Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Material

Edge morphology evolution of graphene domains during chemical vapor deposition cooling revealed through hydrogen etching

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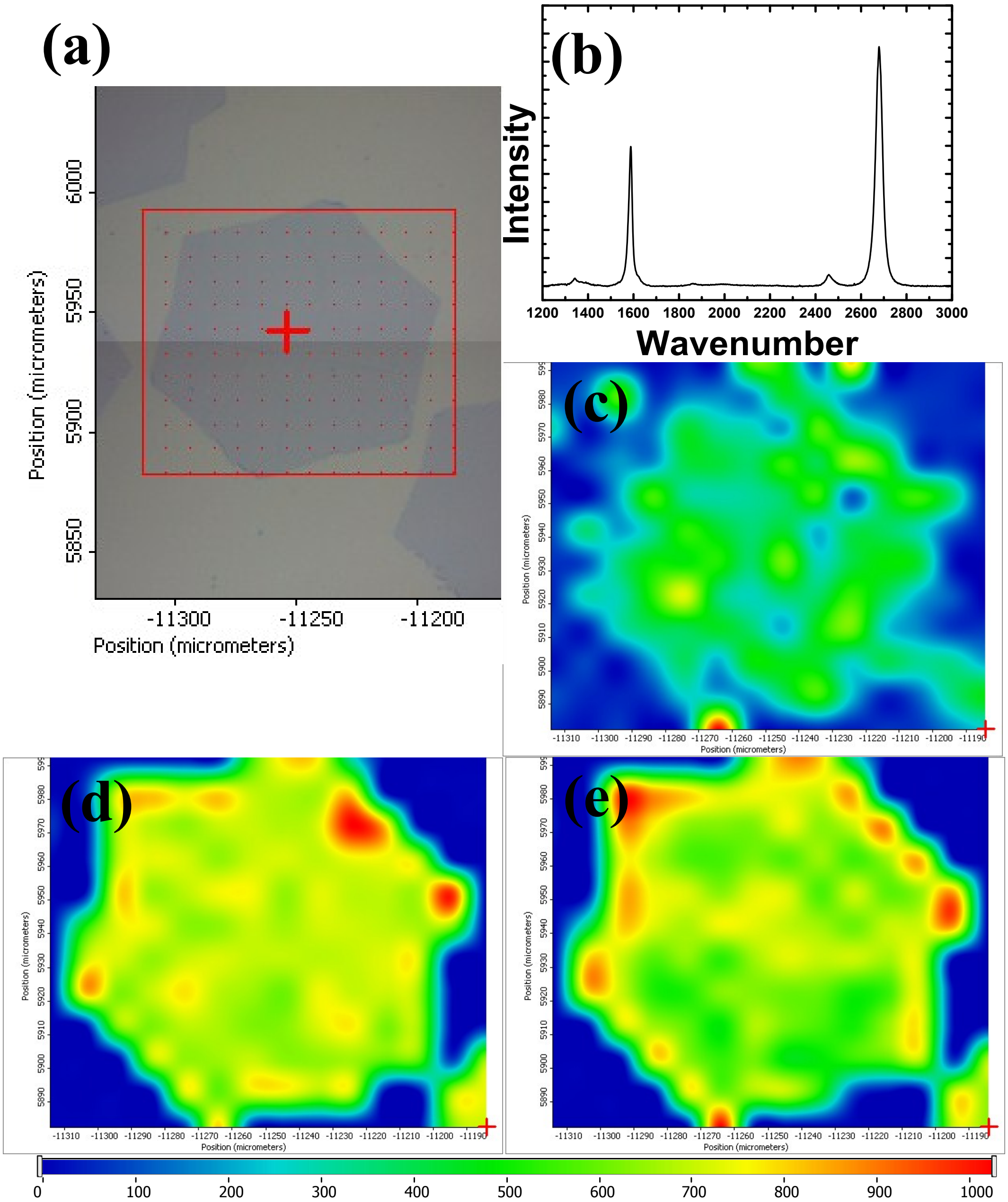

Figure S1. Raman mapping of the synthesized graphene domain: (a) the optical images of the graphene domain; (b) Raman spectrum of the cross location on the graphene in (a); (c) D peak mapping; (d) G peak mapping; and (e) 2D peak mapping.

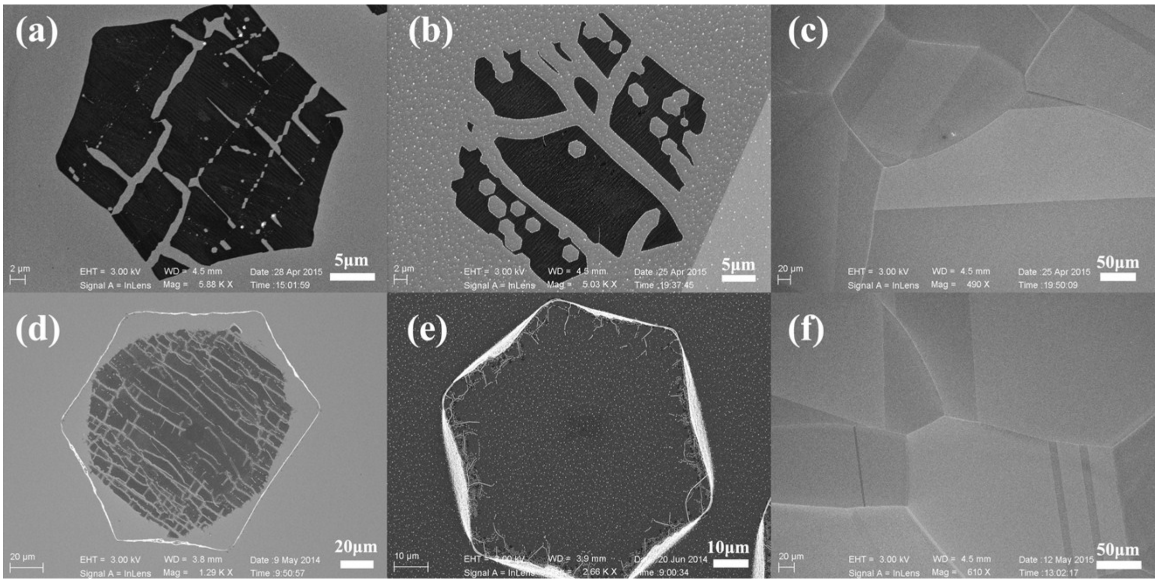

Figure S2. SEM images of the etched domains in different etching temperatures and times. The samples were heated up with 1000 sccm of Ar and 20 sccm of H_2 and annealed with 1000 sccm of Ar and 200 sccm of H_2. (a–c) 10 min, 30 min, and 1 h at 1000 °C, respectively. The etched trenches expand, and hexagonal openings occur. The graphene was fully etched after a sufficient time. (d–f) 30 min, 1 h, and 3 h at 1000 °C, respectively. The graphene was etched from the trenches and domain edge. A white hexagonal trace was left at some of the domain areas. An increasing etching time erased these traces. Scale bar: 20 µm.

A white hexagonal trace appears on some of the domains after hydrogen etching (as shown in Figures S2a to S2f; the etching process details are listed in the caption). All of the samples experienced the growing, cooling, reheating, and etching processes. Figures S2a to S2c show graphene domains etched by hydrogen gas with increasing etching time from 10 min to 1 h at 1000 °C. Etched trenches occur where graphene wrinkles are destroyed by hydrogen (Figure S2a). This observation is consistent with that in the literature. Then, the widths of trenches broaden, and small hexagonal openings are detected (Figure S2b). These hexagonal openings are caused by copper-assisted hydrogen etching along the zigzag direction in graphene. Finally, the graphene domain is fully etched away (Figure S2c). Interestingly, a white hexagonal trace appears around some of the domains, which are etched from the edge (Figure S2d). The graphene residues and the white hexagonal trace are then fully etched, when the etching time is prolonged (Figures S2e and S2f).

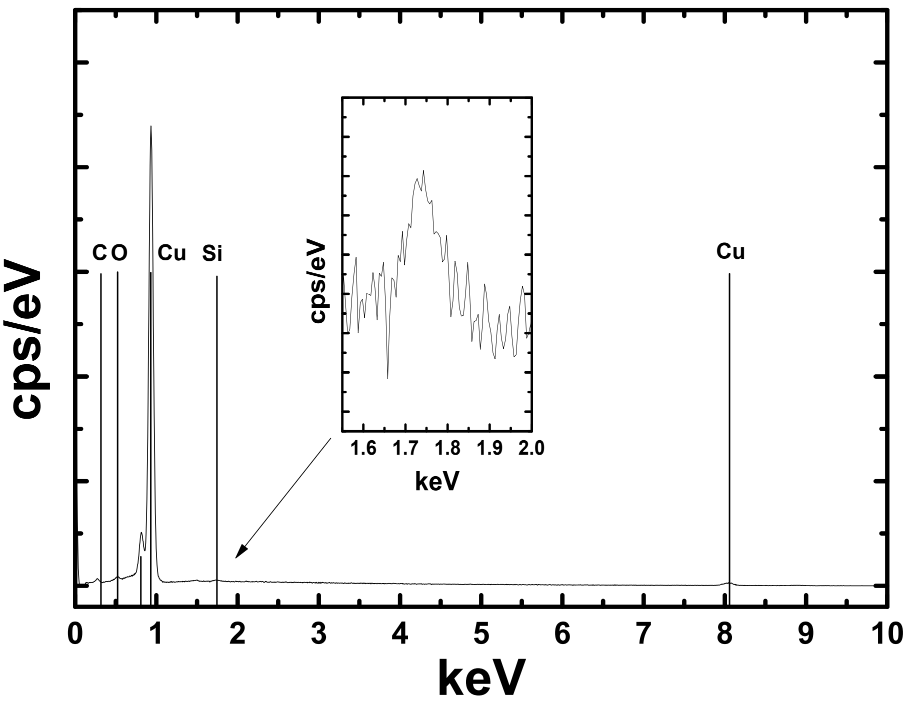

Figure S3. EDS analysis of the white hexagonal trace position after graphene etching.

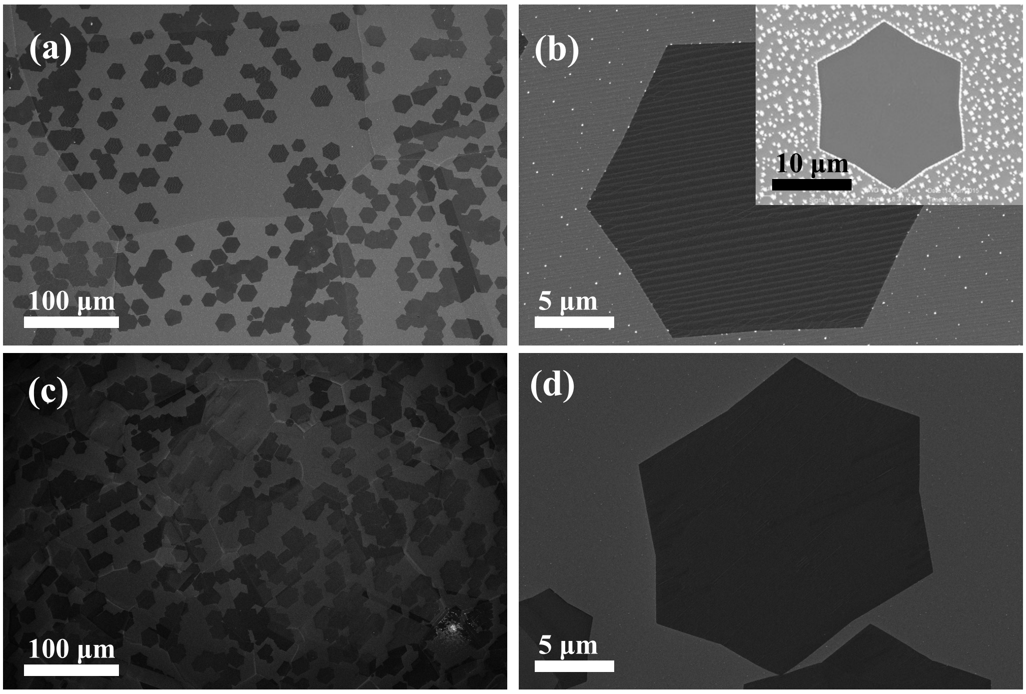

Figure S4. SEM images of graphene domains (a-b) on quartz plate in the alumina tube; (c-d) on alumina plate in the alumina tube. The cooling rate is 10℃/min from 1050℃ to 800℃. The inserted images showed graphene synthesized on the quartz plate in the quartz tube (the cooling rate is the same).

The silicon-containing particles may come from the etching of the quartz tube. For comparison, we put the Cu foils on both quartz (sample A) and alumina plate (sample B) in the alumina tube for graphene growth. The results were shown in Figure S4b. For the same cooling rate, sample in the alumina tube showed less white particles then sample in the quartz ones (Figure S4b). Furthermore, the white particles can be hardly seen in sample B (Figure S4c and S4d). Results indicated that the quartz tube and plate can be the source of the Si-based particles.