## Electronic Supplementary Information

# In-situ construction of multifunctional femtosecond laser-induced graphene on arbitrary substrates

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#### **Supplementary Notes**

**Note S1: COMSOL simulation of temperature distribution in femtosecond laser irradiating PI tape.** The thermal simulations were performed using the Heat Transfer Module based on finite element method (FEM) in COMSOL Multiphysics software. In this model, the temperature after laser pulse irradiation evolution on the surface of PI tape can be described by the thermal conduction equation:

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p u \nabla T + \nabla \cdot (q) = Q$$
(S1)

where  $\rho$  is the density,  $C_p$  is the heat capacity, T is the temperature, **u** is the fluid flow speed, q is the heat flux density, and Q is the local heat source from the laser absorption of PI tape, respectively. Due to the femtosecond scale and extremely high temperature, the effects of heat radiation and heat conduction were mainly considered, while heat convection was neglected. The boundary condition of laser and PI interface is given by the equations:

$$-n \cdot (-k\nabla T) = Q_L \tag{S2}$$

$$-n \cdot (-k\nabla T) = \varepsilon \sigma \left(T_{amb}^{4} - T^{4}\right)$$
(S3)

where k is the thermal conductivity,  $\varepsilon$  is the surface emissivity,  $\sigma$  is the Stefan-Boltzmann constant, T<sub>amb</sub> is the ambient temperature, and Q<sub>L</sub> is the heat flux generated by the laser respectively. The laser was modeled as an area heat source. The following equation gives the laser heat source expression:

$$Q_L = \frac{2AP_L exp\left(-\left(\frac{x^2 + y^2}{r_s^2}\right)\right)}{\pi f_L r_s^2 t_L}$$
(S4)

where A is the laser absorption of LIG,  $P_L$  is the laser power,  $r_s$  is the laser beam radius,  $f_L$  is the laser repetition rate, and  $t_L$  is the laser pulse width, respectively. Additionally, the boundary condition of other interfaces is given by the following equation:

$$-n \cdot (-k\nabla T) = 0 \tag{S5}$$

All the relevant parameters used in the simulation were obtained from experimental data. The time step size used in the FEM calculation is 1 fs. The laser heating time was set as 350 fs.

### **Supplementary Figures**



**Figure S1.** Water droplets placed on the pristine and treated substrates of four substrates (paper, aluminum, ceramic, and silicon); for convenience of observation, the water droplets have been stained with methylene blue.



Figure S2. Detachability test of FsLIG on aluminum.



Figure S3. SEM images of the pristine PI tape surface.



Figure S4. SEM images of pristine a) paper, b) aluminum, c) ceramic, and d) silicon surfaces.



Figure S5. Calculated crystalline sizes (L<sub>a</sub>) of FsLIG on paper, aluminum, ceramic, and silicon.



Figure S6. Dragging process and vertical anti-adhesion measurement.



**Figure S7.** Infrared camera images of P-FsLIG surface under 1.0, 2.0, 3.0, and 4.0 sun illumination. The images were recorded after illumination for 240 s.



Figure S8. I–V linear curve of P-FsLIG indicating a correlation coefficient (R<sup>2</sup>) of 0.999.



**Figure S9.** Infrared camera images of P-FsLIG surface at the voltages of 3, 4, 5, and 6 V. The images were recorded after applied voltage for 240 s.

### **Supplementary Tables**

Materials	Wettability	Optical Property	Electrical Property	Temperature sensing	Reference
FsLIG from PI tape	Superhydrophobic (~153.4°)	~98.8% (400–1600 nm)	132.1 °C at 6 V 297.2 °C cm <sup>2</sup> W <sup>-1</sup>	-0.089% °C <sup>-1</sup> (30-80 °C)	This work
TPE based LIG film	Not discussed	Not discussed	70 °C at 6 V Not given	Not discussed	S1
LIG on mask	Hydrophobic (~141°)	>95% (300–2500 nm)	Not discussed	Not discussed	S2
LIG on glass	Not discussed	~30.0% (300–2500 nm)	~82 °C at 40 V Not given	~-0.1% °C <sup>-1</sup> (30-80 °C)	S3
LIG from Kevlar	Not discussed	96.8% (190–2500 nm)	Not discussed	~-0.068% °C <sup>-1</sup> (30-60 °C)	S4
LIG from wood	Not discussed	Not discussed	127.4 °C at 5 V Not given	~-0.025% °C <sup>-1</sup> (30-70 °C)	S5
LIG from poly(Ph-ddm)	Superhydrophobic (~161°)	>98.0% (250–2500 nm)	~176 °C at 4 V ~222 °C cm <sup>2</sup> W <sup>-1</sup>	Not discussed	S6
FsLIG from PI film	Hydrophilic (~61.3°)	~98.5% (220–1400 nm)	195.7 °C at 4 V 215.9 °C cm <sup>2</sup> W <sup>-1</sup>	Not discussed	S7
LIG from PEI film	Hydrophobic (~97°)	Not discussed	~229 °C at 7 V Not given	Not discussed	S8
AgNPs/LIG (Lased in N <sub>2</sub> )	Hydrophobic (~140°)	~95.0% (500–2500 nm)	~46 °C at 7.5 V Not given	Not discussed	S9
Fluorine- doped LIG	Superhydrophobic (~159.2°)	Not discussed	~95 °C at 8 V ~214 °C cm <sup>2</sup> W <sup>-1</sup>	Not discussed	S10

Table S1. Comparison of the FsLIG multifunctionality with other LIG materials from the literature.

Note: Optical property refers to the absorption of the samples. Electrical property means the stable temperature at a specific voltage and the whole electrothermal conversion efficiency. PI: polyimide. TPE: thermoplastic elastomer. F-LIG: Forest-like LIG. PEI: polyetherimide.

#### **Supplementary Movies**

**Movie S1. Self-cleaning demonstration of the P-FsLIG surface.** The chalk powder was picked up by the rolling water droplets and readily removed.

**Movie S2. A water droplet impacting on the P-FsLIG surface.** The water droplet experienced falling, spreading, retracting, and finally rebounding back into the air.

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