Supplementary information for

Free-standing laser energy converter based on energetic graphene oxide for enhanced photothermic ignition

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This PDF includes:

Supplementary text, Supplementary Figs 1-7, Reference 1-3.

Supplementary text

Statistical analysis of interlayer distance of graphene membranes (GM900) and graphene oxide membrane (GOM) based on the TEM images. For each TEM image, five line cursors which are perpendicular to the layer-stacked direction were addressed at the different location to get comprehensive information of interlayer distance. Then the interlayer distance along the cursors can be obtained automatically by the software. For example, Supplementary Fig. 3 exhibited a typical TEM image of graphene membrane annealed under 900 °C in Argon (GM900), with five line cursors perpendicular to the layer-stacked direction. The location mapping of the line cursor A, B, C, D and E was shown in the Supplementary Fig. 4, each strong positive peak presents one single layer of graphene. Therefore, the position of the graphene layers along the line cursors in GM900 can be obtained by locating all the strong positive peaks. Then the interlayer distance can be further obtained by calculating the difference of interfacing peak position. The detailed data for each interlayer distance for GM900 was listed in the Supplementary Table 1. As another example for statistical analysis of interlayer distance based on the TEM images, the detailed data for GOM was also listed in the Supplementary Figs. 5 and 6 and Supplementary Table 2 for the reader's convenient.



Supplementary Figure 1. Thermogravimetric analysis of GOM and GM900 in air.

The mass loss in the range of 110-230 °C can be ascribed to the removal of oxygen-containing groups, such as C–OH, C=O, and O=C–OH functional groups¹, while the mass loss in the range of 500-600 °C is due to the carbon combustion. As shown in the Supplementary Fig. 1, compared with GM900 that has no mass loss in the range of 110-230 °C, GOM exhibits about 30 wt% of mass loss that can be assigned to the decomposition of the oxygen-containing groups¹. According to the differential scanning calorimetry results for GOM and GM900 in Fig. 3d, extra heat can be released during the the decomposition of the oxygen-containing groups in GOM, which offers GOM unique energetic property and is the key reason for the much higher temperature rise of GOM than GM900 under the irradiation of same laser pulse.



Supplementary Figure 2. (a) X-ray photoelectron spectroscopy (XPS) analysis of GOM, GM200 and GM900. GOM exhibited lowest C/O ratio due to the highest content of oxygen-containing groups. The deconvoluted C1s spectrum for XPS analysis of (b) GOM, (c) GM200 and (d) GM900. The oxygen-containing groups in GOM and GM200 was mainly composited of C-O-C and C=O bond.

X-ray photoelectron spectroscopy analysis was employed to study the composition of oxygen-containing groups in GOM, GM200 and GM900. The characteristic peak that located at 284.5 eV is due to the C=C bonds, while the characteristic peak at 286.5 and 287.9 eV can be ascribed to C-O-C and C=O bond², respectively. This indicated that the oxygen-containing groups in GOM and GM200 was mainly composed of epoxide and carbonyl groups². The atomic ratio between carbon and oxygen is 1.9 for GOM, which is much lower than that of 5.9 for GM900. The much larger content of the oxygen-containing groups in GOM than GM900 is the key factor for the good photothermic effect of the GOM.



Supplementary Figure 3. Typical TEM image of GM900 in Argon, with five line cursors perpendicular to the layer-stacked direction.



Supplementary Figure 4. Location mapping of the line cursors A-E in the Supplementary Fig. 3. Each positive peak presents an individual graphene layer in GM900. The position of each individual graphene layer along the line cursors in GOM was obtained by locating all the positive peaks. The interlayer distance was obtained by calculating the difference of interfacing peak position.



Supplementary Figure 5. Typical TEM image of graphene oxide membrane, with five line cursors perpendicular to the layer-stacked direction.



Supplementary Figure 6. Location mapping of the cursors A-E in the Supplementary Fig. 5. Each positive peak presents an individual graphene oxide layer in GOM. The position of each individual graphene oxide layer along the line cursors in GOM was obtained by locating all the positive peaks. The interlayer distance was obtained by calculating the difference of interfacing peak position.



Supplementary Figure 7. The distribution of interlayer distance for the (**a**) GOM and (**b**) GM900 based on the statistical analysis for the TEM images. The maximum distribution of interlayer distance for GOM and GM900 is 0.760 nm and 0.355 nm, respectively. (**c**) X-ray diffraction results of GOM and GM900, which is in good accordance with the statistical analysis result based on the TEM images.

Based on the statistical analysis from the TEM images, the maximum distribution of interlayer distance for GOM and GM900 is 0.760 nm and 0.355 nm, respectively (Supplementary Fig. 7a, b). As another way to provide proof for the interlayer distances of graphene membranes/films/papers^{2,3}, X-ray diffraction was also performed to GOM and GM900 for comparing with the results from statistical analysis based on the TEM image. As shown in the Supplementary Fig. 7c, the diffraction peak of the GOM was at 11.19° (d=0.787 nm). This is in good agreement with the result from the TEM image analysis (d=0.760 nm). The diffraction peak for GM900 is at $2\theta=25.05^{\circ}$ (d=0.352 nm), which is also in consistence with the result from the TEM image analysis (d=355 nm).

Supplementary Table 3. Comparison for the photothermic performance of GOM with other reported carbon-based materials.

Materials	Highest temperature after the light irradiation	Time to reach the highest temperature	Note
GO soaked with	400, 500 °C	holow 15 mg	Adv. Mater. 2010, 22,
ethanol	400-300 °C	Delow 15 llis	419-423
Carbon	475 °C	30 ms	Carbon, 2007,
nanotubes			45(5):958-964
GO membranes	670 °C	8.7 ms	Our result
GM900	170 °C	8.7 ms	(Wavelength: 0.8 nm.
Graphite paper	34 °C	20 ms	Power: 0.7 W.
Nano-diamond	124 °C	36 ms	Pulse time: 50 ms)

References

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