Supplementary Information

Selective growth of monolayer and bilayer graphene patterns by rapid growth method

Maddumage Don Sandeepa Lakshad Wimalananda, Jae-Kwan Kim[a], Ji-Myon Lee[a]

[a] Department of Printed Electronics Engineering, Sunchon National University, Suncheon, Jeonnam 57922, South Korea.

Surface free-energy calculation

The Van Oss-Chaudhury-Good method is used to calculate SFE values of Cu surfaces.[1]

$$0.5(1+cos\theta)\gamma_L = \sqrt{\gamma_S^{LW}\gamma_L^{LW}} + \sqrt{\gamma_S^{+}\gamma_L^{-}} + \sqrt{\gamma_L^{+}\gamma_S^{-}} \quad (1)$$

$$\gamma_S^{Total} = \gamma_S^{LW} + 2\sqrt{\gamma_S^{+}\gamma_S^{-}} \quad (2)$$

Where,

θ is the contact angle between the solid and measuring liquid,

γ_L is the SFE of the measuring liquid, and γ_S^{LW} and γ_L^{LW} are the Lifshitz-van der Waals components of solid and liquid,

respectively. γ_L^{-} and γ_L^{+} are the basic and acidic constituents of measuring liquid, respectively,

and γ_S^{-} and γ_S^{+} are the basic and acidic constituents of measuring solid, respectively.

γ_S^{Total} is the total SFE of a solid surface.

Surface tension and components for different chemicals are as follows [R],

Diiodomethane (γ_L=50.8 mJm^{-2}, γ_L^{LW} =50.8 mJm^{-2}, γ_S^{-}=0 mJm^{-2}, and γ_L^{+} =0 mJm^{-2})

Deionized water (γ_L=72.8 mJm^{-2}, γ_L^{LW} =21.8 mJm^{-2}, γ_S^{-}=25.5 mJm^{-2}, and γ_L^{+} =25.5 mJm^{-2})

Formamide (γ_L=58 mJm^{-2}, γ_L^{LW} =39 mJm^{-2}, γ_S^{-}=39.6 mJm^{-2}, and γ_L^{+} =2.28 mJm^{-2})

By using these values to equation (1), the following equations are observed.[1,2]

$$\gamma_S^{LW} = 12.7(1 + \text{Cos}^{\theta_D})^2 \quad (3)$$

$$\gamma_S^{+} = \{0.1747 + 6.0634\ \text{Cos}^{\theta_F} - 2.2757\ \text{Cos}^{\theta_{DI}} - 3.6130\ \text{Cos}^{\theta_D}\}^2 \quad (4)$$

$$\gamma_S^{-} = \{3.7386 - 6.0634\ \text{Cos}^{\theta_F} + 9.4840\text{Cos}^{\theta_{DI}} + 0.3180\ \text{Cos}^{\theta_D}\}^2 \quad (5)$$

Where θ_D is the contact angle between the solid and Diiodomethane, θ_{DI} is the contact angle between the solid and deionized water (DI water), and θ_F is the contact angle between the solid and Formamide, respectively.

By using equations (3), (4), and (5) and contact angle measurements, γ_S^{LW}, γ_S^+, and γ_S^- can be calculated and results can be used to calculate surface free energy using equation (2).

	SFE (mJm^{-2})
2 min (200W)	50.34
4 min (200W)	51.91
10 min (400W)	54.34
Oxygen plasma	N/A
12 min	49.86

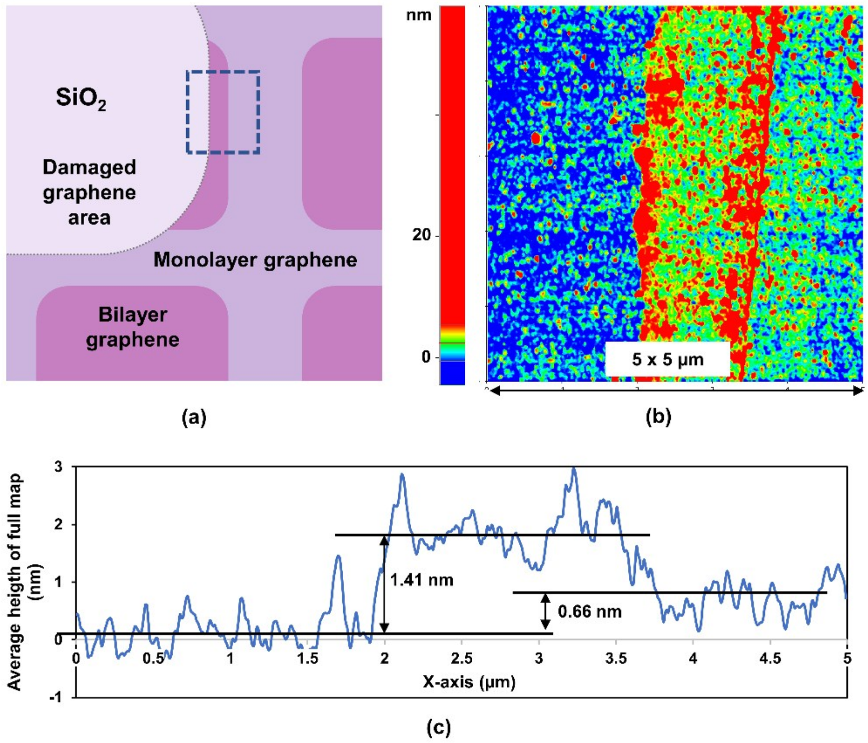

Figure S1. (a) schematic diagram of atomic force microscopy (AFM) mapped area. (b) AFM image. (c) average height profile through the x-axis.

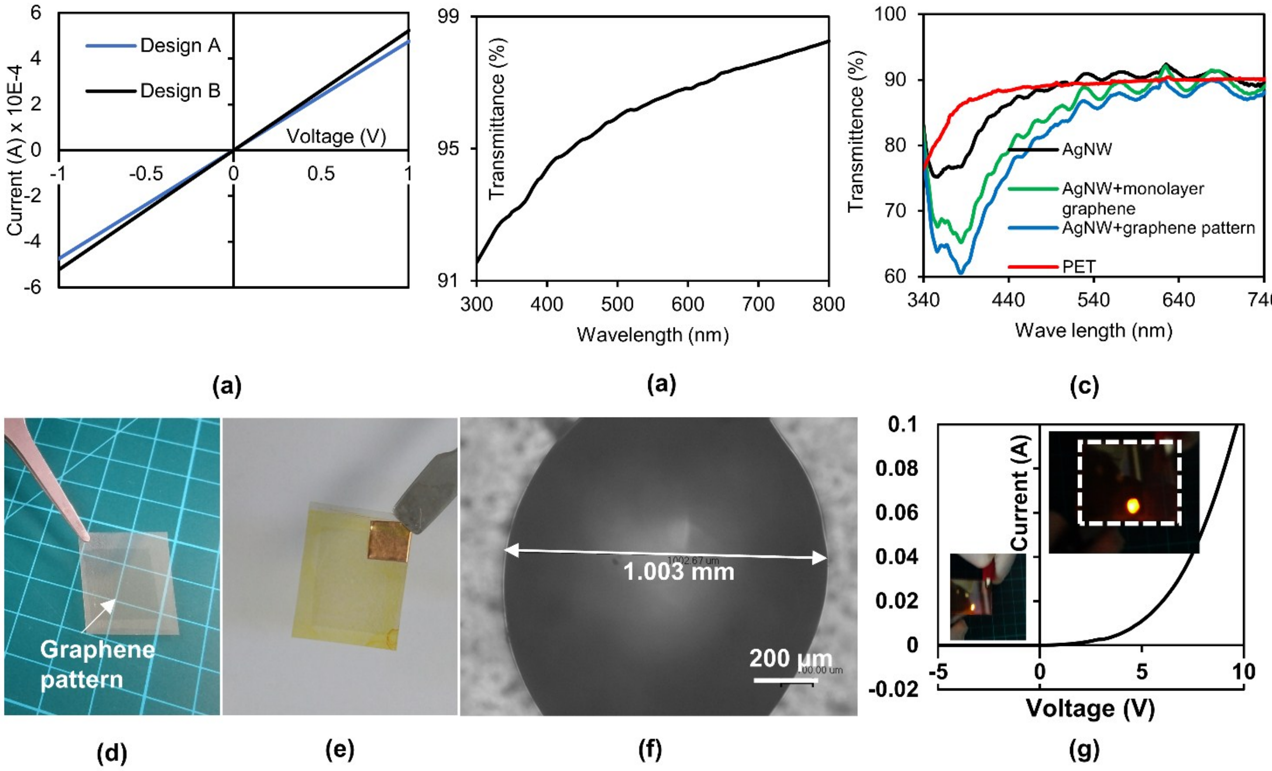

Figure S2. (a) I-V characteristics and (b) optical transparency (only design B) of synthesized graphene patterns. (c) Optical transparency of prepared electrode on PET film and transparency of the PET film. Photographs of (d) AgNW + graphene pattern (anode) on PET film (e) after spin coating of electroluminescent ink. (f) Optical microscopic image of the cathode (Ga:In alloy) area. IV characteristics of the LED prepared by AgNW + graphene pattern [top right corner shows glowing LED at dark (ISO 6400 and shutter speed 100fps) and bottom left corner shows power connection].

According to reports, HOMO (heights occupied molecular orbital) and LUMO (heights occupied molecular orbital) levels of $[Ru(bpy)_3](PF_6)_2$ are 3.1 eV and 5.6 eV, respectively.[3] Therefore, anode and cathode work functions should match with these values. Ga:In alloy is a popular cathode material due to its low work function (4.1 eV). According to reports, AgNW and graphene work functions may vary between 4.2 and 5 eV.[4–6] Therefore, graphene and silver nanowires can be used for the anode.

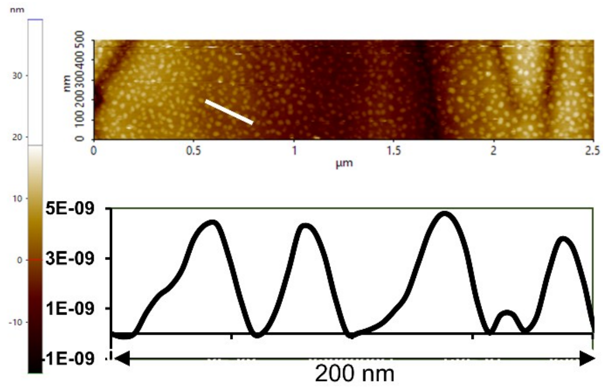

Figure S3. AFM image of Ar plasma treated Cu surface.

References

1 M. Żenkiewicz, *J. Achiev. Mater. Manuf. Eng.*, 2007, **24**, 137–145.

2 M. D. S. Lakshad Wimalananda, J.-K. Kim and J.-M. Lee, *Carbon N. Y.*, 2016, **108**, 127–134.

3 Y. Zhu, C. Gu, S. Tang, T. Fei, X. Gu, H. Wang, Z. Wang, F. Wang, D. Lu and Y. Ma, *J. Mater. Chem.*, 2009, **19**, 3941–3949.

4 R. C. Chiechi, E. A. Weiss, M. D. Dickey and G. M. Whitesides, *Angew. Chemie - Int. Ed.*, 2008, **47**, 142–144.

5 S. R. L. E. B. K. S. K. P. K. Young-Jun Yu Yue Zhao, Y.-J. Yu, Y. Zhao, S. Ryu, L. E. Brus, K. S. Kim and P. Kim, *Nano Lett.*, 2009, **9**, 3430–4.

6 K. Naito, N. Yoshinaga, S. Matake and Y. Akasaka, *Synth. Met.*, 2014, **195**, 260–265.