

Supplementary Information for:

**High electrothermal performance of  
expanded graphite nanoplatelet-based patch heater**

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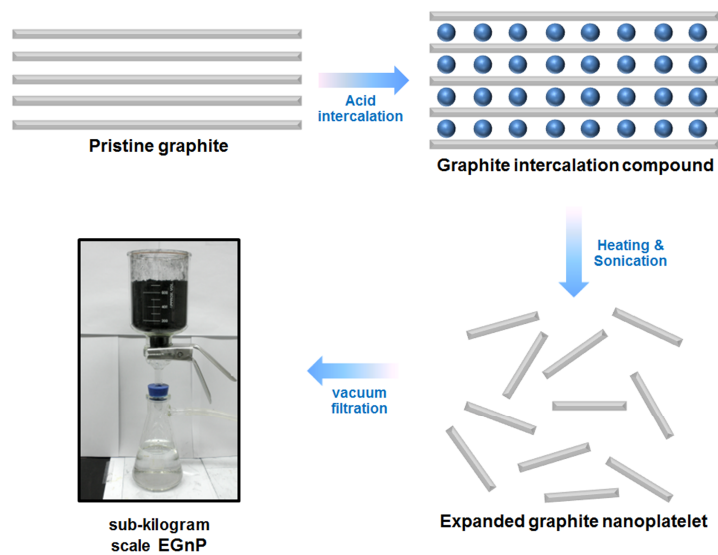
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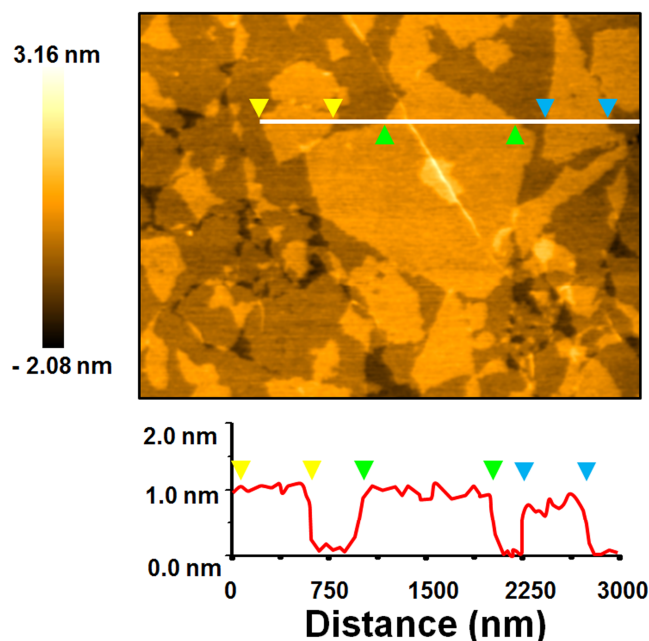
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## 1. Schematic illustration of mechanism for EGnP



**Fig. S1** Schematic illustration of mechanism for sub-kilogram scale of EGnP.

## 2. AFM image of exfoliated GO

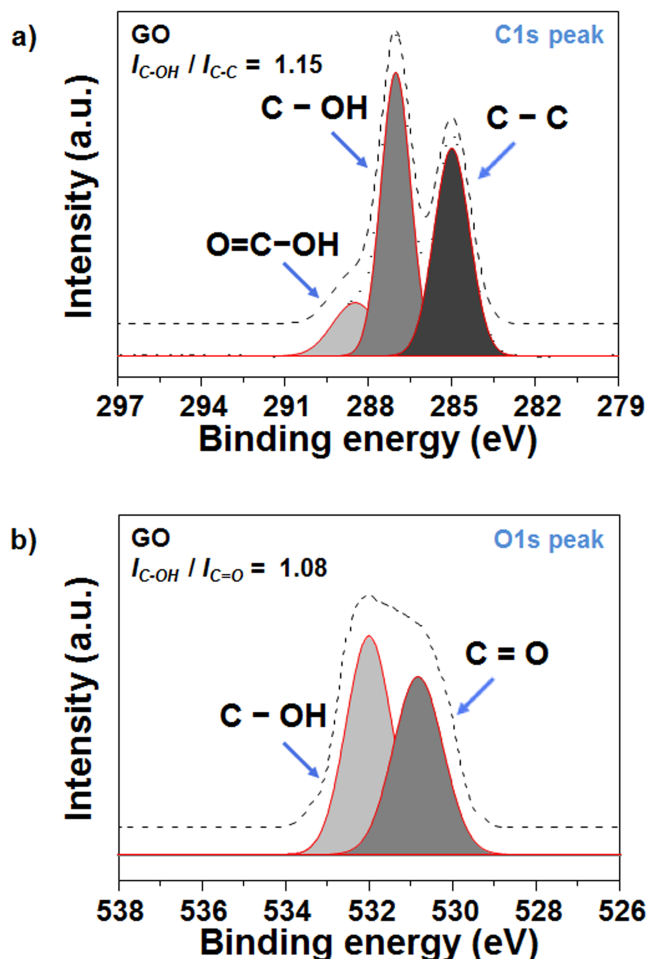


**Fig. S2** Representative atomic force microscopy (AFM) image of the exfoliated GO. For the sample preparation, the exfoliated GO solution was deposited on Si wafer.

We also experimented on the synthesis of exfoliated graphene oxide (GO) to compare with pristine graphite and EGNP. Briefly, unreduced graphene oxide was synthesized from natural graphite by a modified Hummers and Offeman method as originally presented by Kovtyukhova and colleagues.<sup>2</sup> Synthesized purified GO suspensions were dispersed in water. Exfoliation of GO was achieved by sonication of the dispersion for 3 h. The obtained brown dispersion was then washed for 5 cycles of centrifugation at 3,000 r.p.m. to remove any unexfoliated graphite oxide.

The atomic force microscopy (AFM) topography image (Figure S2) displays that the exfoliated GO is composed of a few micrometer-sized monolayers ( $< 1.0$  nm), which means that the GO can be readily exfoliated to form monolayer graphene, dispersing in a stable aqueous solution.

### 3. XPS spectra of exfoliated GO



**Fig. S3** Deconvoluted XPS spectrum of exfoliated GO for a) C1s and b) O1s. The fitted lines of the XPS analysis are labeled as C-C, C-OH and O=C-OH bonds in the C1s region, and C-OH and C=O bonds in the O1s region. For XPS sample preparation, an exfoliated GO aqueous solution was deposited on a Si wafer using spin coating.

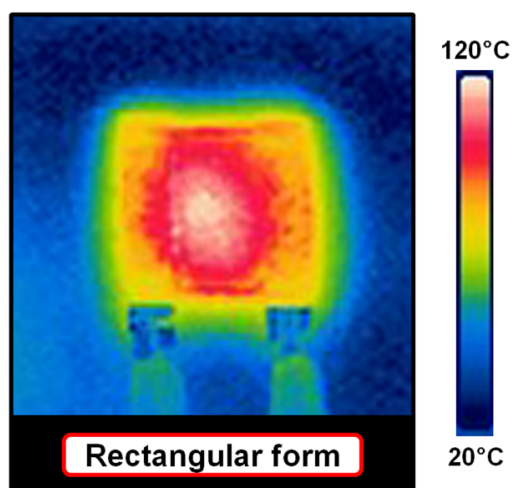
The deconvoluted C 1s and O 1s XPS spectra of exfoliated GO are presented in Figure S3. Generally, the C 1s signal of exfoliated GO mainly consists of three components, which are C - C bond in aromatic rings (284.6 eV), C - OH bond (286.5 eV), and O = C = OH (288.5 eV) peaks (Figure S3a). As shown in C 1s XPS data of pristine graphite and EGnP (Figure 3a), the

ratio of the  $I_{C-OH}/I_{C-C}$  increased from 0.15 (graphite) to 1.15 (exfoliated GO). From the data, it could be conferred that the intensity ratio was proportional to the degree of exfoliation. The O 1s XPS region was fitted to a combination of C – OH bond (532.1 eV) and C = O (531.2 eV) peaks. Interestingly, physical water adsorption peak (533.8 eV) has disappeared, while it presented in O 1s XPS data of graphite and EGnP. It indicates very small amount of trapped water in exfoliated GO relative to the extent of surface functional group such as C – OH and C = O.

In general, the typical d-spacing of completely dehydrated GO was ca. 0.567 nm and this interlayer distance is mainly attributed to the lamellar structure of GO.<sup>3</sup> Water molecules in GO would be strongly bound to the host molecules like several ions at low concentration, being its reorientation highly restricted. However, the rotational freedom of water molecules increases at higher water concentration, since an excess amount of water molecules is surrounded by other water molecules. For this reason, the direct contact with the host molecules can be restricted. From this point of view, the interaction can be key to the ionic conductivity in GO. In other words, the ionic conductivity has maximizing value as the active sites on the GO sheets by increasing water molecules become saturated.

In our experiment, the small amount of trapped water in EGnP clearly enhanced its conductivity with increasing the molar concentration of the oxidizing agent. In the end, EGnP had a low surface resistance,  $10 \Omega\text{sq}^{-1}$  for 150 mM concentration oxidizing agent, which conferred that the active sites on EGnP become saturated. However, our dried GO sample had a very small amount of water molecules on GO sheets and it had a higher interlayer spacing value than that of EGnP (0.341 nm). Therefore, it indicated that the ion conductivity by water molecules in GO was very low and it has little effect on the conductivity of GO. Furthermore, introducing functional groups between carbon layers of graphite by chemical oxidation method could make Van der Waals bond between the carbon layers weaken, which caused the graphene layers to peel off layer by layer from graphite. As  $\text{sp}^2$  hybrid carbon atoms in graphite are partially degraded into  $\text{sp}^2$ – $\text{sp}^3$  hybrid atoms, fully exfoliated graphene oxide possesses less  $\pi$ – $\pi$  stacking stability and poor conductivity.

#### 4. Infrared thermal images of EGnP-based rectangular heater

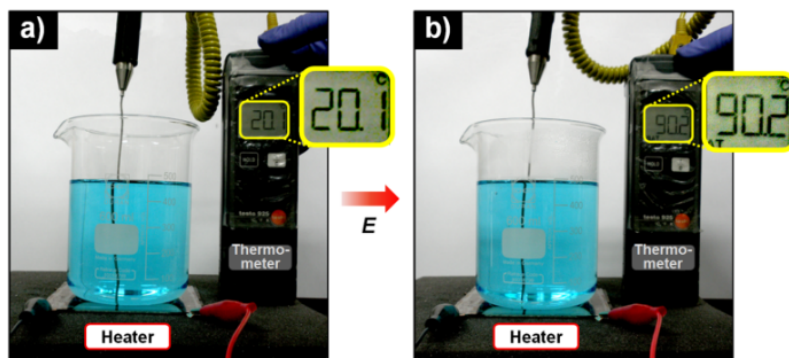


**Fig. S4** The infrared thermal images of EGnP-based heater while applying an input voltage of 3 V in 60 s. The size of rectangular heater was  $4 \times 4$  cm.

## 5. Heat performance of exfoliated and reduced GO-based line-heater

We have compared the screen printing ink condition with fully exfoliated GO and reduced GO (rGO) for fabrication of patterned line-heater. In the case of exfoliated GO for screen printing ink, it can be also well formed on the flexible substrate as desired by formed open area of mesh due to its outstanding dispersibility in aqueous solution. For the reduction process, patterned GO film on flexible substrate was properly cut and located in 95 °C vapor deposition chamber containing hydrazine for 1 h under vacuum. Then, the graphene were immediately formed at low temperature. It clearly demonstrated that the process was more complexed than the screen printing method using EGnP. Most of all, the surface resistance, which direct related to the heat performance, decreased to *ca.* 60  $\Omega\text{sq}^{-1}$  and thus the steady state of temperature could increase to *ca.* 30~40 °C at 12V of applied voltage. Compared to EGnP-based line heater, GO-based line heater had very low heat performance. For using rGO as a screen printing ink, synthesized GO aqueous solution by Hummers method was reduced to rGO using hydrazine and ammonia solution. After the several times washing, rGO aqueous solution was prepared for screen printing ink to pattern the line-heater. As a result, the heat performance of rGO-based line heater was much lower than that of exfoliated GO-based line heater. The concentration of rGO in distilled water can be maximized to *ca.* 0.1~0.2 wt% for use of screen printing ink, since rGO become self-aggregated over the concentration. For this reason, the rGO-based line heater had very high surface resistance ( $> \text{k}\Omega\text{sq}^{-1}$ ) in spite of repetitive screen printing. From this point of view, synthesized EGnP-based line heater had outstanding heat performance compared with exfoliated GO and rGO-based line heater.

## 6. Water heating test of EGnP-based line heater



**Fig. S5** Water heating results a) before and b) after operation of the EGnP-based line heater under an applied voltage of 14 V. The volume of the water used for the boiling test was 500 mL and the time required to raise the water temperature (ca. 90.2°C) was 30 min.



**\* References**

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**2 (a) W. Gao, N. Singh, L. Song, Z. Liu, A.L.M. Reddy, L. Ci, R. Vajtai, Q. Zhang, B. Wei, P.M. Ajayan, *Nature Nanotechnology*, 2011, 6, 496; (b) S. Cervený, F. Barroso-Bujans, A. Alegría, J. Colmenero, *J. Phys. Chem. C.*, 2010, 114, 2604;**