Supplementary information for

Flame synthesis of few-layered graphene/graphite films

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Experimental method, structural characterizations of graphene films:

Movie S1, S2

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Experimental methods

In the first setup (Figure S1a), a laboratory alcohol burner was used as burner 1 and a butanefueled Bunsen burner as burner 2. A piece of Ni foil (20 or 50 μ m thick, >99.5 wt.% pure) of 5× 5 mm² was first fixed above the alcohol burner. The Bunsen burner was ignited to produce a blue color torch flame. The torch flame was then pointed onto the Ni foil, heating the foil up to about 850 °C (measured by thermocouple) and lighting up the alcohol burner. After duration of 10 to 40s, the Bunsen torch flame was switched off and the alcohol burner was then capped off immediately. The Ni foil was first surrounded by the inner flame of alcohol burner, and then kept in a closure space made by the alcohol burner cap. Therefore, the Ni foil was cooled down in an atmosphere with little oxygen content.

In the second setup (Figure S1b), to realize large-area deposition, a stainless steel container was used as burner 1 which could generate a large flame for carburization in Ni foils of relatively larger area. The deposition area is essentially determined by the size of the steel container. In our preliminary experiment, a container with an inner diameter of 2 cm was employed.

See Movie S1, S2 for details.

The nickel foil was floating on the etching solution due to the surface tension during etching. After nickel was etched away, a graphene layer along with a thick and black sheet was left floating on the solution. The thick sheet might come from extra carbon content remaining in the nickel foil after graphene precipitation. After gently peeling off the black sheet, the graphene layer was obtained (Figure S1d) and transferred to deionized water for washing.

Characterizations of FLG films

Flame synthesized graphene/graphite films were characterized using scanning electron microscope (SEM, LEO-1530) and transmission electron microscope (TEM, JEM-2010). The

graphene feature along with their crystallinity of the flame samples were characterized by Raman spectroscope (Renishaw 2000 with a 514 nm laser).

It was found that graphene grown on the back side of the foil has higher crystallinity (see D bands in Figure S2a). The line scanning energy dispersive X-ray (EDX) analysis across the cross section of the Ni foil shows a steep drop of carbon content at the backside Ni/C interface (Figure S2b), revealing a thin layer of carbon precipitation. It is worth noting that the carburizing time in the flame synthesis didn't show obvious influence on the FLG deposition (Figure S3a) which was essentially dependent on the cooling rate. We note that addition of water in ethanol fuel has notably reduced the layer number of graphite films. When varying the water concentration from 0 to 60 %, correspondingly increased I_{2D}/I_{G} ratio and I_{G}/I_{D} were concurrently observed (Figure S3d). This could be attributed to the oxidation effect of water vapor which has been confirmed as a weak oxidizer for carbon etching.

Solar cell assembly

n-type Si (100) wafer (doping density: $1.5 \sim 3 \times 10^{15}$ cm⁻³) was coated with a 300 nm SiO₂ layer. A Ti/Pd/Ag layer was sputtered on the back side of the n-Si as back electrode. Flame synthesized graphene film was then transferred to the top of the patterned wafer and naturally dried to make a conformal coating with the SiO₂ layer and the underlying n-Si through a square window (0.1 cm²). Silver was deposited on the graphene film as upper electrode. Forward bias was defined as positive voltage applied to the graphene. The current-voltage data were recorded using a Keithley 2601 SourceMeter. The solar devices were tested with a Newport solar simulator under AM 1.5 condition.

Ag nanoparticle (NP) deposition on graphene film

The deposition procedure includes: i) transferring the graphene film onto a SiO₂ substrate; i) immersing the film in a 10 mM AgNO₃ solution for about 15h; iii) washing the film surface with

distilled water; iv) drying the film under infrared light. For comparison, CVD grown graphene was also used for Ag NP decoration.

Method	$V_{\rm oc}$ (V)	$J_{\rm sc}~({\rm mA/cm}^2)$	FF (%)	η (%)
CVD-Cu	0.450	8.64	26.0	1.01
CVD-Ni	0.470	12.51	42.0	2.46
Flame1	0.445	11.66	27.6	1.43
Flame2	0.495	13.03	44.7	2.88
Flame2+HNO ₃	0.530	13.75	59.7	4.35

TABLE S1. Photovoltaic properties of G/n-Si solar cells



Figure S1. Photographs of the flame synthesis of FLG films. (a) Lab alcohol burner + Bunsen burner. (b) Stainless steel container + Bunsen burner. (c) Schematic phase diagram of the Ni-C system. (d) FLG film floating on the water surface. (d) A FLG film transferred on PET.



Figure S2. (a) Inset: Ni foils inserted in the flame. (b) SEM image and energy dispersive X-ray

(EDX) analysis across the yellow line marked along the thickness.



Figure S3. Raman spectra. (a) Flame-synthesized samples of different carburizing durations. (d) Raman spectra of the flame-synthesized samples using different carbon sources. Inset: the I_D/I_G intensity ratio is increasing with the carbon content. (c) Raman spectra of flame-synthesized samples from acetonitrile and pyridine. Inset: XPS spectrum of FLG from pyridine flame. (d) Flame-synthesized samples from ethanol/water mixture.



Figure S4. (a) Transmission spectra of the flame synthesized FLG films. (b) Sheet resistance versus transmittance (at 550 nm) for different thin films. Data for other thin films were captured from Ref [S1].



Figure S5. 10-day stability of the HNO₃ treated graphene/n-Si solar cell.



Figure S6. (a) XPS spectrum of the flame graphene, SEM images of Ag nanoparticle (NP) deposition on (b) flame graphene and (c) CVD graphene.

Reference:

[S1] Bonaccorso, F.; Sun, Z.; Hasan, T.; Ferrari, A. C. Nature Nanotechnol. 2010, 4, 611.