

Supporting Information

Facial Fabrication of Few-Layer Functionalized Graphene with Sole Functional Group Through Diels–Alder Reaction by Ball Milling

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S1 Morphologies and Structures of ball-milled graphite (G-BM) without any additives

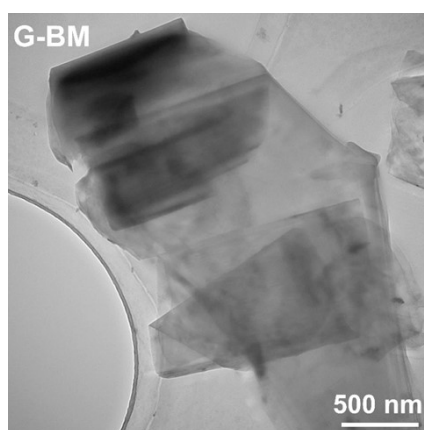


Fig. S1 Transmission electron microscopy (TEM) image of G-BM.

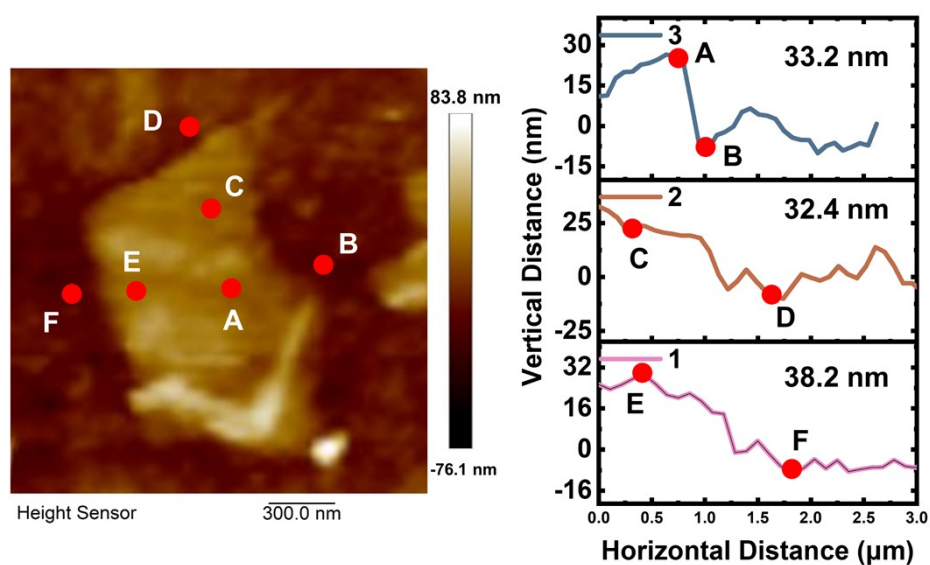


Fig. S2 Atomic force microscopy (AFM) tapping-mode image of G-BM.

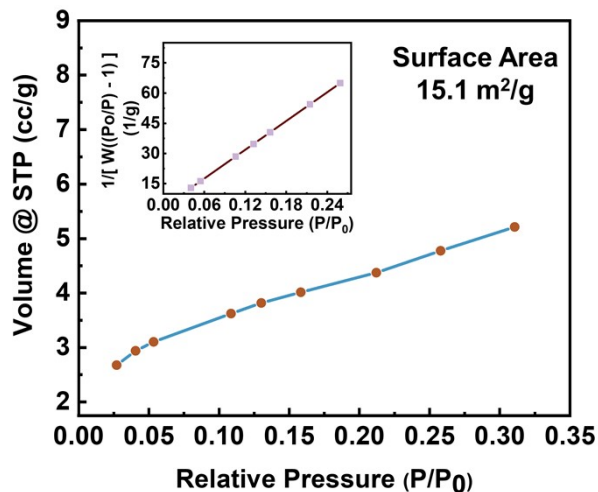


Fig. S3 Nitrogen adsorption/desorption isotherm of G-BM.

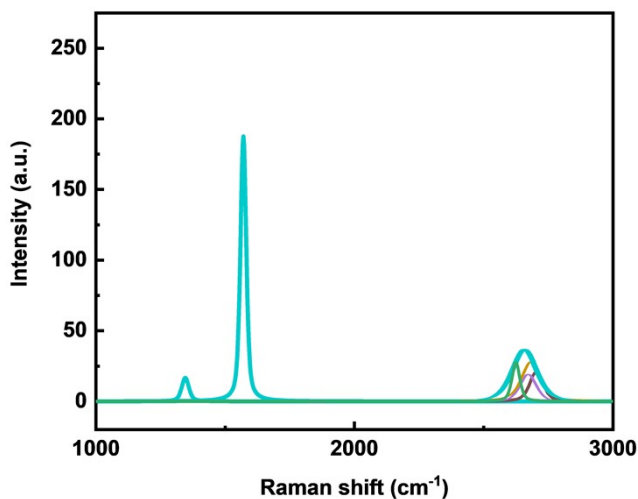


Fig. S4 Raman spectra of G-BM.

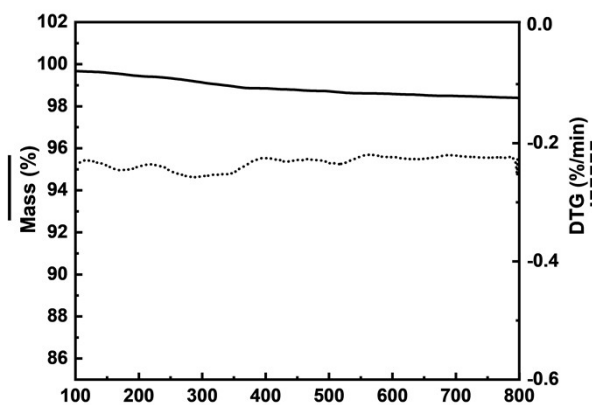


Fig. S5 Thermogravimetric analysis (TGA) curve of G-BM.

S2 Dispersion of graphite and functionalized graphene in polyethylene terephthalate (PET)

The dispersibility of nanomaterials such as graphene in the polymer matrix affects the performance of polymer composites. The better the dispersion of graphene in the polymer matrix, the better the compatibility

and the higher the physical and mechanical properties of the composite material will be. The dispersion of graphite/functionalized graphene in the PET composite fiber was observed through SEM (Fig. S6). As shown in the Fig. S6, the surface of pure PET fiber is smooth. After 0.2 wt% graphite was added, a large number of protrusions were produced on the surface of the fiber. When the same loading of functionalized graphene was added, the surface smoothness was significantly improved, and the convex particles became smaller, indicating that the degree of agglomeration of the functionalized graphene was reduced. Among several composite fibers, PET/GFAL-BM composite fibers had the best surface smoothness, indicating that GFAL-BM had the best dispersion and compatibility in the PET matrix, which means composite fiber could have the best mechanical properties. When 2 wt% of GFAL-BM was added, the surface roughness of the fiber was significantly increased, showing a dense agglomeration of particles. The agglomeration of graphite flakes would cause local stress concentration of the composite fiber, thereby reducing the tensile strength of the composite fiber.

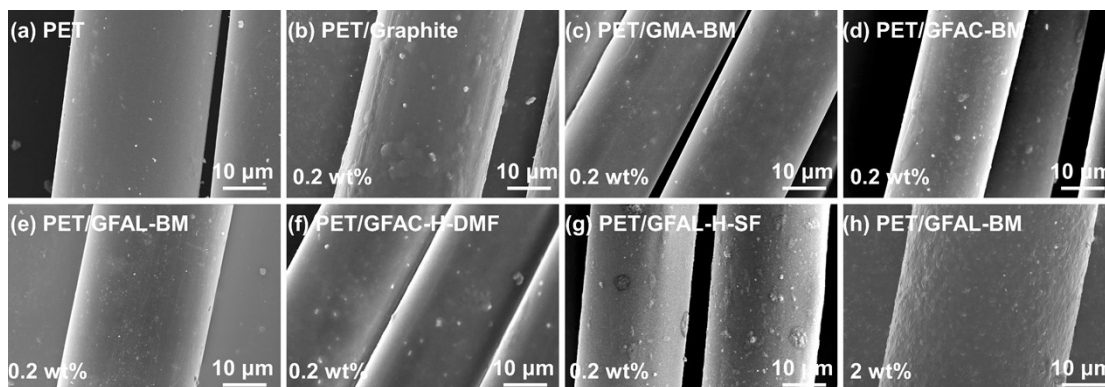


Fig. S6 SEM images of the surface of PET composites fibers.

S3 Effect of PET/GFAL-BM composite fiber on the crystalline properties

It could be judged from the SEM images that the dispersion effect of PET/GFAL-BM in PET was the best. Therefore, in this section, the melting and crystallization behavior of PET/GFAL-BM composite fiber was analyzed and characterized. DSC melting and cooling curves and test data are shown in Fig. S7 and Table S1, respectively. Among them, the crystallinity (X_c) was calculated²⁰. The results showed that the melting temperature (T_m) of the fiber decreases slightly with the addition of graphite (0.2 wt%). This was due to the lack of functionalization, the lamellae were easy to agglomerate, which increased the local heat transfer rate of the fiber, thereby promoting the accumulation of heat required for melting of PET at a relatively low temperature. The melting temperature of the composite fiber with the same loading of GFAL-BM was the highest, indicating that GFAL-BM might be more conducive to the crystallization of PET.

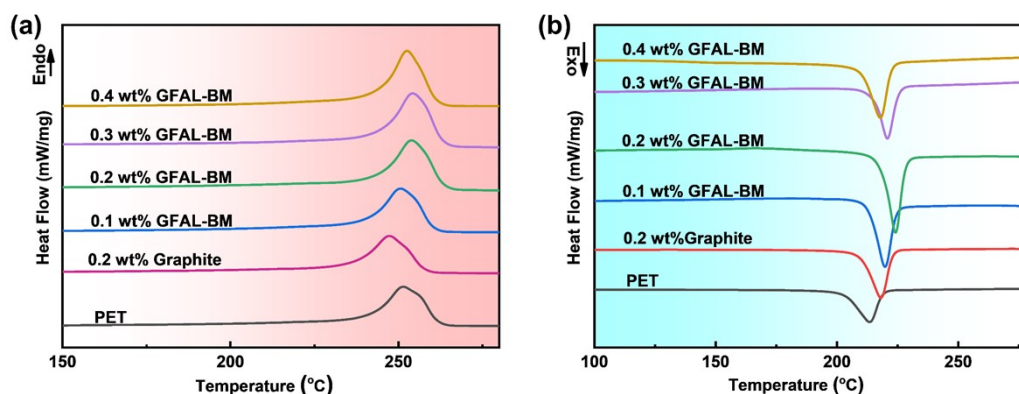


Fig. S7 DSC curves of the (a) melting and (b) cooling behavior in PET and PET composites fibers

When the loading of GFAL-BM exceeded 0.2 wt%, the melting temperature of the fiber decreased. The high concentration of GFAL-BM was prone to agglomeration and stacking in the PET matrix. After adding graphite or GFAL-BM, the crystallinity was improved, indicating that GFAL-BM could be an effective heterogeneous nucleating agent to promote crystallization. Using 0.2 wt% of GFAL-BM could obtain the highest degree of

crystallinity (35.3%) of the fiber, indicating that 0.2 wt% of GFAL-BM was more conducive to the crystallization of PET. The degree of supercooling (ΔT) of GFAL-BM fiber was the lowest (29.4°C), and the crystallization temperature (T_c) was the highest (224.4°C), indicating that the crystallization rate was accelerated. When an excessive amount of GFAL-BM (>0.2 wt%) was added, the sheets would re-stack and hinder the movement of the PET molecular chain, which was not conducive to the growth of crystals, thereby reducing the crystallinity of the fiber. Since crystallinity is a key factor affecting the mechanical properties of composite fibers, it could be inferred from the crystallinity of the fiber that the mechanical properties of GFAL-BM composite fibers should be the best with loading of 0.2 wt%.

Table S1. DSC data of PET and their composite fibers

Fibers	ΔH_m (J/g)	T_m (°C)	T_c (°C)	ΔT (°C)	X_c (%)
PET	34.3	251.5	213.6	37.9	24.5
0.2 wt% Graphite	37.6	247.4	218.1	29.3	26.9
0.1 wt% GFAL-BM	39.8	250.6	220.3	30.3	28.4
0.2 wt% GFAL-BM	49.4	253.8	224.4	29.4	35.3
0.3 wt% GFAL-BM	46.5	253.4	220.7	32.7	33.2
0.4 wt% GFAL-BM	44.8	252.7	218.4	34.3	32.0