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Supporting Information

Enhanced Multifunctional Performance of Flash Graphene-Polymer Composites via Nitrogen Doping

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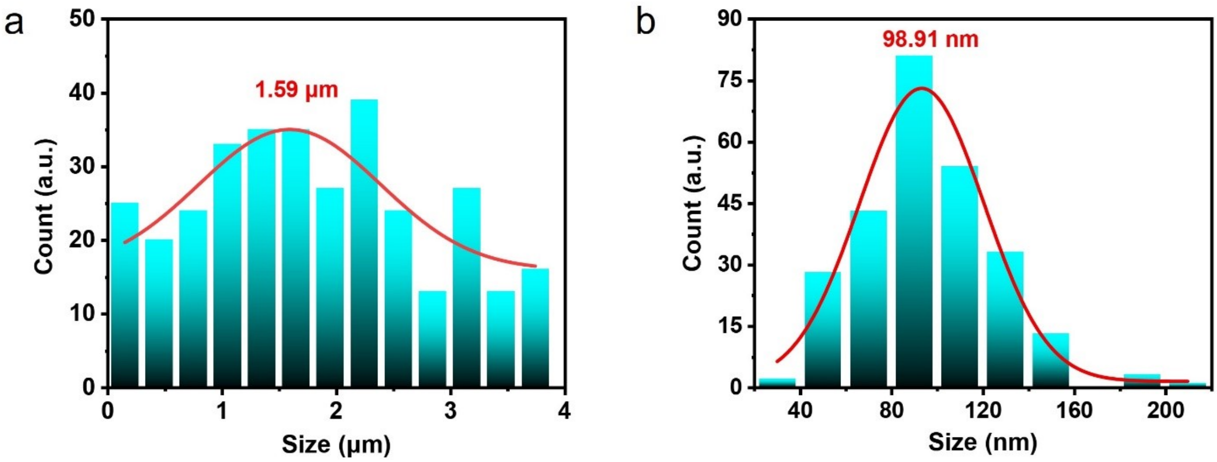

Fig. S1 (a) Size distribution of FG. (b) Size distribution of N-FG.

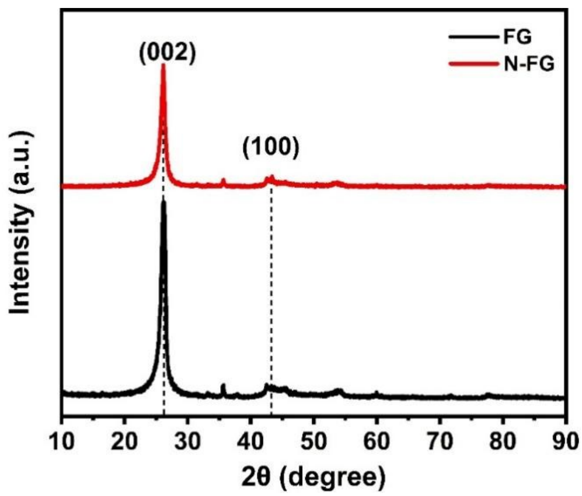

Fig. S2 XRD spectra of FG and N-FG.

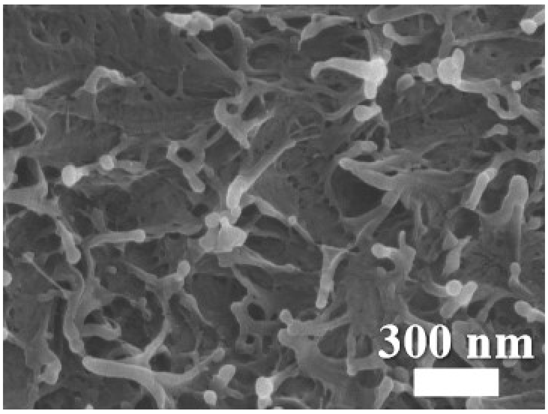

Fig. S3 Cross-sectional SEM image of 1 wt% FG/HDPE.

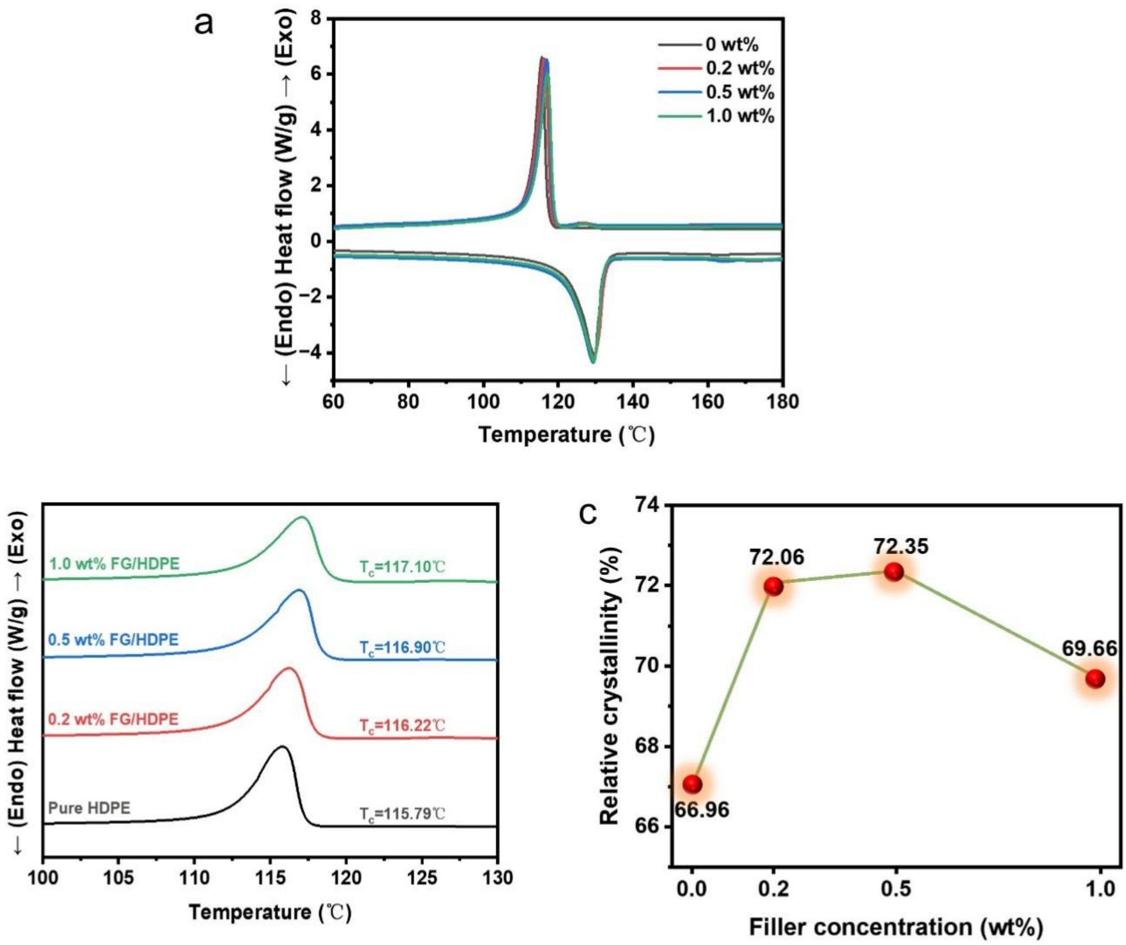

Fig. S4 (a) DSC curves of FG/HDPE. (b) Crystallization peak of FG/HDPE. (c) Comparison of the crystallinity of FG/HDPE.

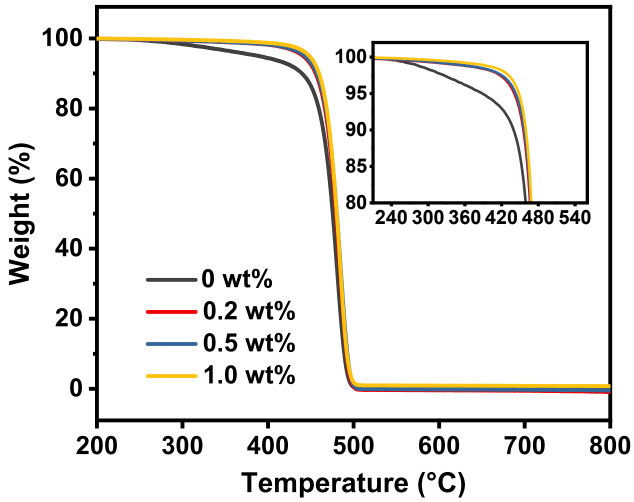

Fig. S5 TGA curves of FG/HDPE.

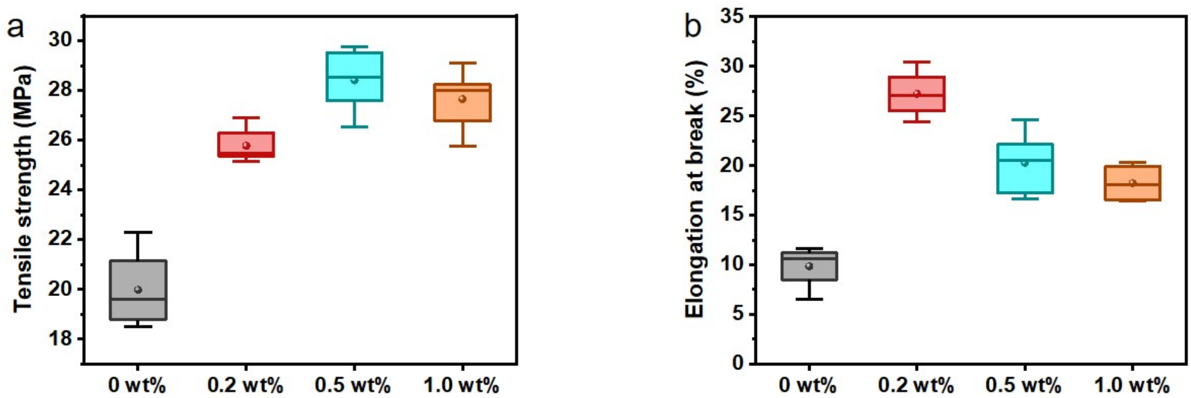

Fig. S6 Mechanical properties of FG/HDPE composites. (a) Tensile strength. (b) Elongation at break.

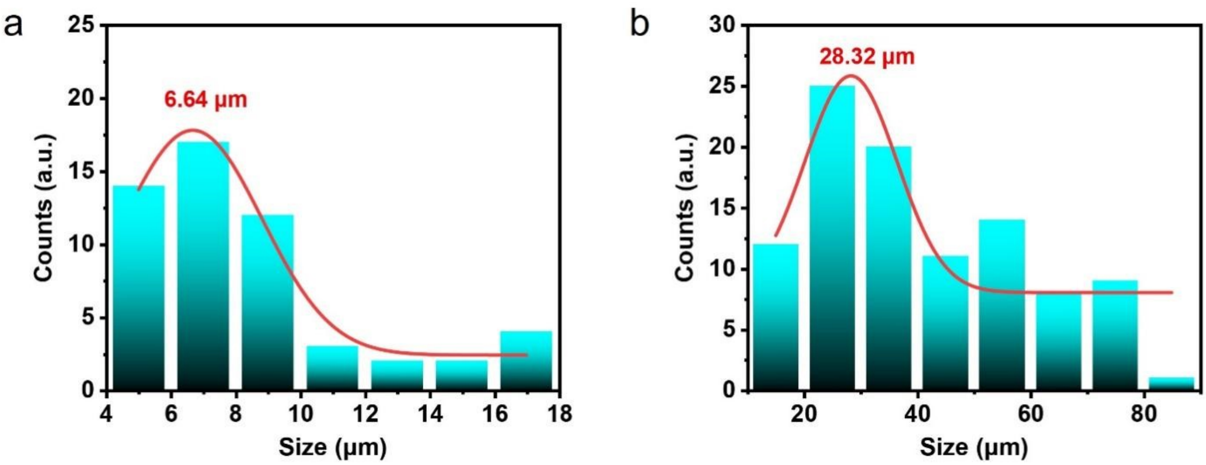

Fig. S7 (a) Size distribution of MMT. (b) Size distribution of GO.

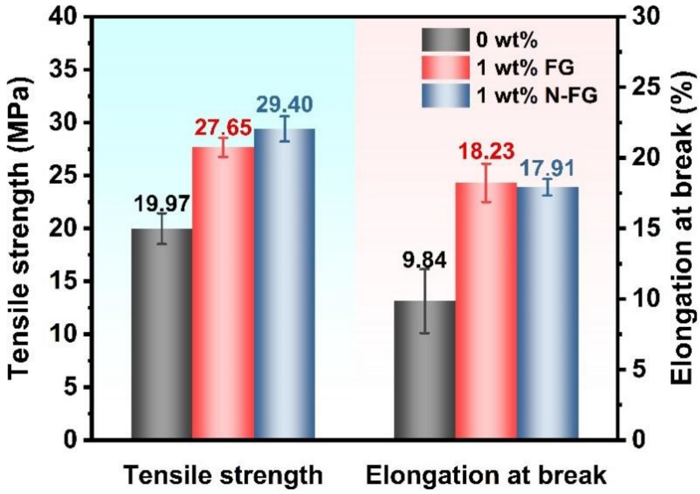

Fig. S8 Mechanical properties of FG and N-FG/HDPE composite materials.

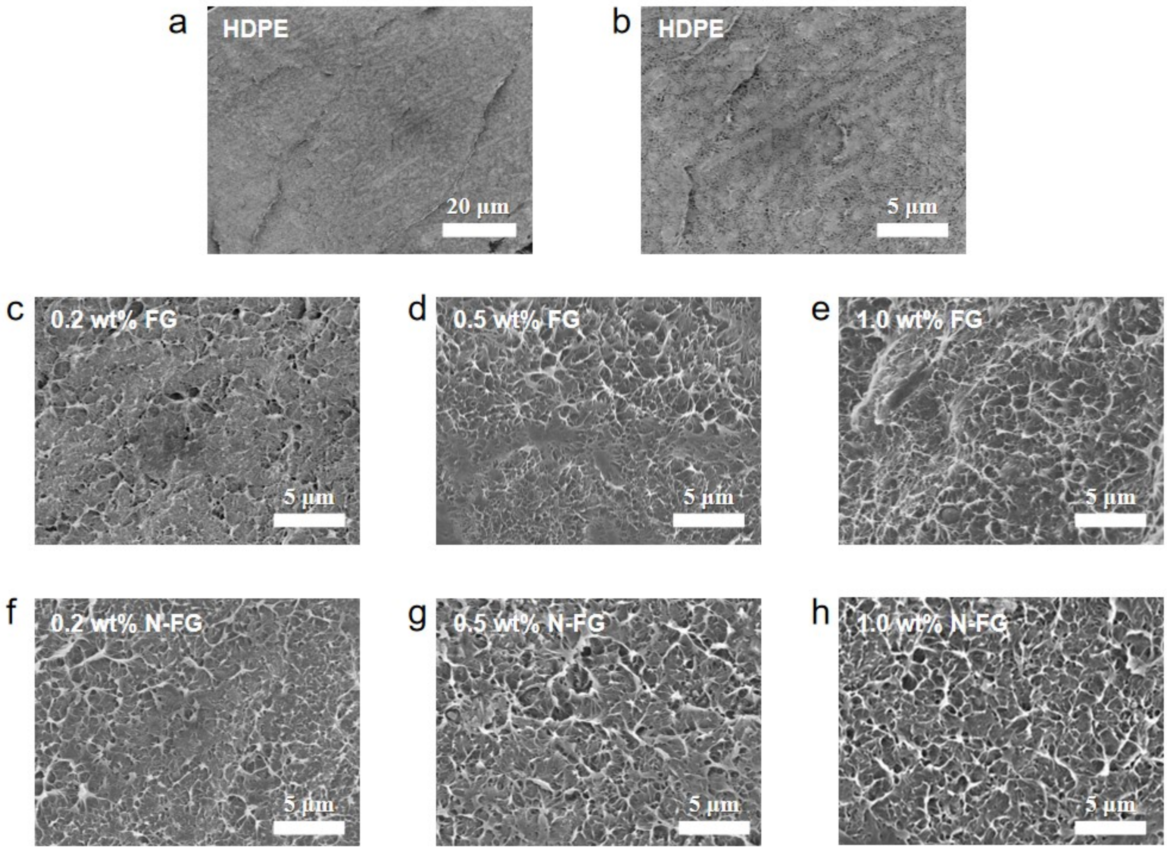

Fig. S9 Cross-sectional SEM images of HDPE composites at different concentrations of FG and N-FG. (a) HDPE. (b) HDPE (Magnified images). (c) 0.2 wt% FG/HDPE. (d) 0.5 wt% FG/HDPE. (e) 1.0 wt% FG/HDPE. (f) 0.2 wt% N-FG/HDPE. (g) 0.5 wt% N-FG/HDPE. (h) 1.0 wt% N-FG/HDPE.

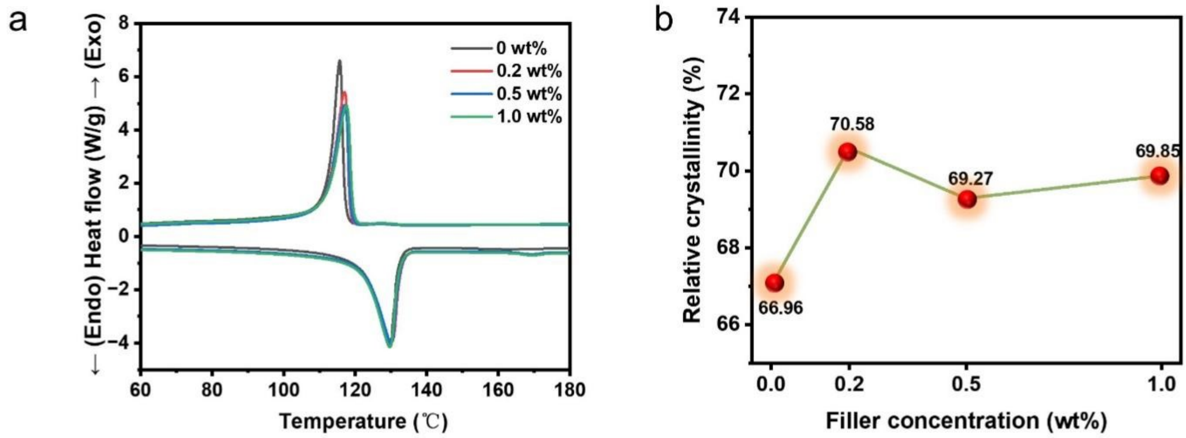

Fig. S10 (a) DSC curve of N-FG/HDPE composites. (b) Crystallinity of N-FG/HDPE composites at different N-FG loadings.

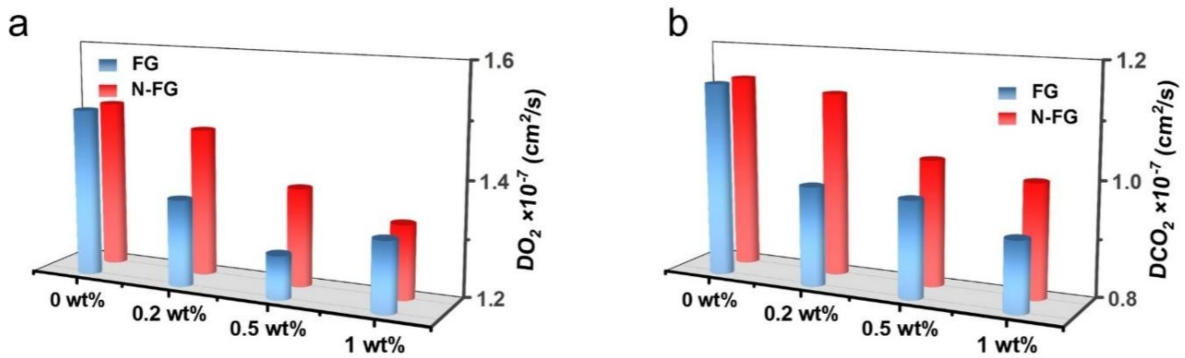

Fig. S11 Gas diffusion coefficient of FG/HDPE and N-FG/HDPE composite films. (a) Oxygen. (b) Carbon dioxide.

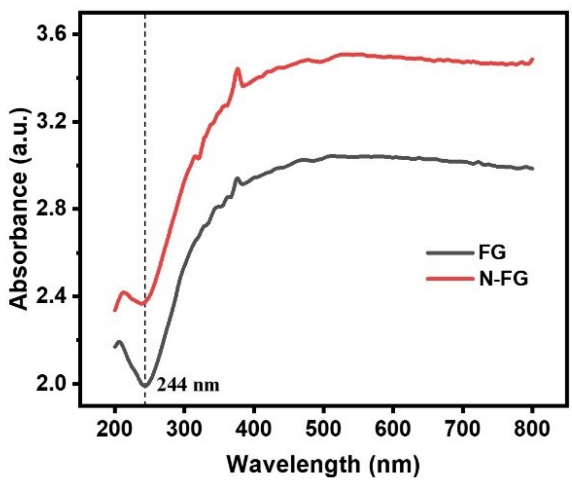

Fig. S12 UV-Vis spectra of FG and N-FG.

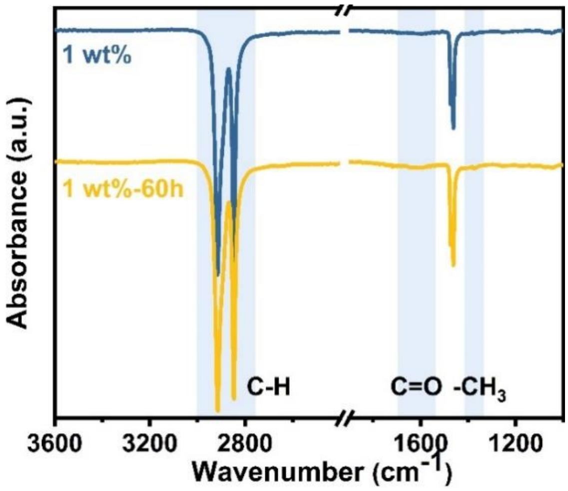

Fig. S13 FTIR spectra of 1 wt% N-FG/HDPE before and after aging.

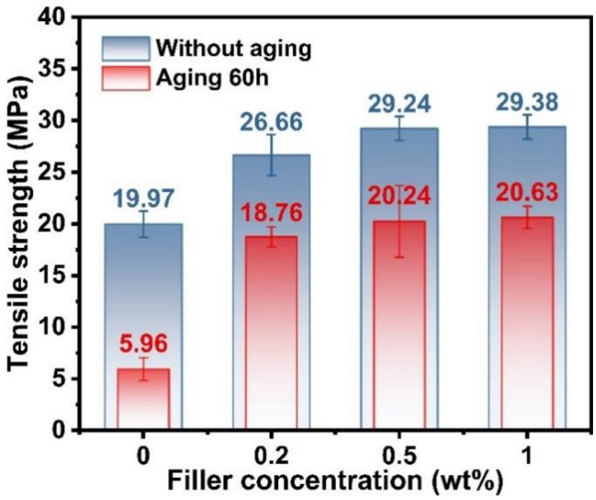

Fig. S14 Comparison of tensile strength of N-FG/HDPE before and after aging.

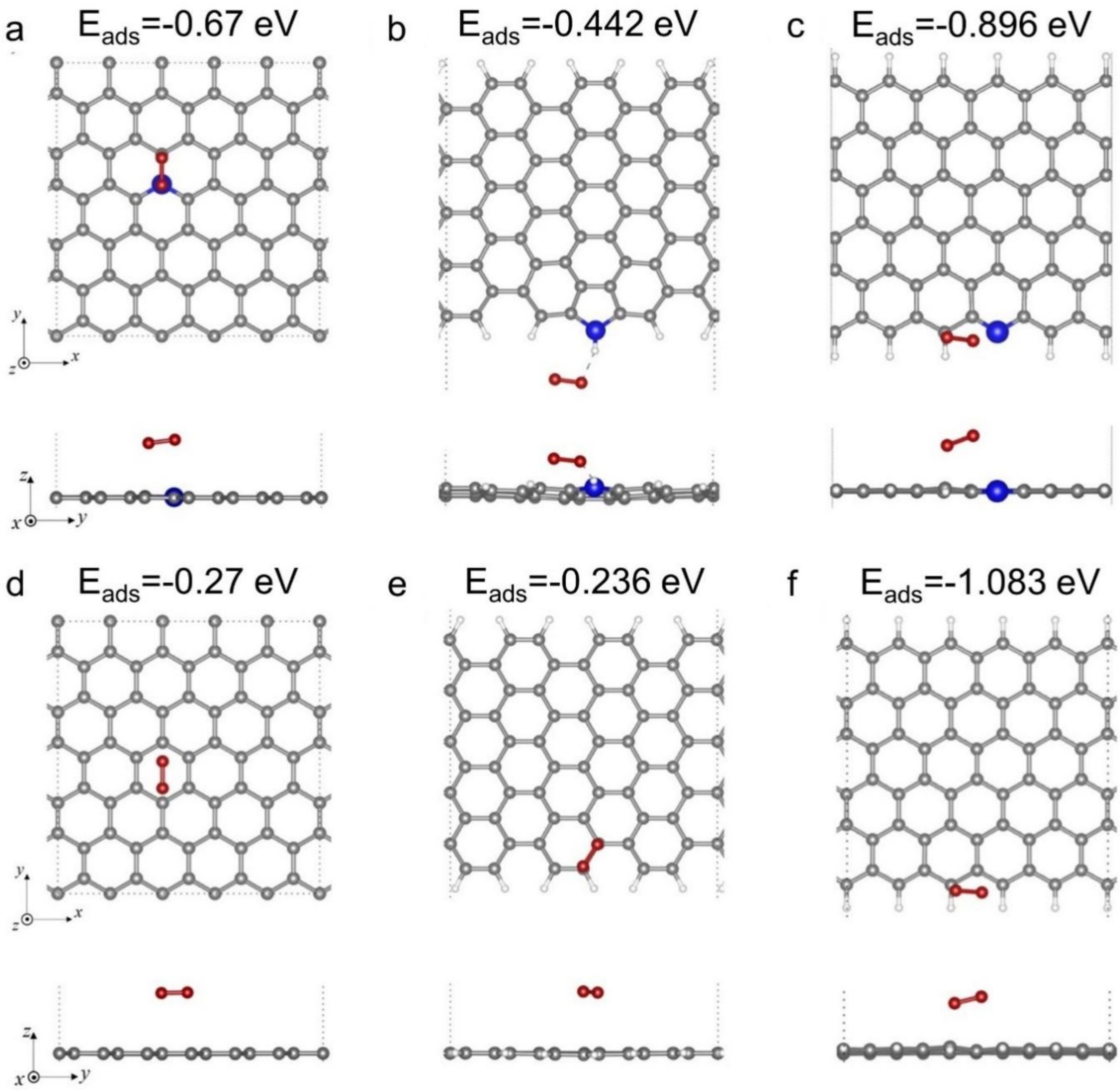

Fig S15 Optimal O_2 adsorption position at N-doped graphene and graphene. (a) O_2@ Graphitic-N. (b) O_2@ Pyrrolic-N. (c) O_2@ Pyridinic-N. (d) O_2@Graphene. (e) O_2@AC-Graphene. (f) O_2@ZZ-Graphene.

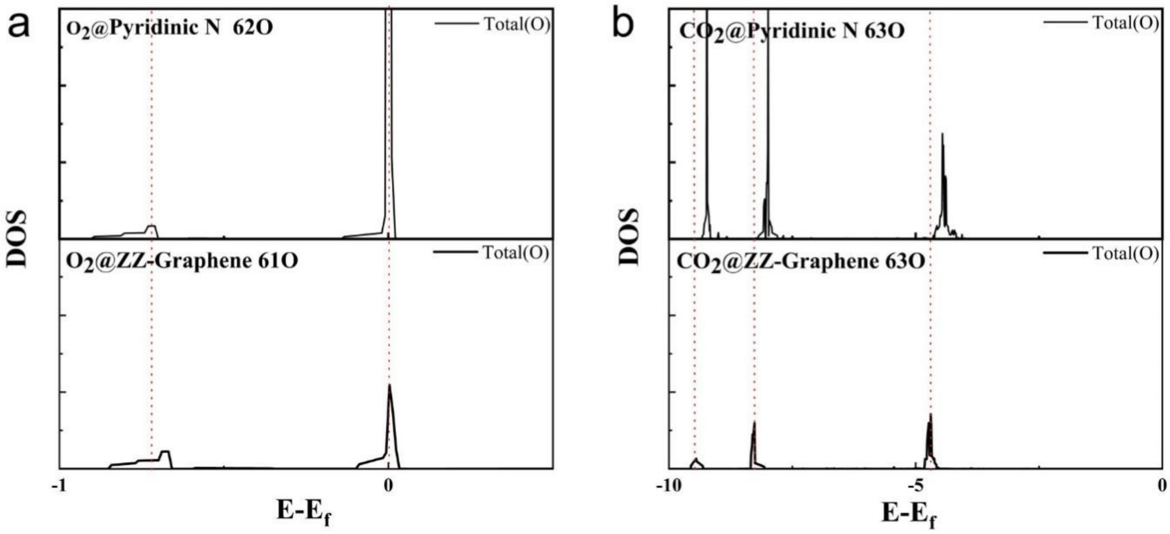

Fig S16 Density of states of the nearest adsorbed atom. (a) Nearest adsorbed O atom in O_2@Pyridinic-N and O_2@ZZ-graphene. (b) Nearest adsorbed O atom in CO_2@Pyridinic-N and CO_2@ZZ-graphene.

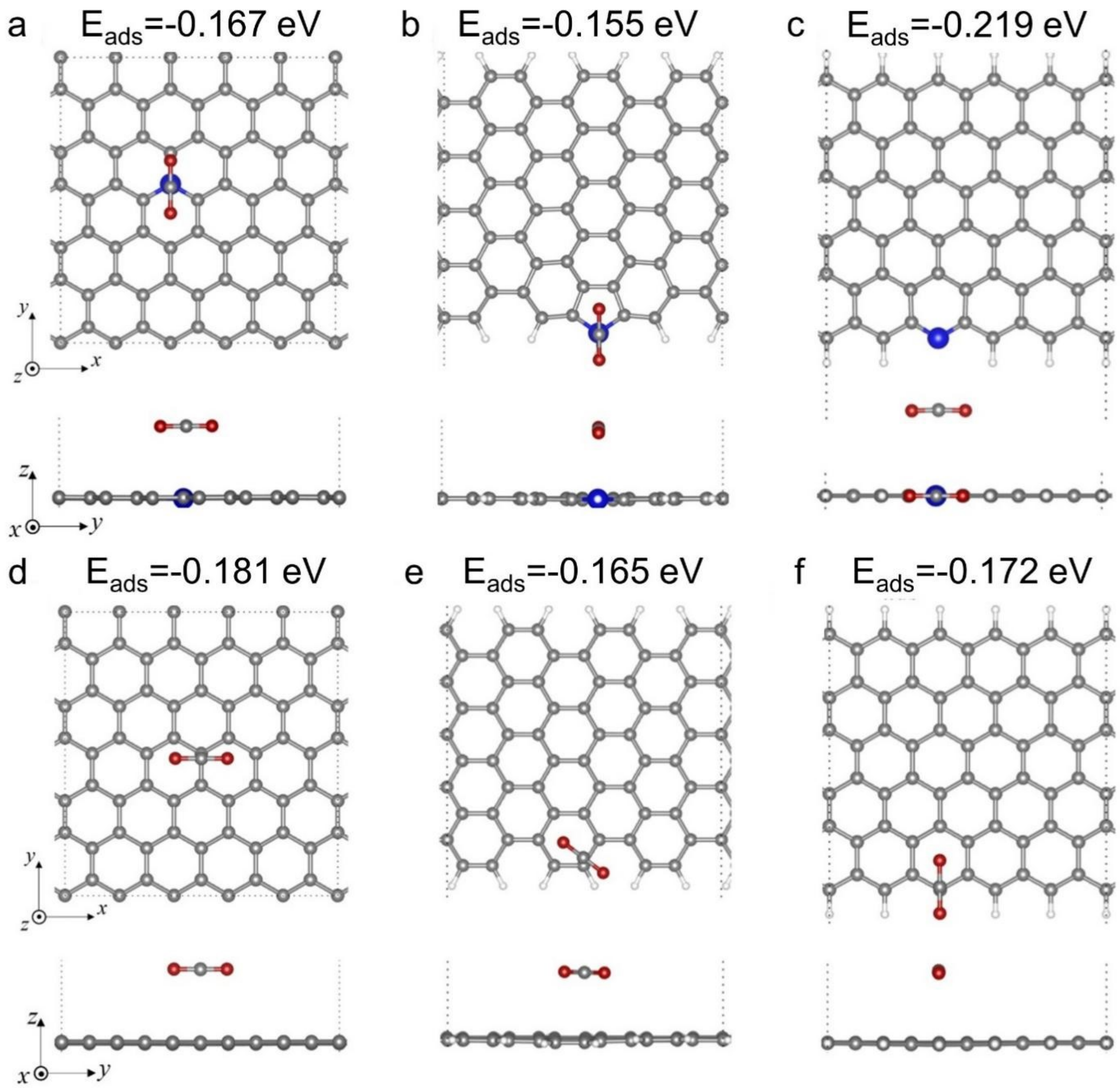

Fig S17 Optimal CO_2 adsorption position at graphene and N-doped graphene. (a) CO_2@ Graphitic-N. (b) CO_2@ Pyrrolic-N. (c) CO_2@ Pyridinic-N. (d) CO_2@Graphene. (e) CO_2@AC-Graphene. (f) CO_2@ZZ-Graphene.

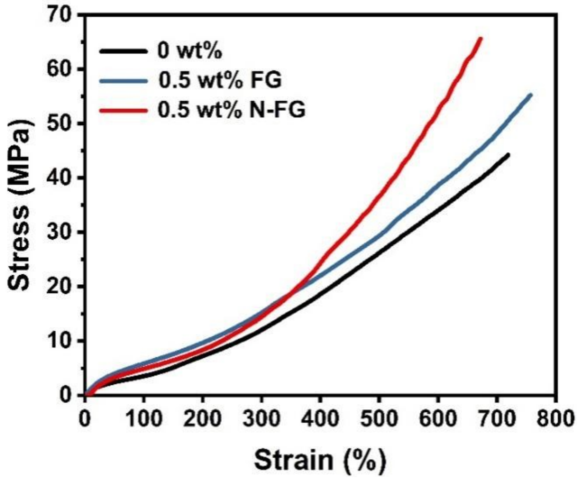

Fig. S18 Stress-Strain curves of 0.5 wt% FG/TPU and 0.5 wt% N-FG/TPU.

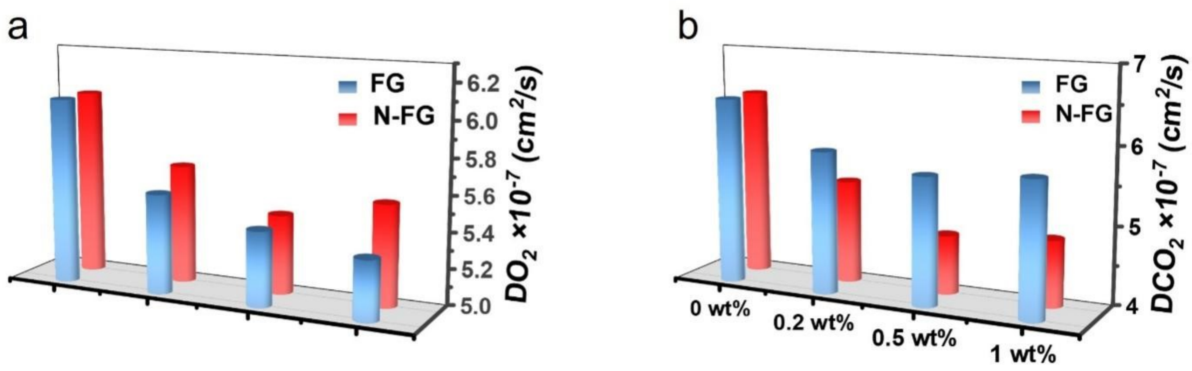

Fig. S19 Gas diffusion coefficient of FG/TPU and N-FG/TPU composite films. (a) Oxygen. (b) Carbon dioxide.

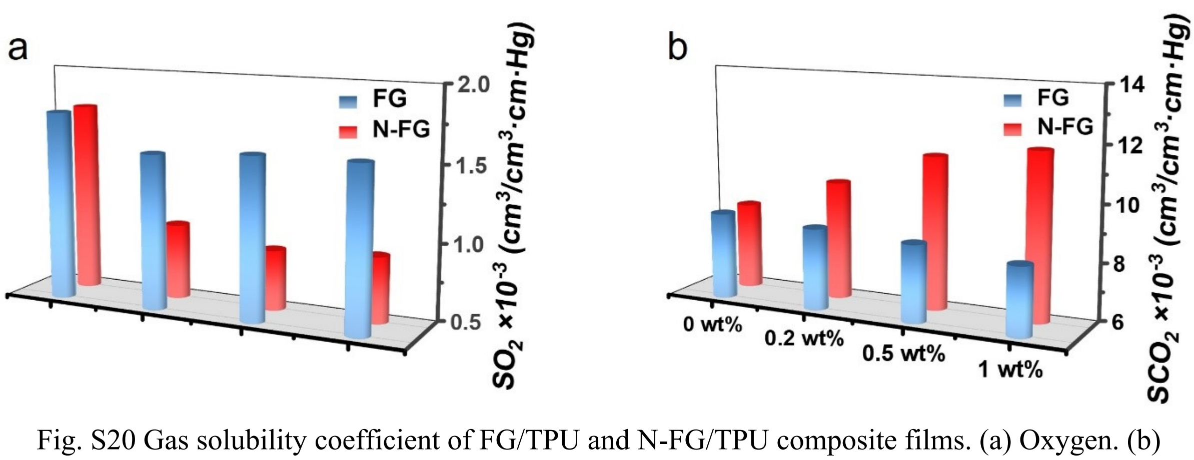

Fig. S20 Gas solubility coefficient of FG/TPU and N-FG/TPU composite films. (a) Oxygen. (b) Carbon dioxide.

Table S1 Cost of fillers required to achieve a 20% reduction in O_2 permeability (Calculated for 1 ton of composite).

Filler	Loading amount (wt%)	Unit price ($/t)	Sum ($/t)
GO	0.435	5900000[1]	25665.00
MMT	1.499	3580[2]	53.66
FG	0.198	189.5[3]	0.38

Table S2 Cost of fillers required to achieve a 15% reduction in CO_2 permeability (Calculated for 1 ton of composite).

Filler	Loading amount (wt%)	Unit price ($/t)	Sum ($/t)
GO	0.501	5900000[1]	29559.00
MMT	8.489	3580[2]	303.91
FG	0.177	189.5[3]	0.34

Table S3 Carbonyl index of pure HDPE, 1 wt% FG/HDPE and 1 wt% N-FG/HDPE before and after aging.

Sample type	$A_{C\text{-}H}$	$B_{C=O}$	CI
HDPE	33.43	1.24	0.038
HDPE-aging	38.64	3.17	0.082
1 wt% FG/HDPE	32.58	0.84	0.026
1 wt% FG/HDPE-aging	31.63	1.29	0.041
1 wt% N-FG/HDPE	19.10	0.14	0.0075
1 wt% N-FG/HDPE-aging	19.72	0.21	0.0105

Table S4 Comparison of mechanical performance: FG, N-FG, and other fillers (Based on recent literature).

Types of graphene	Polymer	Loading amount (wt%)	The increase of tensile strength (%)	The increase of elongation at break (%)	Reference
Graphene nanoplatelet (GNP)	HDPE	10	23.0	-85.0	[4]
GO	HDPE	1.0	11.0	-10.0	[4]
GO	HDPE	0.1	10.3%	-0.4	[5]
GO	HDPE	0.5	7.4	12.6	[6]
Graphene nanosheets (GNS)	PE	1.4	-3.2	-24.5	[7]
Graphene nanoplatelets(GNP)	HDPE	0.5	23.0	-20.1	[8]
MWCNT	HDPE	2.5	-54.5	-92.9	[9]
FG	HDPE	1	38.5	85.3	This work
N-FG	HDPE	1	47.2	82.0	This work

Table S5 Comparison of gas barrier performance: FG, N-FG, and other fillers (Based on recent literature).

Types of graphene	Polymer	Loading amount (wt%)	PO_2 decrease rate (%)	Reference
Graphene nanoplatelets (GNP)	HDPE	2	29.8	[10]
			35.7	[11]
Multiwalled carbon nanotubes (MWCNTs)	HDPE	2.5	50.38	[9]
GO	HDPE	1.0	28.3	[12]
MWCNTs	HDPE	1.0	20.1	[12]
FG	HDPE	1.0	27.1	This work
N-FG	HDPE	0.5	38.5	This work

Table S6 Atomic charge transfer of adsorbed gas molecules at the edges of ZZ-graphene and pyridine-N. Highlighted atoms are those adsorbed in the nearest position.

System	Adsorption of gas	Q (e)		ΔQ (e)
ZZ-Graphene	O_2	Atom 61	6.1634	-0.1634
		Atom 62	6.1983	-0.1983
Pyridinic N		Atom 61	6.1651	-0.1651
		Atom 62	6.1484	-0.1484
ZZ-Graphene	CO_2	Atom 61	2.3849	1.6151
		Atom 62	6.6988	-0.6988
		Atom 63	6.9393	-0.9393
Pyridinic N		Atom 60	2.1197	1.8803
		Atom 62	6.9506	-0.9506
		Atom 63	6.9441	-0.9441

[1]Industrial-Grade Graphene Oxide, ACS Material, https://www.acsmaterial.com/industrial-grade-graphite-oxide.html, (accessed July 2025).
[2]Montmorillonite Clay, Alibaba, https://www.alibaba.com/product-detail/Advanced-Montmorillonite-Clay-Organic-Montmorillonite-Clay_1601431204782.html, (accessed July 2025)
[3]M. Du, P. A. Advincula., X. Ding, J. M. Tour. C. Xiang. *Adv. Mater.*, 2023, **35**, 2300129.
[4]S. Lin, M. A. S. Anwer, Y. Zhou, A. Sinha, L. Carson and H. E. Naguib, *Compos. Part B*, 2018, **132**, 61.

[5]Y. Lin, J. Jin and M. Song, *J. Mater. Chem.*, 2011, **21**, 3455.
[6]N. Yan, H. Xia, J. Wu, Y. Zhan, G. Fei and C. Chen, *J. Appl. Polym. Sci.*, 2012, **127**, 933-941.
[7]F. d. C. Fim, N. R. S. Basso, A. P. Graebin, D. S. Azambuja and G. B. Galland, *J. Appl. Polym. Sci.*, 2012, **128**, 2630.
[8]A. Dargahi, M. Duncan, J. Runka, A. Hammami, T. Wen, X. Wang, W. Chen and H. E. Naguib, *Ind. Eng. Chem. Res.*, 2025, **64**, 5359.
[9]K. Chrissafis, K. M. Paraskevopoulos, I. Tsiaoussis and D. Bikiaris, *J. Appl. Polym. Sci.*, 2009, **114**, 1606.
[10]K. Honaker, F. Vautard, L. T. Drzal, *Adv. Compos. Hybrid Mater.*, 2021, **4**, 492.
[11]K. Honaker, F. Vautard, L. T. Drzal, *Mater. Sci. Eng. B*, 2017, **216**, 23.
[12]Y. Q. Gill, J. Jin and M. Song, *Polym. Polym. Compos.*, 2019, **28**, 562.