SUPPORTING INFORMATION

Direct Measurement of Adhesion Energy of Monolayer Graphene As-Grown on Copper and Its Application to Renewable Transfer Process

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1. Graphene syntheses

The chemical vapor deposition (CVD) process with inductively coupled plasma (ICP) is used for the synthesis of the monolayer graphene, as it enables the growth of graphene on copper at a reduced temperature. A 300-nm-thick copper film is deposited on top of a 4-inch Si wafer with 300-nm-thick SiO_2 and is loaded into an ICP CVD chamber. After heating to 725°C at 50 mtorr in Ar ambient, the sample is treated with H_2 plasma at a gas flow rate of 40 sccm and a RF plasma power of 50 W for 2 min. A mixture of Ar and C_2H_2 ($Ar:C_2H_2$=40:1 sccm) is then flowed into the chamber with 150 W of RF plasma for the graphene synthesis process.

2. Characterization of graphene by Raman spectroscopy

The left inset of Figure S1 shows the synthesized graphene layer on a 4-inch copper (300 nm)/SiO_2 (300 nm)/Si (525 μm) wafer. The typical graphene layer transfer technique using copper wet chemical etching is applied for the characterization of the synthesized graphene quality. Figure S1 shows the Raman spectra of three representative points of the graphene layer (denoted as ⓐ, ⓑ, and ⓒ in the left inset of Fig. S1). Negligibly small D (~1350 cm^{-1}) peaks indicate that high-quality graphene is synthesized across the 4-inch wafer. Single-Lorentzian-shaped 2D (~2650 cm^{-1}) peaks and high 2D/G peaks ratio suggest that the synthesized graphene is indeed a monolayer. The right inset of Figure S1 shows the Raman spectrum of as-grown graphene on a copper surface. It shows a different Raman background due to the use of copper as compared to that of graphene transferred onto a SiO_2/Si wafer, serving as a reference spectrum for the high-quality monolayer graphene on copper.

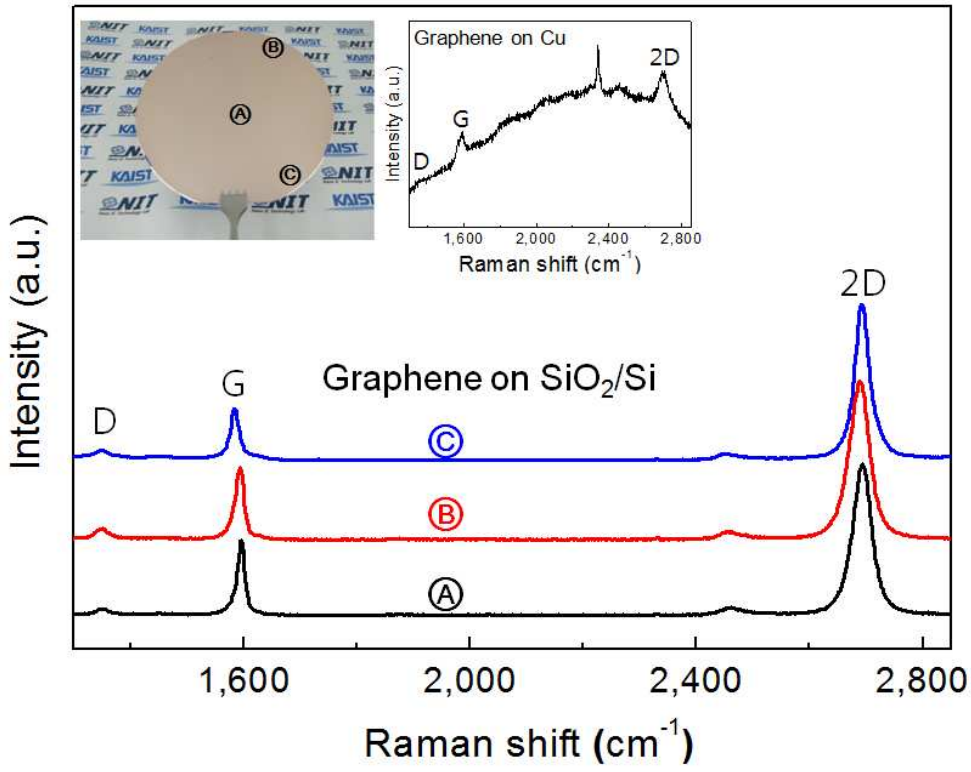

Figure S1. Raman spectra of three representative points (as denoted in the left inset) of graphene as transferred onto a SiO_2 (300 nm)/Si substrate. The left inset shows a photograph of the as-grown graphene on a 4-inch copper (300 nm)/SiO_2 (300 nm)/Si wafer. The Raman spectrum of the as-grown graphene on the copper surface is shown in the right inset.

3. Optical transmittance curves of graphene

To confirm the graphene monolayer quantitatively, the graphene layer is released from the copper and is transferred to soda lime glass (inset of Fig. S2) to measure the optical transmittance (UV2550, Shimadzu). Figure S2 shows nine curves of optical transmittance with respect to the wavelength for nine points of the graphene. They are nearly identical, and the transmittance at a wavelength of 550 nm is 97.4%. This is evidence of uniform and monolayer graphene.

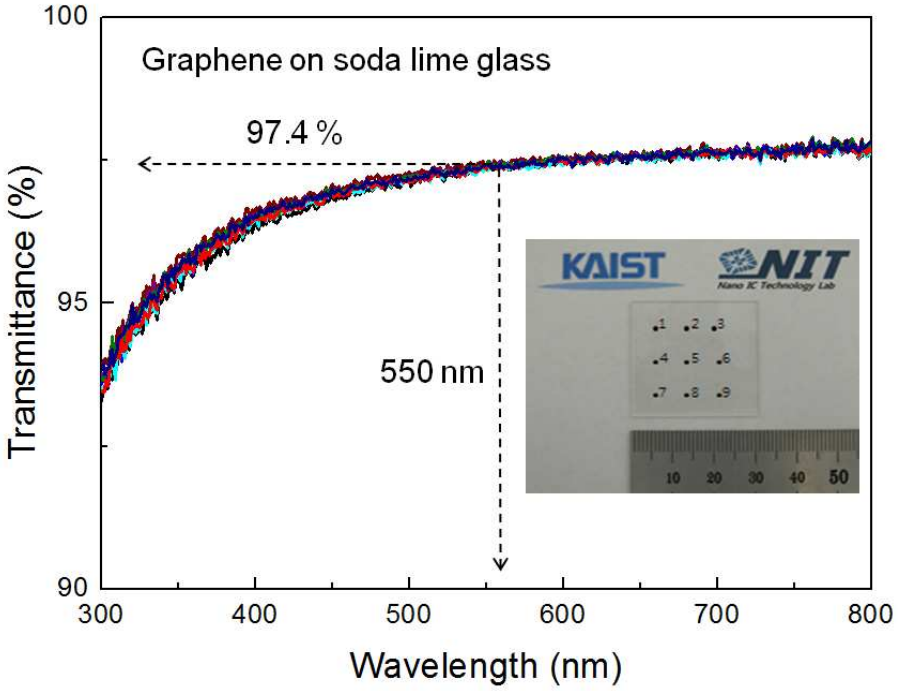

Figure S2. Optical transmittance curves at nine points of graphene on soda lime glass (as numbered in the inset). The inset shows the transferred graphene onto the soda lime glass (30 mm x 30 mm) as used to measure the optical transmittance. The optical transmittance is 97.4% at a wavelength of 550 nm, which coincides with the value for monolayer graphene.

4. DCB specimen preparation

Specimens were fabricated by cutting a 5-mm wide and 30-mm long rectangular beam from a graphene/Cu/SiO_2/Si wafer and bonding it to a SiO_2/Si counterpart (Fig. S3a). A selected epoxy (Epo-Tek 353ND consisting of bisphenol F and imidazole; Epoxy Technology) was used for bonding and a ~5-mm long pre-notch was naturally formed by controlling the application of the epoxy (Fig. S3b). The pre-notch is where the epoxy was not deposited, and therefore the beams were not bonded in this region. Then, the two substrates were attached together with constant clamping pressure (150 kPa), and the epoxy was cured for 2 hrs at 125°C in convection oven (Fig. S3c). This curing conditions result in 1-μm

thick epoxy layer. Aluminum loading tabs containing low-friction ruby jewel bearings were attached to the DCB specimen to measure the low adhesion energy accurately by reducing the friction between the loading tabs and the pins. A commercial epoxy adhesive (DP-420, 3M) was used to attach the tabs on the DCB specimen (Fig. S3d).

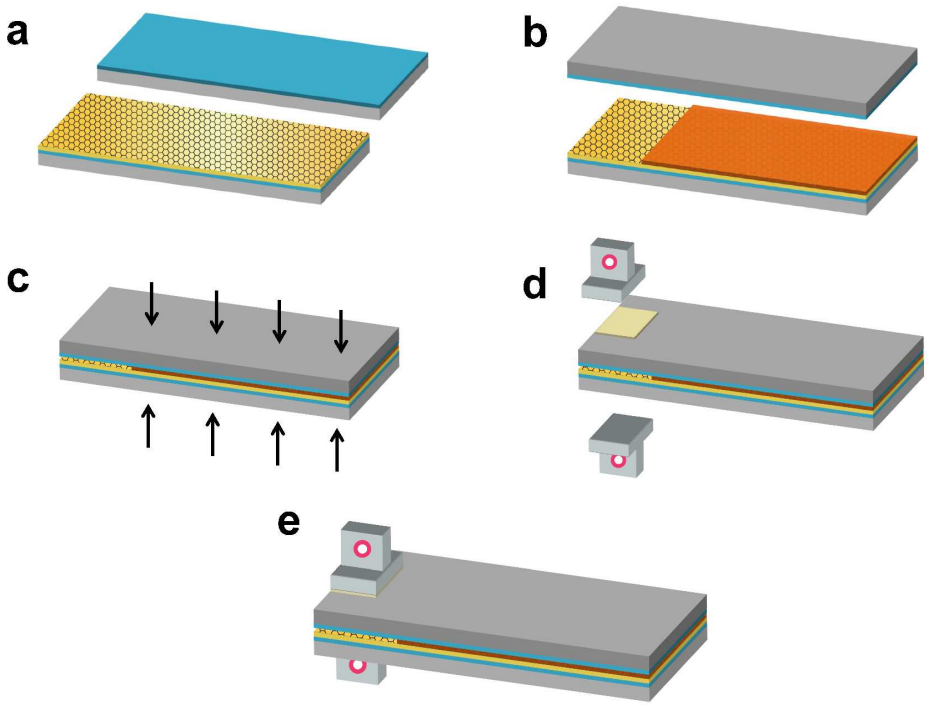

Figure S3. DCB specimen preparation procedures. (a) A silicon substrate containing graphene and a counterpart silicon substrate are cut with same size. (b) Applying epoxy adhesive on selected area of the substrate. (c) Attaching two substrates and curing under constant clamping pressure. (d) Aluminum loading tabs are attached with epoxy adhesive. (e) Completed DCB specimen.

5. DCB testing and data analysis

A photograph of the DCB test equipment is shown in Figure S4. The equipment consists of a linear actuator, loading grips, and a load cell. Aluminum loading tabs of the DCB specimen are inserted in the loading grips with steel pins. Each specimen was loaded and unloaded under a constant displacement rate of 5 μm s^{-1}. The obtained load-versus-displacement curve from a specimen is shown in Figure 1 of the main text.

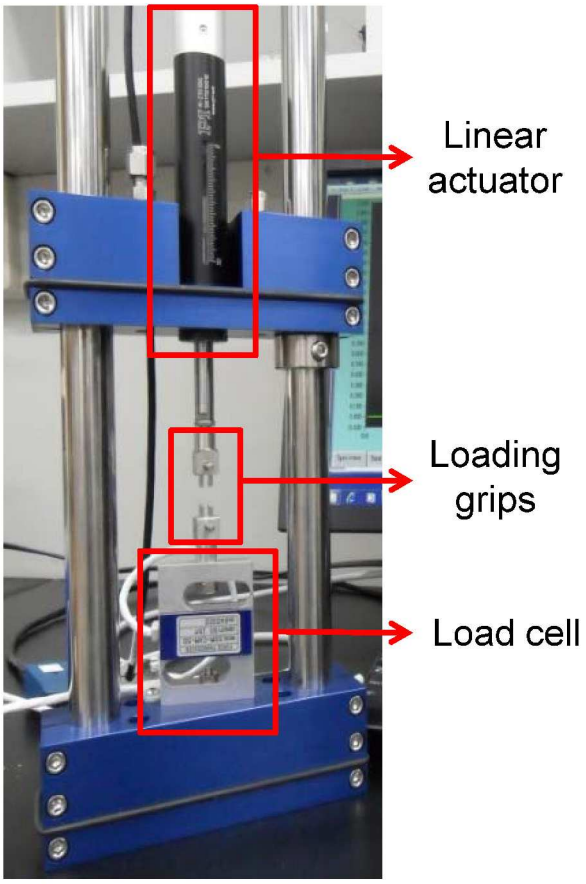

Figure S4. Photograph of the DCB test equipment.

The crack length *a,* and the adhesion energy G_c can be calculated by following equations[1, 2],

$$a = \left(\frac{CE'Bh^3}{8} \right)^{1/3} - 0.64h \tag{1}$$

$$G_c = \frac{12P_c^2 a^2}{E'B^2 h^3} \left(1 + 0.64 \frac{h}{a} \right)^2 \tag{2}$$

where C is the specimen compliance, $d\delta/dP$, δ is the total displacement of the beam ends, P is the applied load, and P_c is the critical load where crack-growth starts in Figure 1. E' is the plain strain modulus of the beam (silicon substrate), B is the specimen width, and h is the half height of the specimen. Several loading/crack-growth/unloading cycles were performed to measure multiple values of the crack length and the adhesion energy from a single specimen.

6. FET fabrication

Source and drain electrodes consisting of Cr (5 nm)/Au (30 nm) were deposited by thermal evaporation onto the graphene directly transferred to PI and were patterned using a lift-off process. Oxygen plasma was used to define the graphene channel region. For the top-gate stack formation, 20 nm thick aluminum oxide (Al_2O_3) was deposited on top of the graphene using atomic layer deposition (ALD) process at a low temperature (130°C). Cr (5 nm) / Au (100 nm) was used as the gate electrode.

REFERENCES

1. Kanninen, M. F. *International Journal of Fracture* **1973,** 9, (1), 83-92.
2. Hohlfelder, R. J.; Maidenberg, D. A.; Dauskardt, R. H.; Wei, Y.; Hutchinson, J. W. *Journal of Materials Research* **2001,** 16, (1), 243-255.