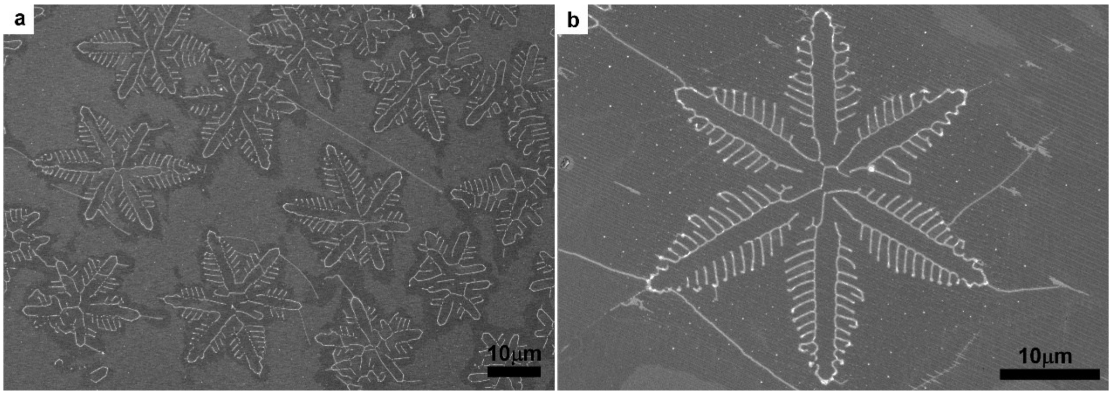

Figure S2. The SEM images of etched fractal graphene grown on liquid Cu surface under the CVD condition as listed in Table S1.

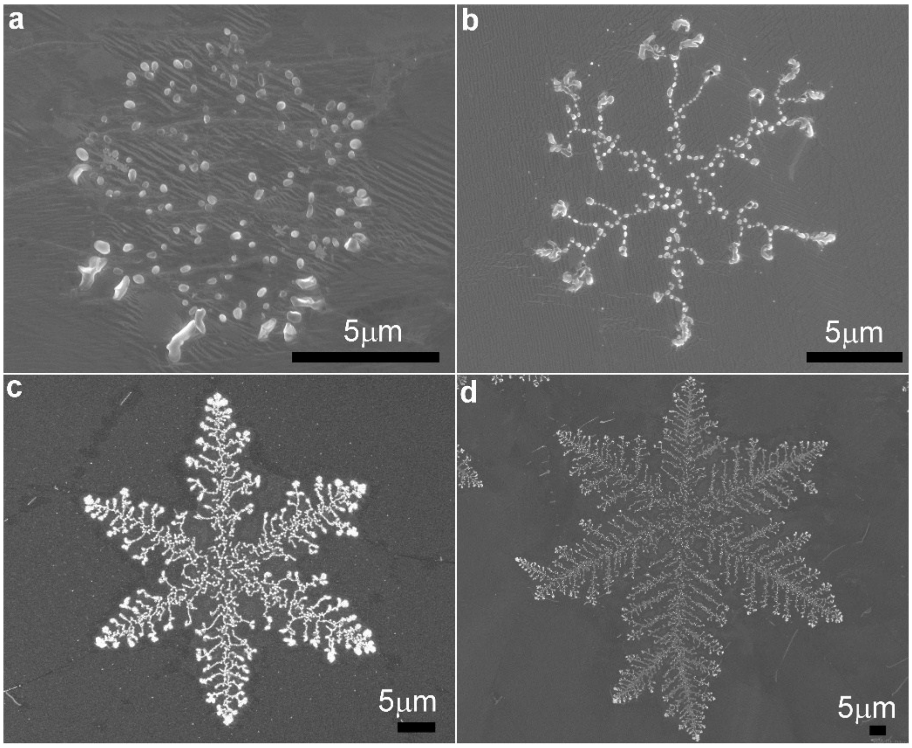

Figure S3. Time-dependent shape evolution of fractal SiO_x. (a-d) SEM image of the shape evolution of fractal SiO_x as time increased. The time are 10 min, 30 min, 60 min and 90 min, respectively.

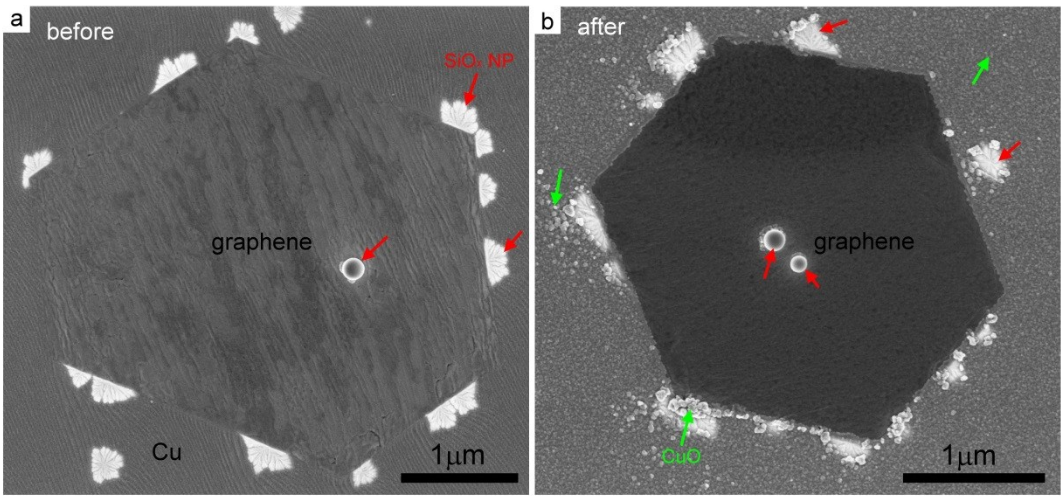

Figure S4. SEM images of SiO_x nanoparticles in graphene (a) just obtained and (b) placed in air for one week from a. Note that no visible structural change was observed while the neighboring Cu substrates were covered with cooper oxide, indicating their high thermal and chemical stability.

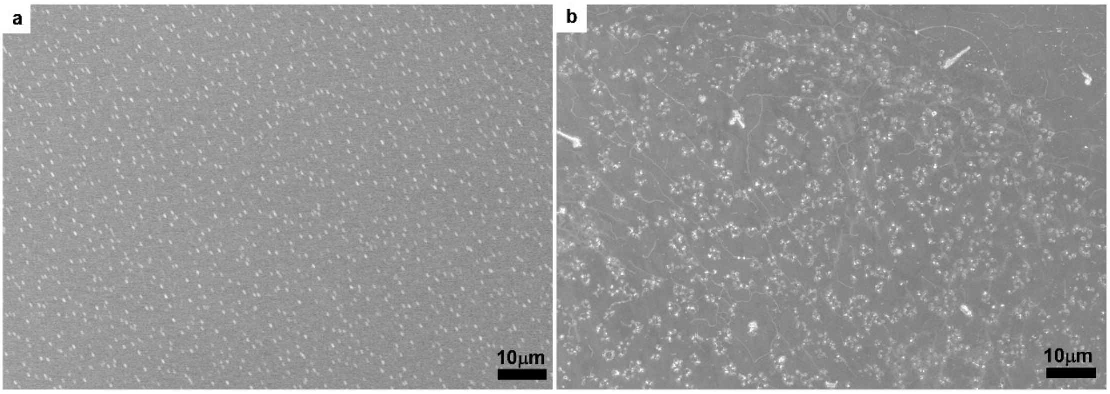

Figure S5. The non-regular shaped SiO_x patterns deposited on liquid Cu surface and graphene film without etching, respectively.

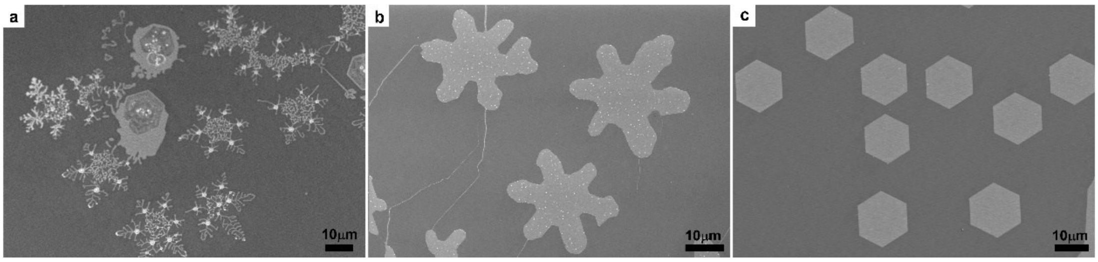

Figure S6. The SEM images of large-area three kind graphene etching patterns, in which the white areas indicate the complete etching of graphene and thus the holes are left.

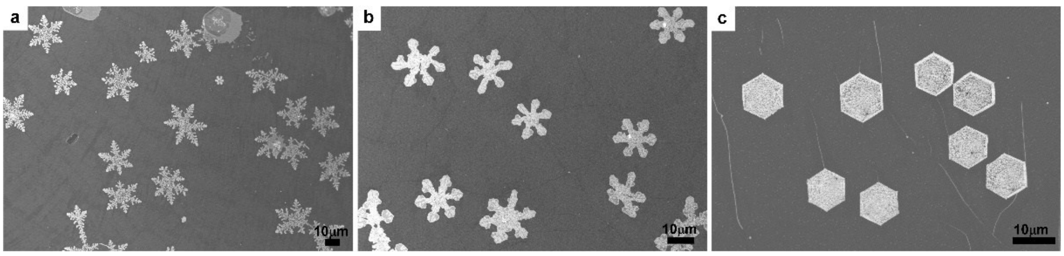

Figure S7. The SEM images of large-area three typical kinds as-assembly SiO_x patterns on the corresponding graphene etched holes as shown in Figure S6.

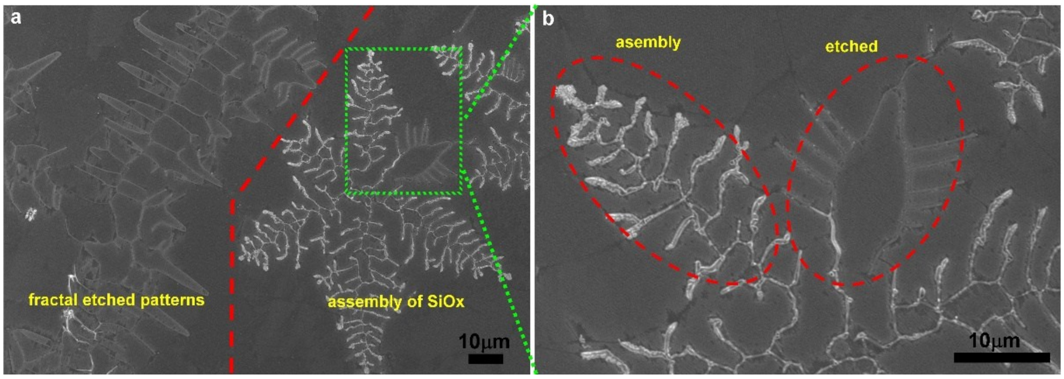

Figure S8. The observation of both graphene fractal etching patterns and assembly of SiO_x NPs on the one sample, indicating the relation between the two processes.

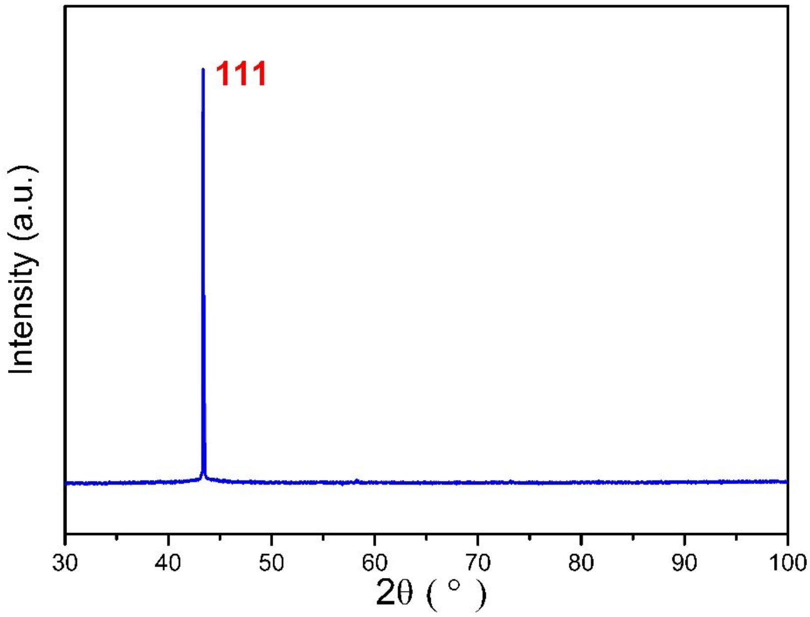

Figure S9. The XRD measurements of the Cu through liquid process, indicating that the liquid Cu owns uniform owned only the Cu (111) crystalline orientation and is beneficial for the growth of SiO_x NPs.

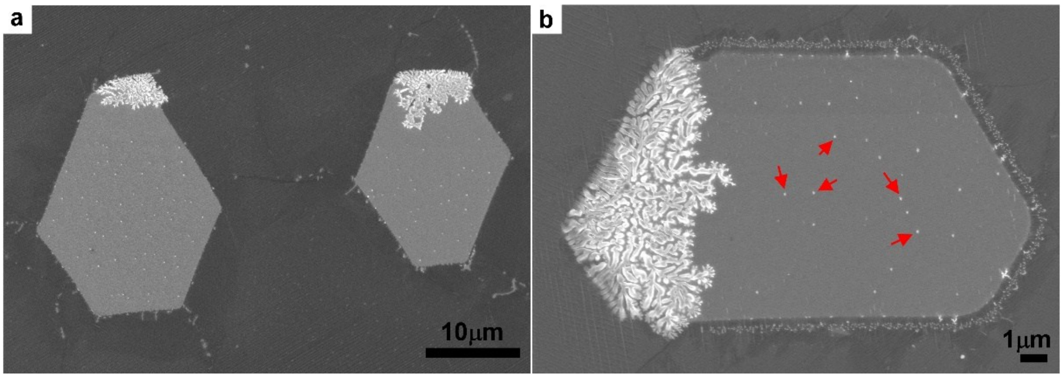

Figure S10. The growth rule of SiO_x NPs on the etched holes in graphene, in which the nuclei usually firstly located at the edges of holes.

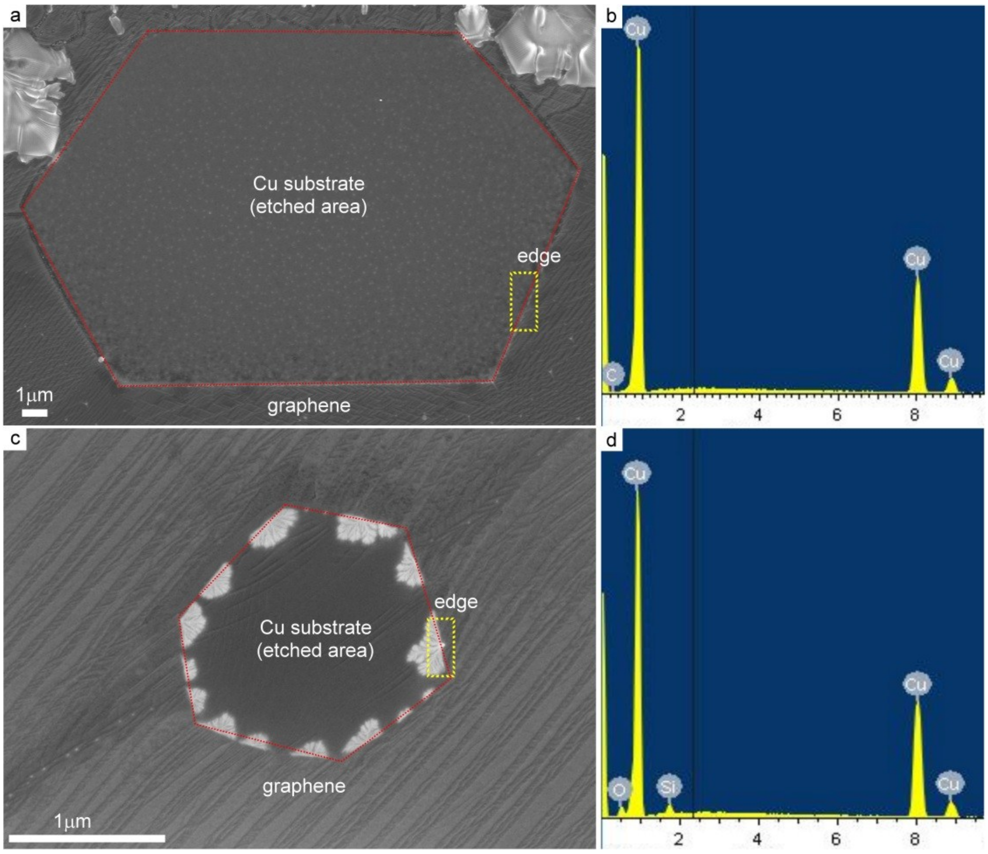

Figure S11. Elemental determination of edges of etched graphene. (a and c) SEM image of hexagonal etched area of graphene on Cu substrate. (b and d) EDX measurements of etched edge, showing the signal of C originated from graphene, while the signal of Si and O, giving a direct evidence for the presence of SiO_x structures.

Table S2. Elemental determinations of the sample 1 and 2, respectively.

Samples	Cu (Mass%/Atom%)	C (Mass%/Atom%)	Si (Mass%/Atom%)	O (Mass%/Atom%)
1	97.79/89.32	2.21/10.68	0	0
2	89.27/75.1	2.32/10.31	2.34/4.46	6.07/10.13

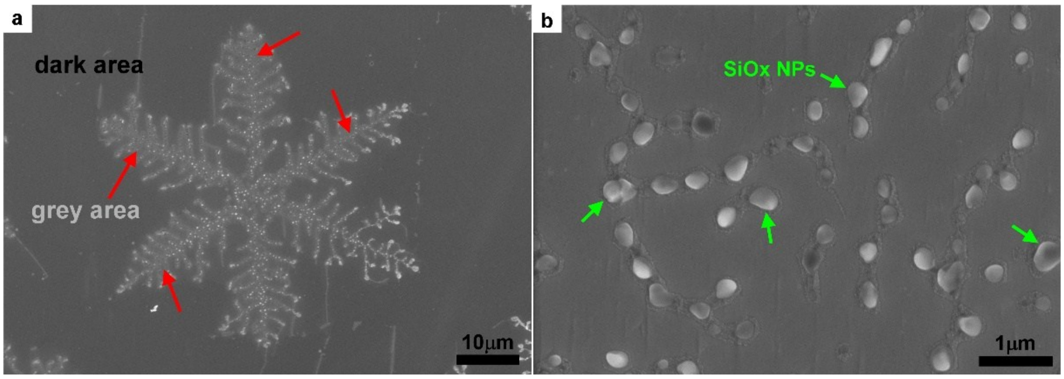

Figure S12. The middle phase of the fractal SiO_x patterns, in which the original NPs are directly found in the graphene.

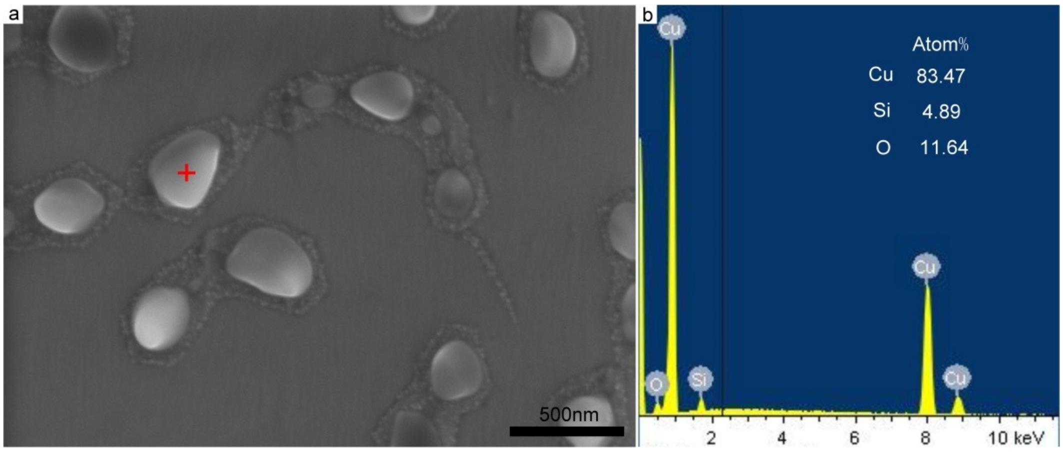

Figure S13. (a) SEM image of SiO_x nanoparticles. (b) EDX spectroscopy of single SiO_x nanoparticle as marked in (a).

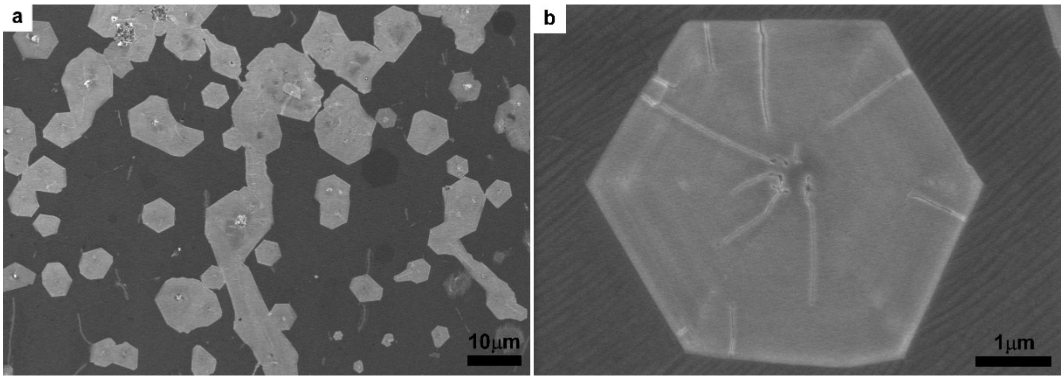

Figure S14. The SEM images of as-grown compact SiO_x patterns in graphene.