

Supporting Information for

The effect of doping graphene oxide on the structure and property of polyimide-based graphite fibre.

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1. Analysis data for Fig.3 in the paper

Table. S1 The 5% weigh loss temperature(Td5) and 10% weight loss temperature (Td10) of PI fibre and GO /PI composite fibre in TG curve.

sample	Pure PI	0.3wt%	0.5wt%	1.0wt%
T _{d5} /°C	459	474	478	497
T _{d10} /°C	469	489	492	509

2. Analysis data for Fig.4 in the paper

Table. S2 Peak temperature and intensity of exothermic during carbonization process in oxygen atmosphere in DSC curve.

Sample	Pure PI	0.3wt%	0.5wt%	1.0wt%
Exothermic peak temperature of / °C	503	530	531	539
Exothermic peak intensity of / (mW/mg)	-92.44	-24.11	-22.67	-28.01

3. FT-IR for GO/PAA fibres

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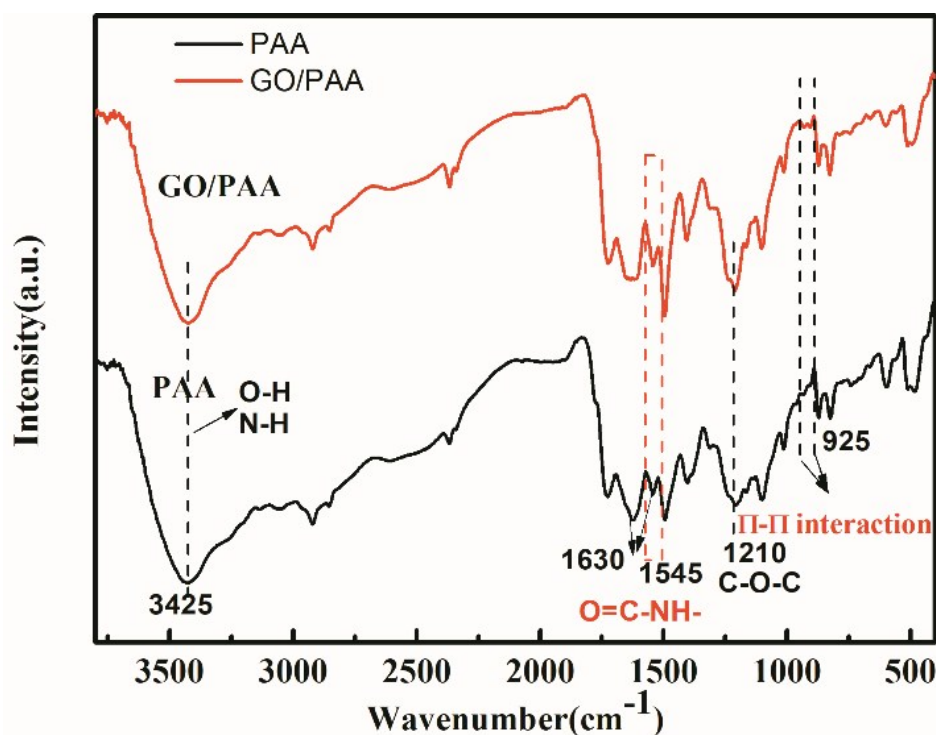


Fig. S1 FT-IR spectrum of PAA and GO/PAA fibres.

Fig.S1 displays the FT-IR spectrum of PAA and GO/PAA fibres. The broad peak around 3425cm⁻¹ is the stretching vibration of carboxyl group on the molecular chains and amino groups at the end of molecular chains. The two adjacent peaks at 1545cm⁻¹ and 1630cm⁻¹ indicate the existence of the amide group on the molecular chains. The sharp peak at 1210cm⁻¹ indicate the existence of ether group. The intensity of the peak around 925cm⁻¹ was strengthened after GO was added in to the PAA fibres, which indicate that PI molecular chains were attached to GO sheets due to π - π interaction.

4. TG-DSC curves for GO/PAA fibres

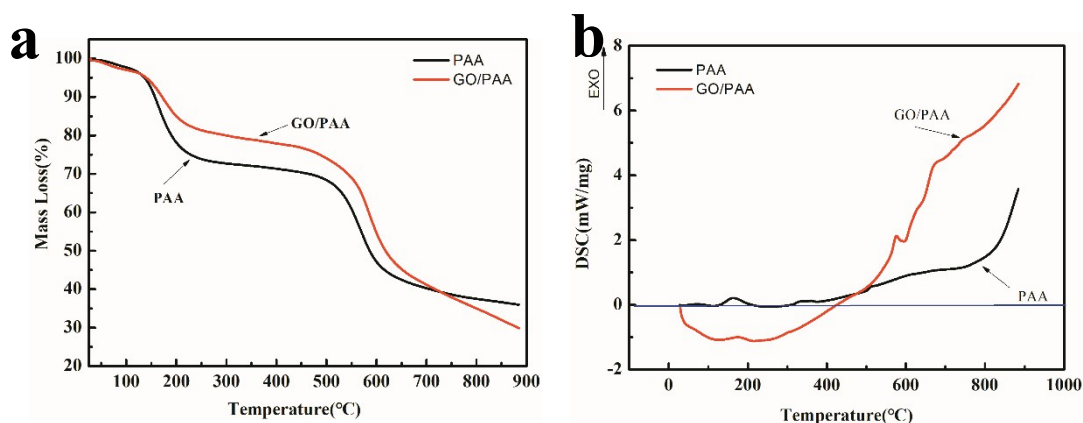


Fig. S2 TG (a) and DSC (b) curves of PAA fibres and GO/PAA composite fibres.

TG-DSC is used to analyze the thermal stability of PAA fibres and GO/PAA composite fibres. As shown in Fig.S2(a), the PAA fibres and GO/PAA fibres all experienced a similar weight loss strategy. From room temperature to 300°C, PAA fibres were transformed into PI fibres with the generation and removal of H₂O. The residual mass and the pyrolysis starting temperature of GO/PAA fibres is higher than that of pure PAA fibres, which indicate that GO/PAA fibres have a better thermal stability than PAA fibres. Due to the structural instability of GO, the carbon yield of GO/PAA fibres is lower than that of PI fibres.

Fig.S2(b) shows the endothermic and exothermic data of PAA fibres and GO/PAA fibres. The GO/PAA fibres have a endothermic peak from room temperature to 500°C, indicating a reaction between GO and PAA molecular which increase the thermal stability of the composite fibre.

5. TG-DTA curves for GO/PI composite fibres

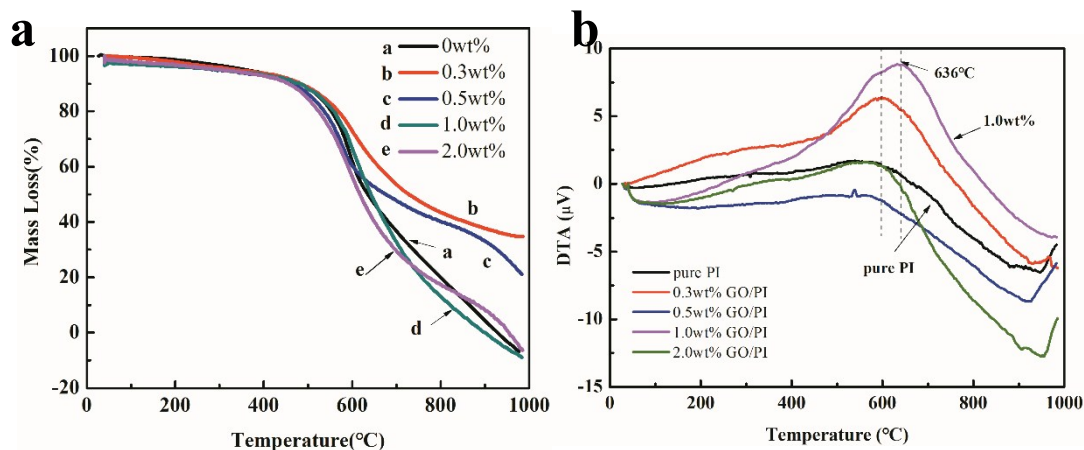


Fig. S3 TG (a) and DSC (b) curves of GO/PI composite fibres.

The same result can be confirmed by TG/DTA analysis in pure-nitrogen atmosphere of GO/PI composite fibre with different GO contents. In Fig. S3(a), the pyrolysis starting temperature and carbon yield of GO/PI composite fibre with 0.3wt% GO content are the highest, which indicate the better thermal stability of the fibre. The improved thermal stability of GO/PI composite fibres may have two reasons: firstly, GO sheets may have a strong reaction with PAA molecular thus increase the thermal stability; secondly, GO sheets may hinder the movement of PAA molecular chains thus increase the energy to break the molecular chains.

As shown in Fig. S3(b), the pyrolysis starting temperature of GO/PAA composite fibre with 1.0wt% GO content is 636°C, which is 76°C higher than that of pure PI fibres. The result of TG-DSC confirmed that adding GO increased the thermal stability of GO/PAA composite fibres.

6. XRD curves for GO/PI composite fibres

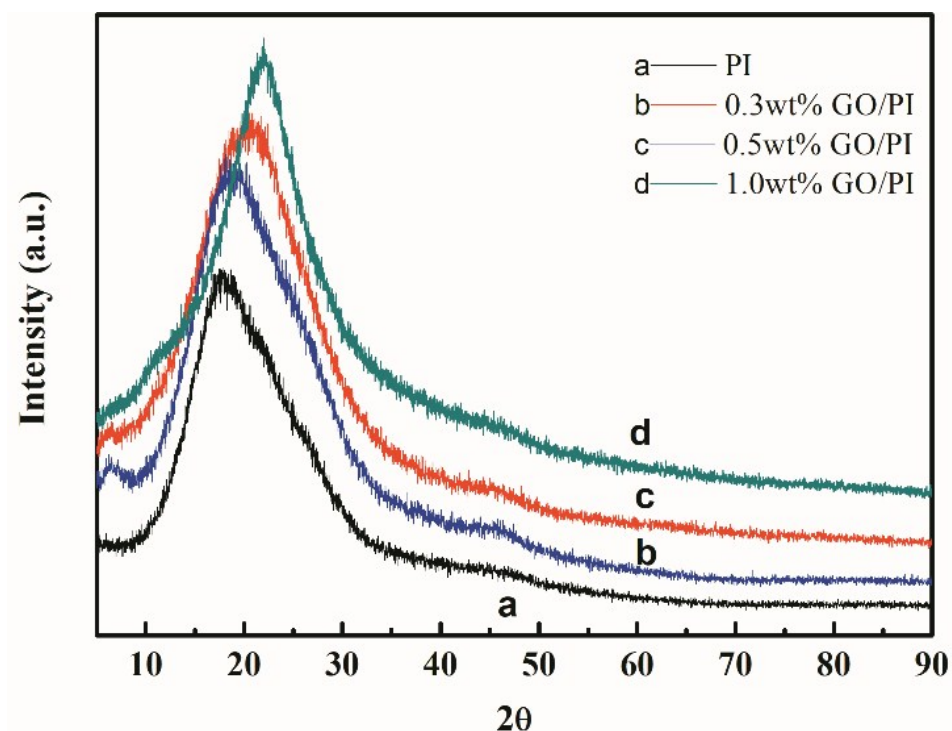


Fig. S4 XRD spectrum of GO/PI composite fibres with different GO contents.

XRD was used to study the crystal structure of the GO/PI composite fibre. As shown in Fig. S4, the crystal structure of GO/PI fibre was amorphous semi-crystalline because the molecular chains of GO/PI composite fibre are too rigid to crystallize. It was obvious to see that the diffraction peak moved to higher angle with the increase of GO contents. The diffraction peak of pure PI is at $2\theta=18.11^\circ$, while the diffraction peak with 1.0wt% GO content was $2\theta=21.95^\circ$ which shifted 3.84° to high angle. According to the Bragg equation $2d \sin \theta = n\lambda$, it could be indicated that the interplanar spacing of PI crystal gradually decreases with the increase of GO contents. The diffraction peak becomes sharper and has a trend to continue to crystallize with the increase of GO contents. It could be inferred that GO can induce the crystallization of composite fibre and improve the orientation of composite fibre.

7. Pyrolysis strategy of graphite fibres

Table. S3 The element composition of composite carbon fibre with 0.3wt%GO content at different HTT.

C/%	N/%	C/%	O/%	N/C/%	O/C/%
800	4.66	78.77	14.50	5.92	18.40
1000	3.53	83.39	11.14	4.23	13.36
1200	2.53	89.61	6.49	2.82	7.25
1500	0.41	98.40	0.81	0.42	0.83
2800	0.12	99.01	0.55	0.12	0.56

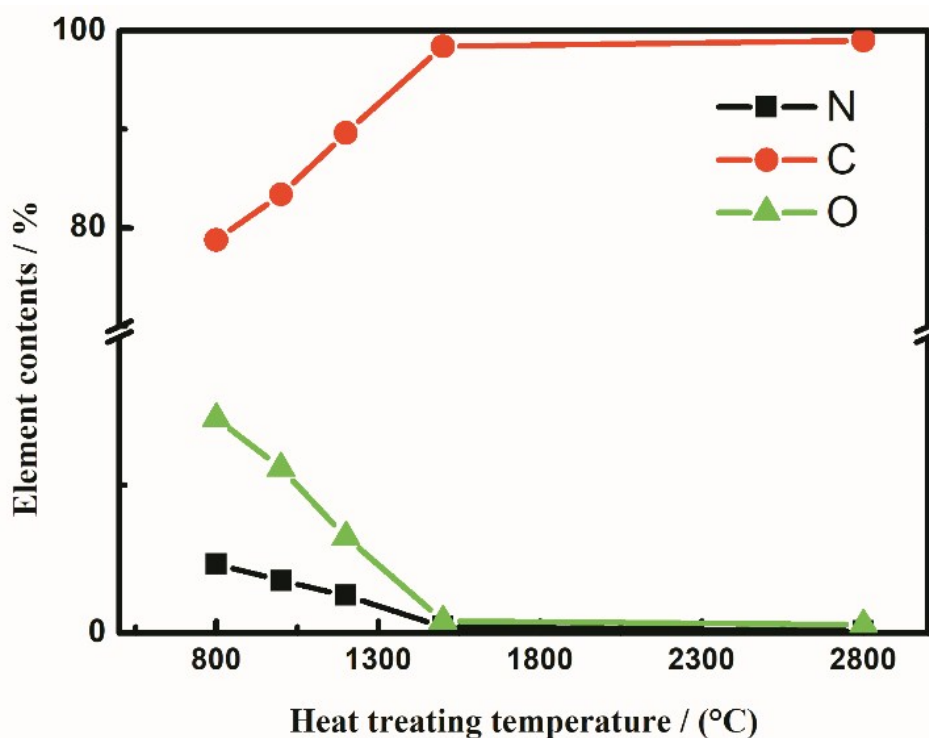


Fig. S5 The element composition of composite graphite fibre with 0.3wt% GO content at different HTT.

Fig. S5 shows the change of element contents of GO/PI based carbon fibre with 0.3wt% GO at different HTT. It can be confirmed in Fig. S5 and Table. S3, the content of C is increasing while the content of O and N are decreasing with the increase of heat treating temperature(HTT). It can be inferred that the chemical bonding of C-H and C-N will break down during carbonization and give out the small molecular like N_2 , CO and CO_2 . During graphitization process, the element of N were removed from the crystal structure of graphite fibres, which increased the degree of graphitization.

8. Tensile modulus of graphite fiber with different content of GO.

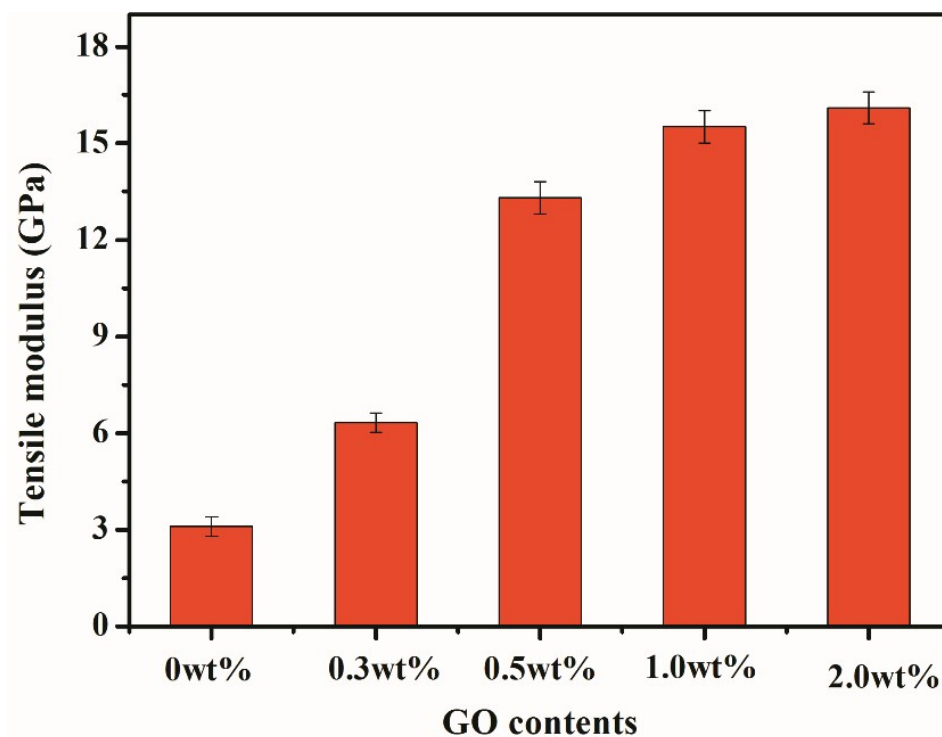


Fig. S6 Tensile modulus of graphite fiber with different content of GO.

As is shown in Fig. S6, the tensile modulus of graphite fiber is enhanced with the increasing of GO.

9. Thermal conductivity at different HTT

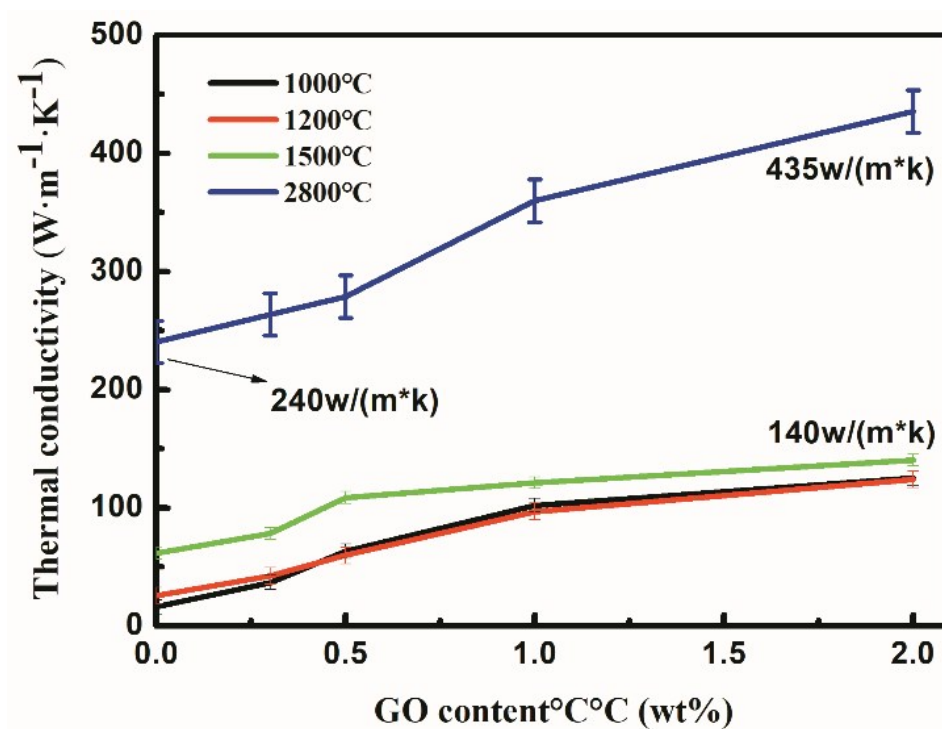


Fig. S7 Thermal conductivity of GO/PI based carbon fibres and graphite fibres with different GO content.

The thermal conductivity of carbon fibres and graphite fibres increases as the heat treating temperature grows. The electrical resistance and thermal conductivity of mesophase carbon fibre has an empirical equation, which is $\lambda = 1261/\rho$. Using electronic meter to measure the electrical resistance, then calculate the thermal conductivity of the GO/PI based graphite fibre.

As shown in Fig. S7, it is obvious that the thermal conductivity of composite graphite fibre increases as GO content grows. At 2800°C, the thermal conductivity of GO/PI based graphite fibre with 2.0wt% GO adding is almost twice of that of pure PI based graphite fibre. The increase of heat treating temperature can increase the thermal conductivity also. The adding of GO can induce the crystallization, increase the size of microcrystals, improve the orientation and change the structure into an orderly manner, thus can increase the thermal conductivity of composite graphite fibre.

10. TEM of the cross-section and longitudinal-section of graphite fiber with 2.0wt% GO content.

Frozen ultra-thin slicing method was used to slice the fiber into pieces with a thickness of 50nm to show the cross section and longitudinal section of the composite graphite fiber. Then, we use TEM to investigate the nanostructure of 2.0wt% GO/PI composite fiber.

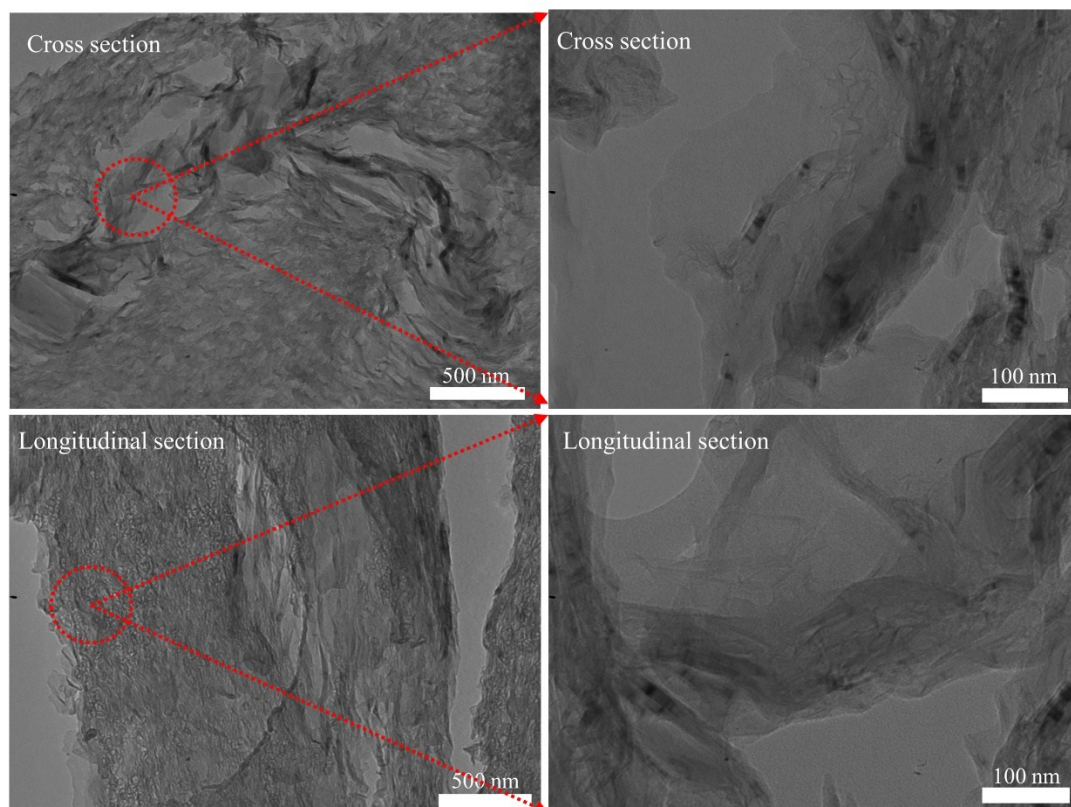


Fig. S8 TEM of the cross-section and longitudinal-section of graphite fiber with 2.0wt% GO content.

From the cross-section TEM graph of graphite fiber, we can see the formation of graphite crystallites. The darker part of the cross-section indicate graphite crystallite induced by rGO. The graphite crystallite induced by rGO is apparently larger than that of other part, which indicate GO doping induced the formation and growing of graphite crystallite.