Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary Information

SI 1.

The detailed FE-SEM speciation, measuring conditions (Table SI 1) and calculations of resolution in this work.

Table SI 1. the FE-SEM speciation and measuring conditions in this work

Model	Hitachi, S-4300SE
acceleration voltage	0.5 ~ 30 kV / in this work : 5kV
minimum resolution	1.49 nm (at 15kV) / in this work 2.26 nm (at 5kV)
Magnification	x50 ~ 500,000 / in this work x50,000
Vacuum	2x10^{-7} Torr

-Spherical Aberration-Limited Resolution (Practical Resolution)

The resolution is defined as the Rayleigh criterion, where the minimum resolvable distance, r_{th}, spherical aberration, C_s, between two incoherent point sources is given by

$$r_{th} = 0.91(C_s \cdot \lambda^3)^{1/4} \qquad (SI\,1-1)$$

where λ is wavelength of the accelerated electrons related through

$$\lambda = \left[\frac{12.26}{V(1+0.9778\times10^{-6})^{\frac{1}{2}}}\right](\text{Å}) \qquad (SI\,1-2)$$

for V = 5 kV applied acceleration voltage, wavelength of the accelerated electrons is obtain as $\lambda = 0.173(\text{Å}) = 0.0173(nm)$

From the eq. (SI 1-1) with $C_S = 7.36 \times 10^6$, we have

$$r_{th} = 0.91 \times (7.36 \times 10^6 \cdot 0.0173^3)^{1/4} = 2.26\,nm$$

SI 2.

- Calculation of diffusion coefficients and diffusion length during annealing for alloying and growth process

The diffusion coefficient, D (cm^2/sec), is related to temperature by an Arrhenius equation as below

$$D = D_0 exp\left(\frac{-Q}{kT}\right) \qquad (SI\ 2-1)$$

where D_0 is pre-exponential factor (cm^2/s), Q is the activation energy (in units of eV) for diffusion of species under consideration, k is the Boltzmann's constants (8.62 X10^{-5} eV/K), and T is the absolute temperature (kelvin).

The diffusion length, L (cm), is given as

$$L = \sqrt{D \cdot t} \qquad (SI\ 2-2)$$

where t is the diffusion time (sec)

Table SI 2. Diffusion lengths during annealing for alloying and growth process at 1173 *K*.

Process	Time *t (*sec)	Diffusion (Sample)	*L* (nm)	*D* (cm^2/sec)	D_0 (cm^2/s)	Q (eV)
Annealing for alloying	1,200	Ni in Cu	1,106	3.727X10^{-12}	2.0X10^{-7}	1.0
		Cu in Ni	668.7	1.019 X10^{-11}	6.0X10^{-8}	0.98
growth	20	C in Ni (S1)	38,249	7.31 X10^{-7}	0.013	0.990
		C in S2	29,726	4.42 X10^{-7}	0.013†	1.041‡
		C in S3	24,556	3.02 X10^{-7}	0.013†	1.080‡
		C in S4	16,646	1.39 X10^{-7}	0.013†	1.158‡
		C in S5	12,012	7.21 X10^{-8}	0.013†	1.224‡
		C in S6	5,931	1.76 X10^{-8}	0.013†	1.367‡
		C in Cu (S7)	3,153	4.97 X10^{-9}	0.013†	1.495

† D_0 for carbon diffusion in Cu and Ni_xCu_{1-x} alloy is assumed to be the same as that of C in Ni because Cu and Ni have the same crystalline structure (FCC) and the diameter of Cu atom is almost similar to that of Ni atom.

‡ The activation energies for Ni_xCu_{1-x} alloys are obtained by applying Vegard's law from ref. [SI 2.1 and SI 2.2].

[SI 2.1] J. F. Shackelford, W. Alexander, *Materials Science and Engineering Handbook*, CRC Press, 3rd edn., 2001, ch. 4, pp. 331.

[SI 2.2] S. Dorfman, D. Fuks, Sensors and Actuators A, 1995, **51**, 13

SI 3.

- The Raman mapping (area of 15 μm×15 μm) of intensities of the G and 2D band.

Figure SI 3 shows that the results of the Raman mapping of the G and 2D band in each samples. For measurement, a lab-made laser confocal microscope with a spectrometer was used. With a 0.9 NA objective, the focused spot diameter of the laser light was approximately 300 nm. Scattered light was collected using the same objective and guided to a 50 cm long monochromator equipped with a cooled CCD through an optical fiber with a 200 μm core diameter, which acted as a confocal detection pinhole. The excitation lasers were the 532 nm laser line of diode-pumped solid state laser, with a typical laser power applied to the sample of 0.1 mW and an acquisition time of 250 ms per pixel.

All of Raman mapping image using in this work consist of the 4096 spectra (64 pixel X 64 pixel) in 15 μm × 15μm area for ~300 nm-spatial resolution of confocal microscopy.

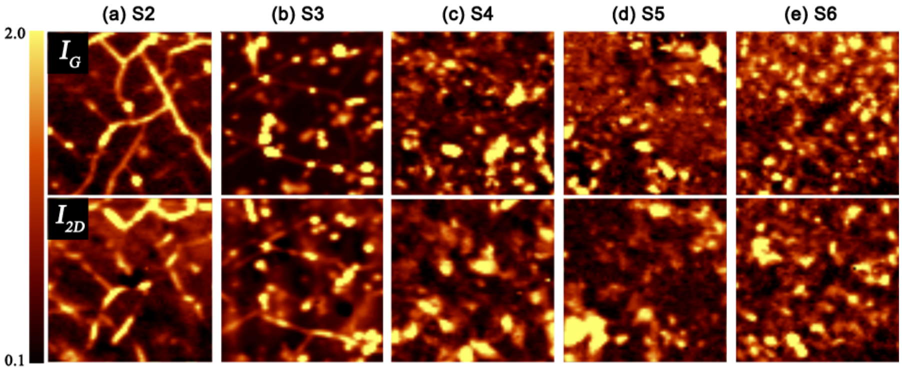

Figure SI 3. The Raman mapping of intensities of the G and 2D band of the S2, S3, S4, S5 and S6 ((a) ~ (e)) with same scale (15 μm×15 μm) and scale bar (0.1~2.0).

SI 4.

- The sheet resistance compared to literature values.

Figure SI 4 shows that the results of the sheet resistances, R_s, compared to literature values. For measurement, 4-point probe (C4S-54/1S, Cascade Microtech) was used for the R_s measurement. The 250 µm radius osmium 4 probes are arranged in a collinear configuration with 1 mm spacing such that a constant current is supplied between the two outer probes and voltage drop is measured using the two inner probes under 40-70 gms spring force (FPP-HS8, DASOL).

Figure SI 4 shows that various R_s value according to the number of graphene layers.

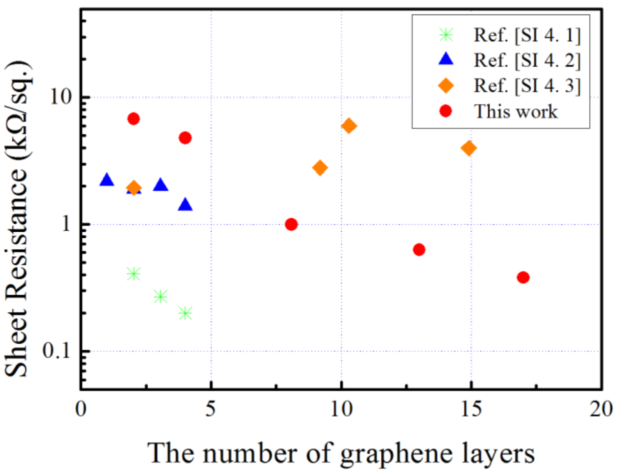

Figure SI 4. The various Rs values according to the number of graphene layers.

[SI 4.1] S. Chen, W. Cai, R. D. Piner, J. W. Suk, Y. Wu, Y. Ren, J. Kang, R. S. Ruoff, Nano Lett., 2011, 11, 3519.

[SI 4.2] X. Li, Y. Zhu, W. Cai, M. Borysiak, B. Han, D. Chen, R. D. Piner, L. Colombo, R. S. Ruoff, Nano Lett., 2009, **11**, 4359.

[SI 4.3] Y. Wang, Y. Li, L. Tang, J. Lu, J. Li, Electrochem. Comm., 2009, **11**, 889.

SI 5.

- Cross-sectional HRTEM images of the S3, S4, and S5.

Figure SI 5 shows that the layer thickness and uniformity of selective graphene samples (S3, S4 and S5).

In order to support the uniformity and crystal structure of the synthesized graphene layers, we use high resolution TEM with an aberration corrector operating at 200 kV. Cross-sectional HRTEM (JEOL, 2200FS) images of four, eight, thirteen graphene layers are shown corresponding to the S3, S4 and S5, respectively.

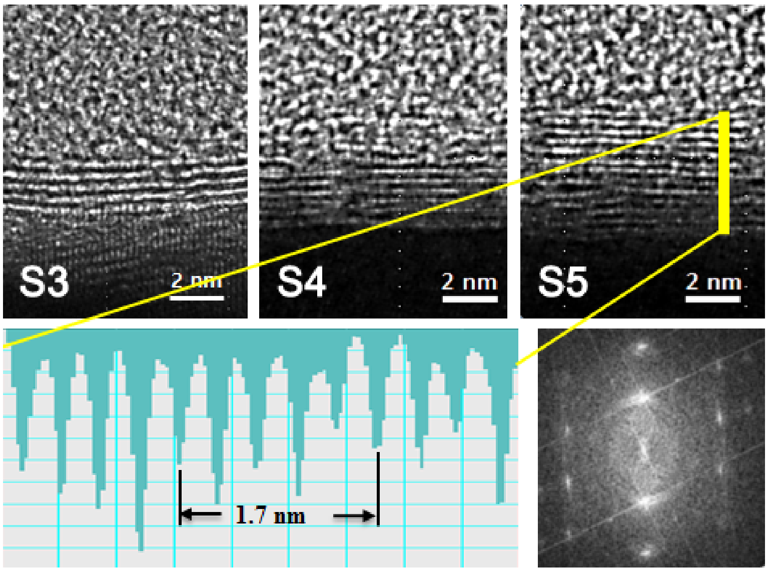

Figure SI 5. Cross-sectional HRTEM images of four, eight, thirteen graphene layers are shown corresponding to the S3, S4 and S5, respectively.

It is shown damaged and uncleared interface between surface and graphene layer by the focused ion beam (FIB) treatments. But these results are consistent with both the raman mapping and transmittance results. The line profile measured at the HRTEM image indicates that the spacing of each layer with 0.34 nm corresponds to the spacing of graphite layers.