Supporting Information

**Construction of highly aligned graphene-based aerogels and their epoxy composites towards high thermal conductivity**

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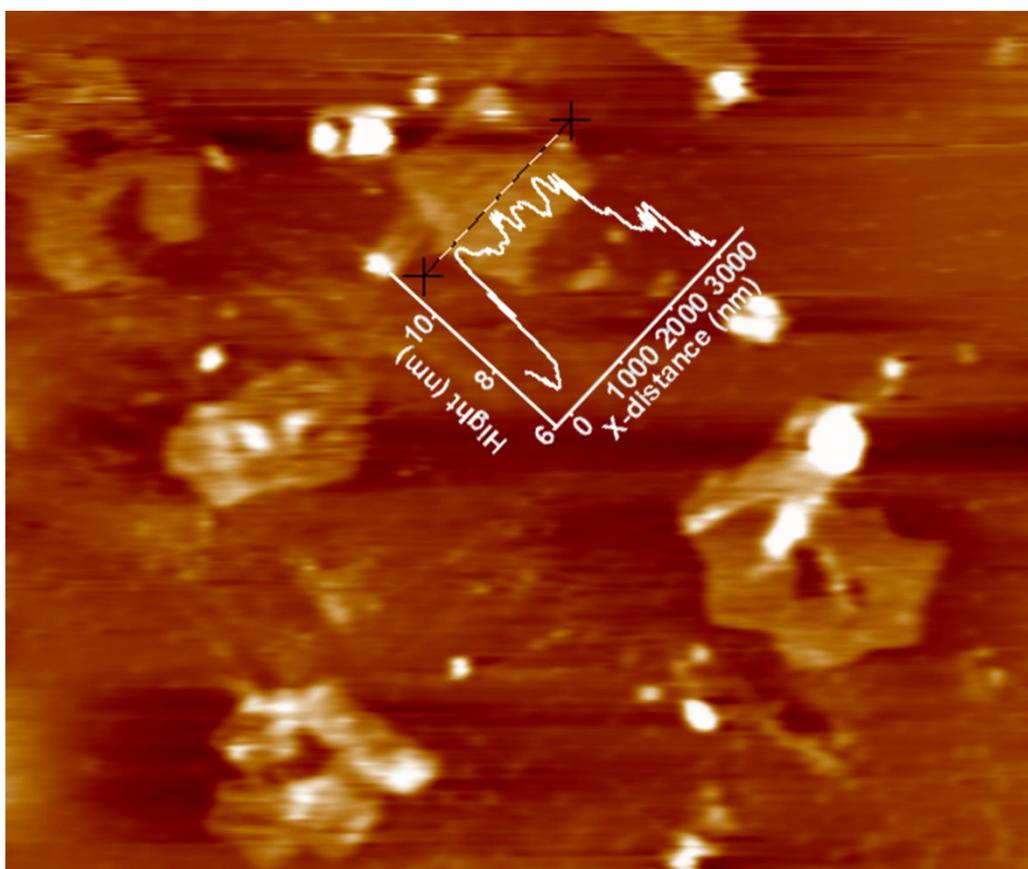

**Fig. S1.** Atomic force microscope image of GO sheets.
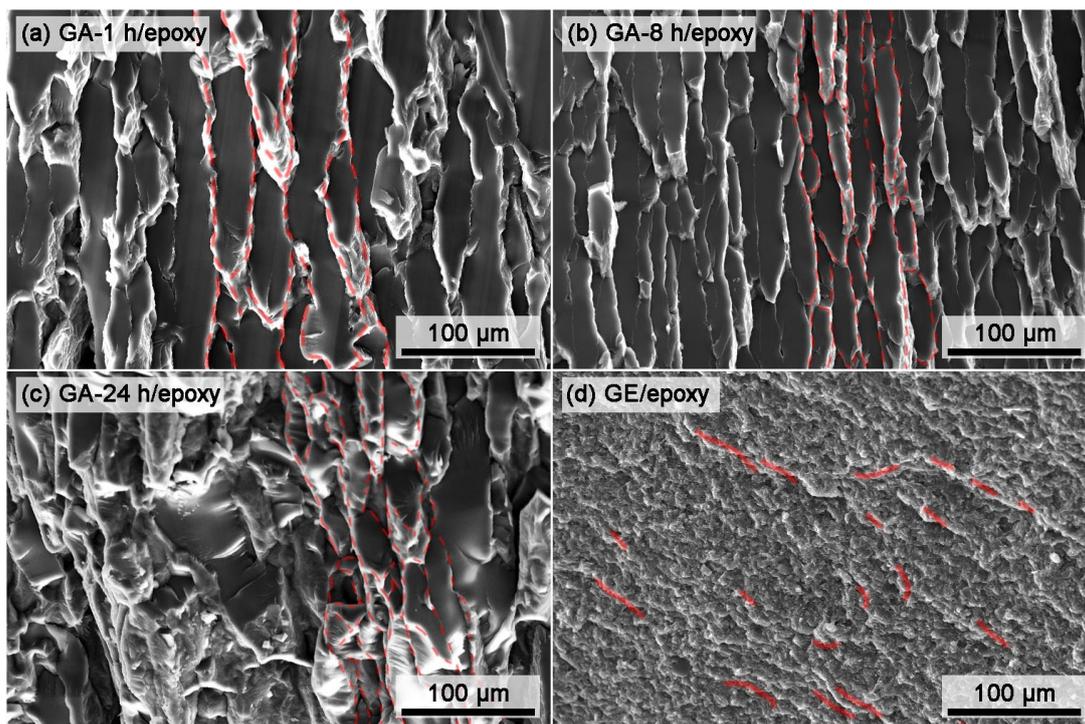
Fig. S2. The thermal conducting channels consisted of GE sheets in the GA/epoxy or GE/epoxy composites. SEM images of the fracture surfaces of epoxy composites with (a) GA-1 h (a), (b) GA-8 h and (c) GA-24 h. The red dash line indicates the location of GE sheets in epoxy.
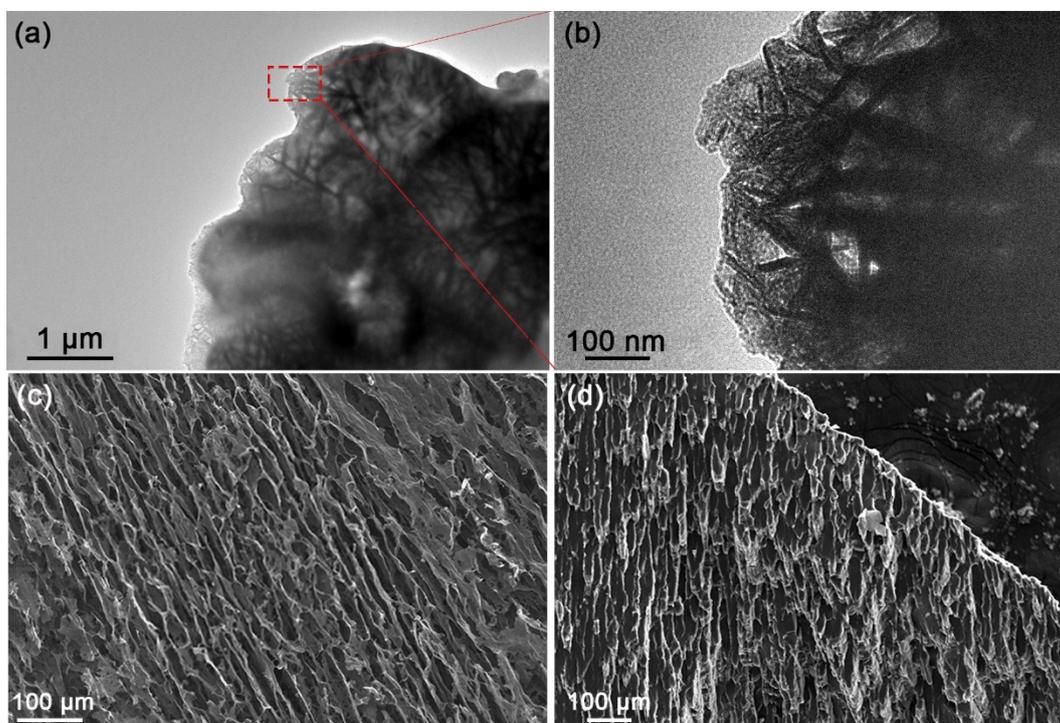

Fig. S3. The microstructures of GA after the infiltration of epoxy. (a) A TEM image of GA-24 h/epoxy, (b) An enlarged view of the red square box in Fig. S3a; (c) A SEM image of GA before be infiltrated with epoxy, (d) A SEM image of GA-24h/epoxy composite.
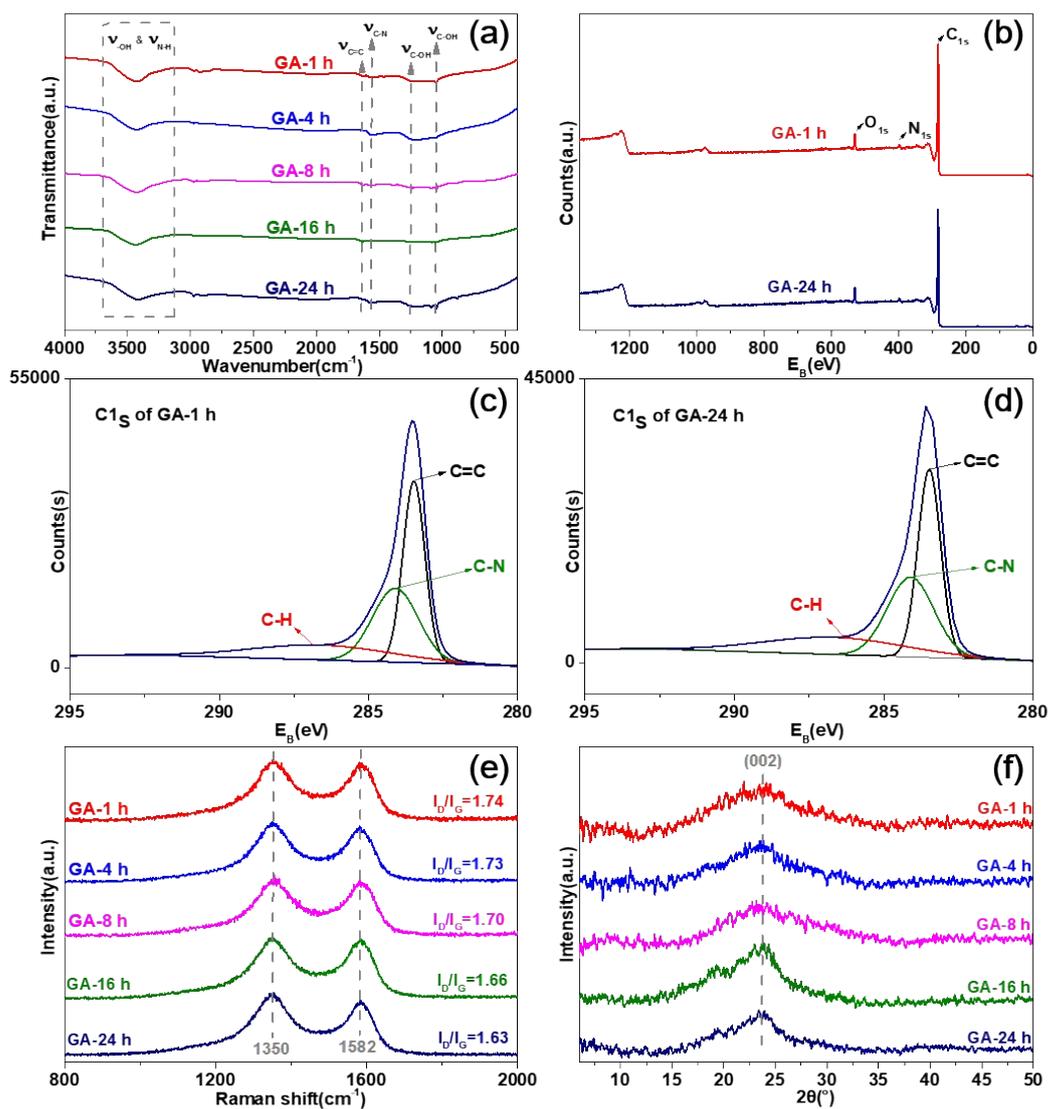
Fig. S4. The microstructures of GAs after the thermal reduction. (a) FT-IR spectra of GO and GAs; (b) XPS general spectra of GA-1 h and GA-24 h. (c-d) C1s deconvoluted spectra of GA-1 h and GA-24 h. (g) Raman spectra and (h) XRD patterns of GO and GAs after thermal reduction.
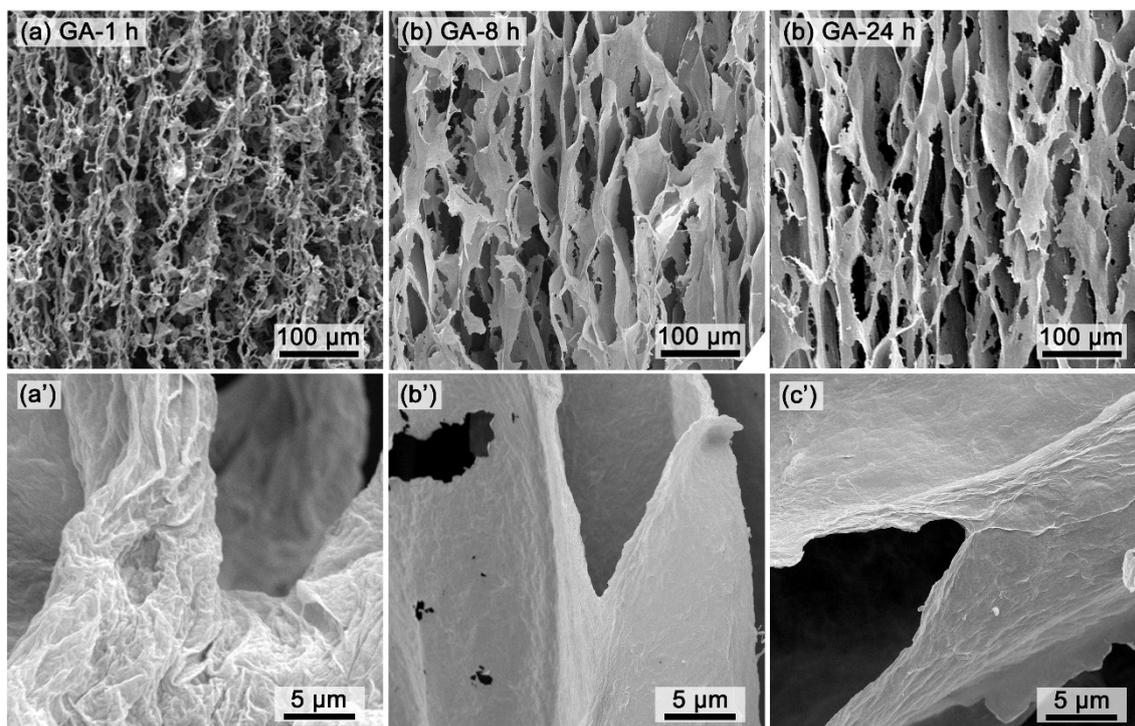
Fig. S5. Morphologies of GAs after thermal reduction. SEM images of the morphologies of (a) GA-1 h, (b) GA-8 h and (c) GA-24 h. SEM images with higher magnifications showing the contact and the thickness of GE sheets in (a') GA-1 h, (b') GA-8 h and (c') GA-24 h, respectively.
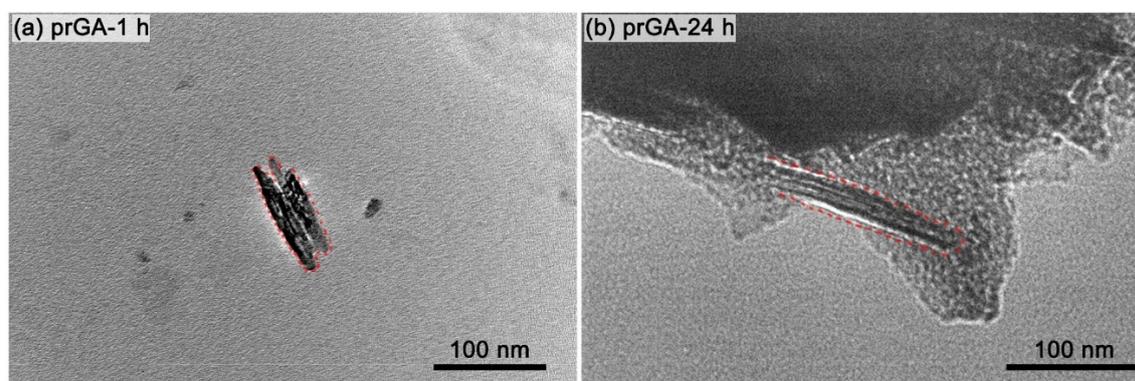

Fig. S6. TEM images of cross-sections of the GE walls in (a) prGA-1 h and (b) prGA-24 h.

Molecular dynamics simulation process

The molecular dynamics of the two-phase system was carried out using the Materials Studio (Version 8.0) suite of programs by Accelrys5. The initial structure of water and GE layers were geometry optimized using the MS DMol3 molecular dynamics module and the simulating system boxes were sketched using MS Amorphous Cell module. Then the boxes were geometry optimized by MS Forcite module (COMPASS II force fields, Atom based summations). Every MD simulation system consisted of 1000 water molecules. These systems were then heated form 0 K to 77 K for 100 ps in NVT ensemble with thermostat of Velocity Scale, pre-equilibrated at 77 K for 100 ps in NPT ensemble with Barostat of Parrinello and equilibrated at 77 K for 100 ps in NVT ensemble with thermostat of Velocity Scale. The interaction parameters for water and GE layers were taken from the COMPASS II forcefield. The energy summation method of electrostatic and van der Waals were both atom based and the van der Waals forces were truncated at 15.5 Å. After simulation, the radical distribution function of GO sheets and water molecules is calculated with cutoff of 20.0 Å and interval of 0.02 Å. Besides, to better distinguish the reducing degree of GO to the interactions with water, the model GO sheets is set with different number of oxygen-containing groups as Fig. S6, and denoted as GO-1 h and GO-24 h.
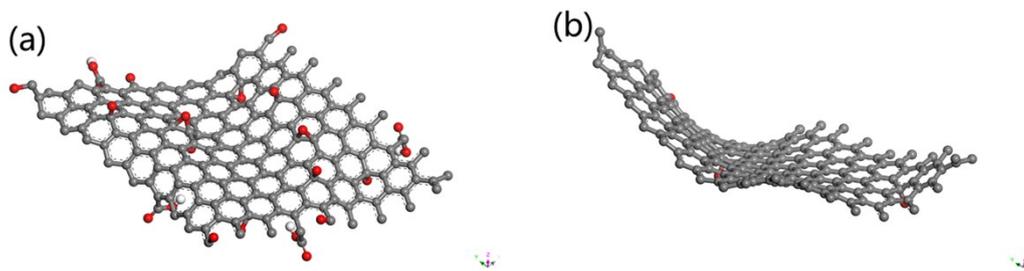
Fig. S7. Models of GE layers in (a) GO-1 h and (b) GO-24 h used in molecular dynamics

simulation.
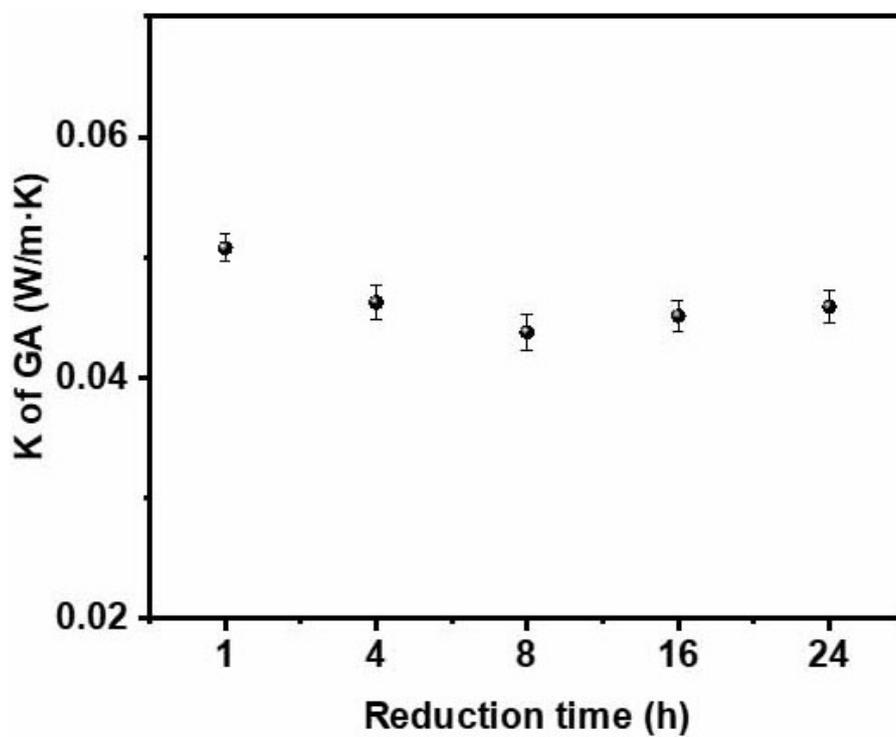
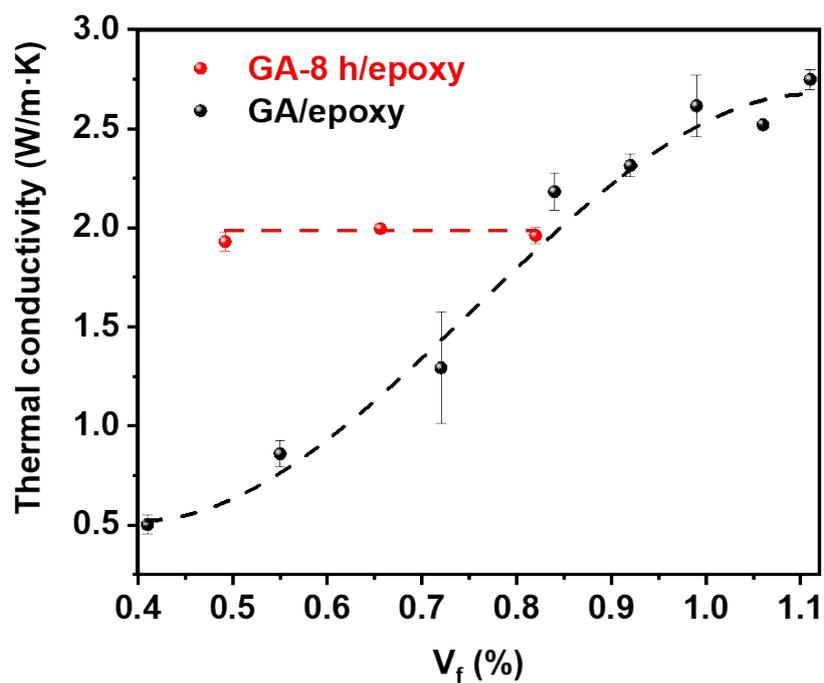Fig. S8. The thermal conductivity of GA reduced for different time.

Fig. S9. The comparation of thermal conductivity among GA-8 h/epoxy with different volume fraction for GA ranging from 0.5-0.8% and GA/epoxy with reduced for different time.
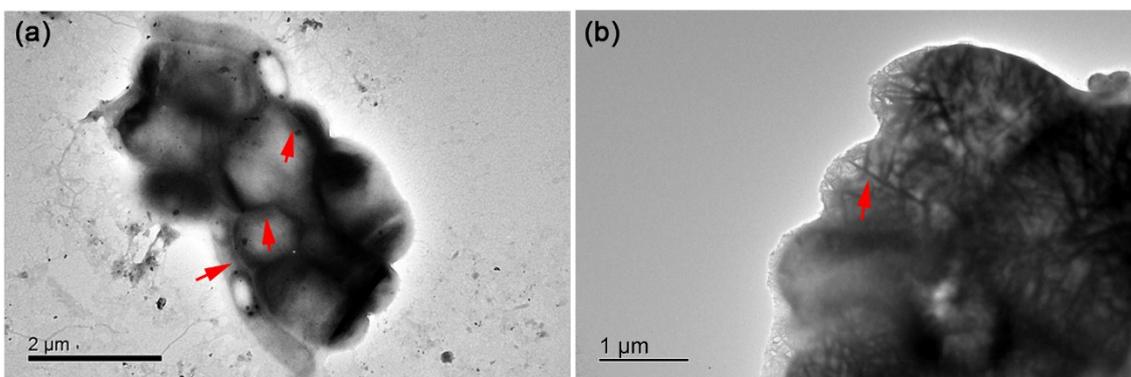

Fig. S10. The TEM images of GA-1 h/epoxy (a) and GA-24 h/epoxy (b). The red arrows are pointed to the GA skeleton.

Appendix

All the fitting processes is conducted on MATLAB R2012a.

Based on the EMI model, the thermal conductivity of GA/epoxy composites can be expressed as the following equation:

$$K = \frac{2 + V_f \dfrac{K_f}{K_m}(1 + \langle cos^2\theta \rangle)}{2 - V_f \dfrac{K_f h - K_m h - R_b K_f K_m}{K_f h}(1 - \langle cos^2\theta \rangle)} K_m$$

The meaning and value is given below:

K: Thermal conductivity of composite (experimental)

K_f: Thermal conductivity of filler (experimental, average 2 W/m·K)

K_m: Thermal conductivity of matrix (experimental, average 0.23 W/m·K)

V_f: Filler loading (experimental)

h: Thickness of filler (experimental)

R_b: Thermal boundary resistance between fillers and matrix (fitted)

Assume:

R_b: $5*10^{-7}$; $5*10^{-8}$; $5*10^{-9}$; $5*10^{-10}$

Variants:

V_f: 0.4-1.1

Code:

```
% This script is applied to analyse the thermal property

close all; clc; clear;

%% Thermal property analysis
```

```matlab
% PARAMETERS

% K:thermal conductivity of composite

% Kf£°thermal conductivity of filler

% Km:Thermal conductivity of matrix

% Vf: filler loading

% h: thickness of filler

% Rbd: thermal boundary resistance between fillers and matrix

Kf=2;

theta=pi/2;

Km=0.2301;

h=8.0478;

Vf=.4:.005:1.1;

Rbd=[1e-10 1e-9 1e-8 1e-7 1e-6];


len=length(Rbd);

fontsize=11;


for i=1:len

K(i,:)=Km*(2+Vf.*(Kf/Km).*(1+(cos(theta)).^2))./(2-Vf.*((Kf*h-Km*h-

Rbd(i)*Kf*Km).*(1-(cos(theta)).^2))/(Kf*h));

% Rbd=(h.*(Kf-Km)-(2*Kf.*h-
```

```
(Kf.*h./K).*(2*Km+Vf.*Kf.*(1+(cos(theta).^2)))))./(Vf.*(1-

cos(theta).^2)))./(Kf.*Km);

end
```

For the nonlinear model:

$$K(V_f, \alpha) = K_0(V_f - V_c(\alpha))^{t(a)}$$

K_o: Preexponential factor ratio, which is the expected contribution of filler skeleton alone

a: The aspect ratio of filler

$V_c(\alpha)$: The critical volume fraction of skeletal filler

$a \gg 1$, $V_c(\alpha) = \dfrac{0.6}{a}$

$a \to 0$, $V_c(\alpha) = 0.29$

$a \to \infty$, another theory

for the average lateral size of GO is around 2500 nm and the thickness is about 8 nm, we konw the aspect ratio a is much higher than 1, thus $V_c(\alpha)$ can be calculated by 0.6/α

$t(\alpha)$: A conductivity exponent depending on the aspect ratio of fillers

$\alpha \leq 1$, $t(\alpha) = 2$

$10^2 \leq \alpha \leq 10^3$, $1.2 \leq t(\alpha) \leq 1.6$

for $10^2 \leq \alpha \leq 10^3$, we set $t(\alpha)$ to be a value in the range of 1.2-1.6

code:

```
% This script is applied to analyse the thermal property

close all; clc; clear;

%% Thermal property analysis

% PARAMETERS
```

```matlab
% K:thermal conductivity of composite

% Kf£°thermal conductivity of filler

% Km:Thermal conductivity of matrix

% Vf: filler loading

% h: thickness of filler

% Rbd: thermal boundary resistance between fillers and matrix

Vf=.4:.005:1.15;

Ko=[1 2 2.5 2.75 3]

Vc=0.00189;


len=length(Ko);

fontsize=11;


for i=1:len

K(i,:)=Ko(i)*((Vf-Vc).^1.2)

%K(i,:)=Km*(2+Vf.*(Kf/Km).*(1+(cos(theta)).^2))./(2-Vf.*((Kf*h-Km*h-

Rbd(i)*Kf*Km).*(1-(cos(theta)).^2))/(Kf*h));

% Rbd=(h.*(Kf-Km)-(2*Kf.*h-

(Kf.*h./K).*(2*Km+Vf.*Kf.*(1+(cos(theta).^2)))))./(Vf.*(1-

cos(theta).^2)))./(Kf.*Km);

end
```