

## **Supplementary Materials**

# **Synthesis of High-Efficiency, Eco-friendly, and Synergistic Flame Retardant for Epoxy Resin**

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((Optional Dedication))

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## 1. Supplementary Figures

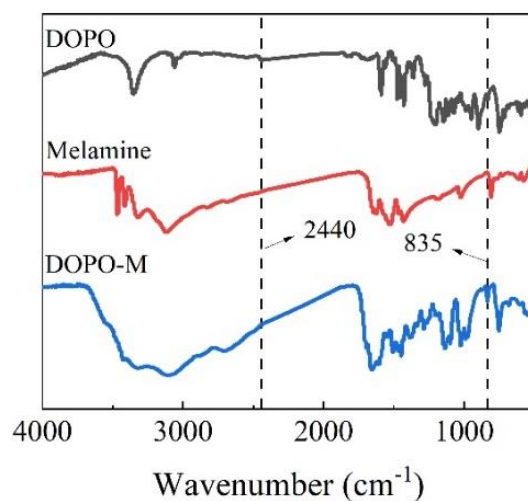


Figure S1. The FTIR spectra of DOPO, Melamine, and DOPO-M.

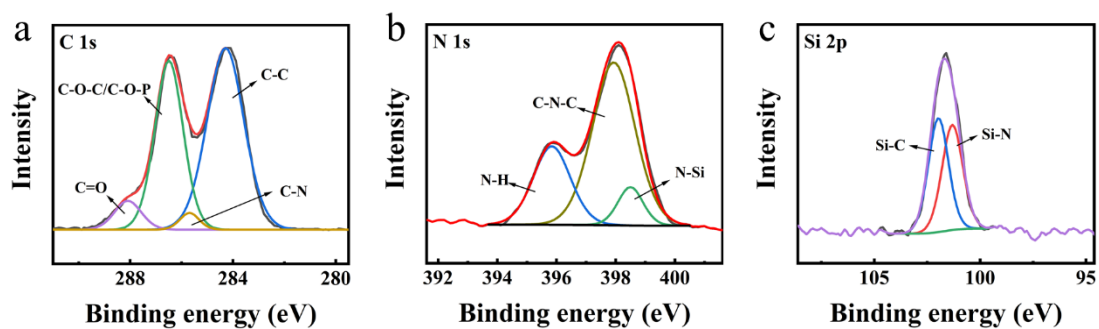


Figure S2. High-resolution XPS spectra for (a) C 1s; (b) N 1s; and (c) Si 2p of DOPO-M-rGO.

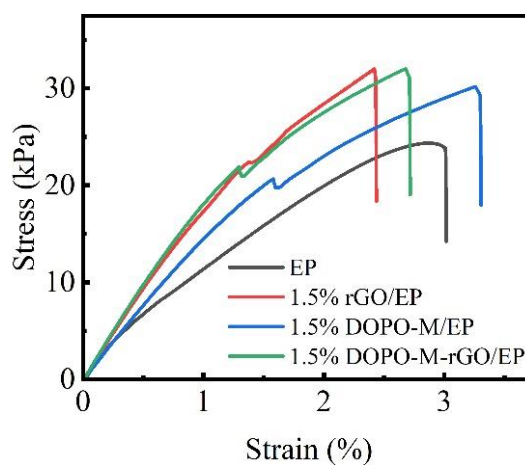


Figure S3. Stress-strain curves of pure EP, 1.5% rGO/EP composite, 1.5% DOPO-M/EP composite, and 1.5% DOPO-M-rGO/EP composite.

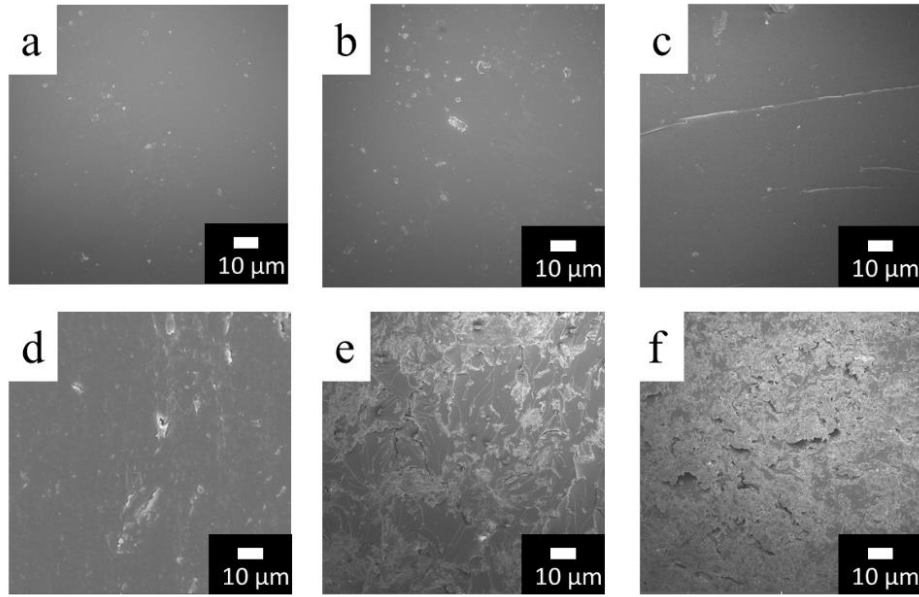


Figure S4. SEM images of (a) 0.5% DOPO-M-rGO/EP; (b) 1% DOPO-M-rGO/EP; (c) 1.5% DOPO-M-rGO/EP; (d) 2% DOPO-M-rGO/EP; (e) 3% DOPO-M-rGO/EP; (f) 5% DOPO-M-rGO/EP.

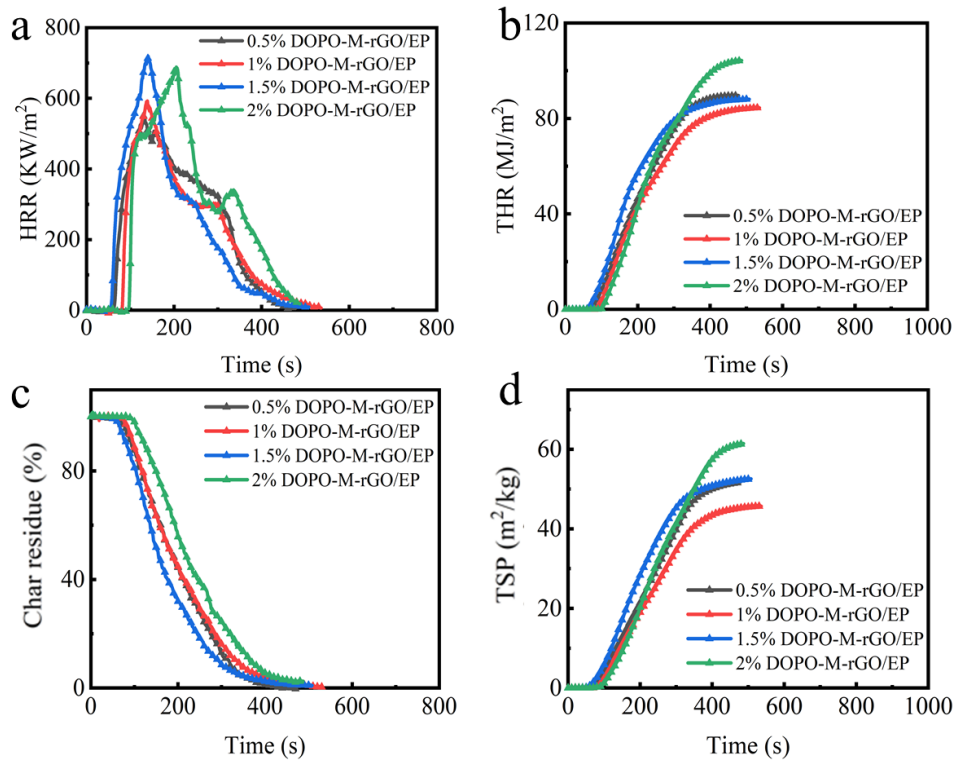


Figure S5. (a) Heat release rate; (b) total heat release; (c) char residue curves; (d) total smoke production of DOPO-M-rGO/EP composites with different DOPO-M-rGO contents under an external heat flux of 35 kW/m<sup>2</sup>.

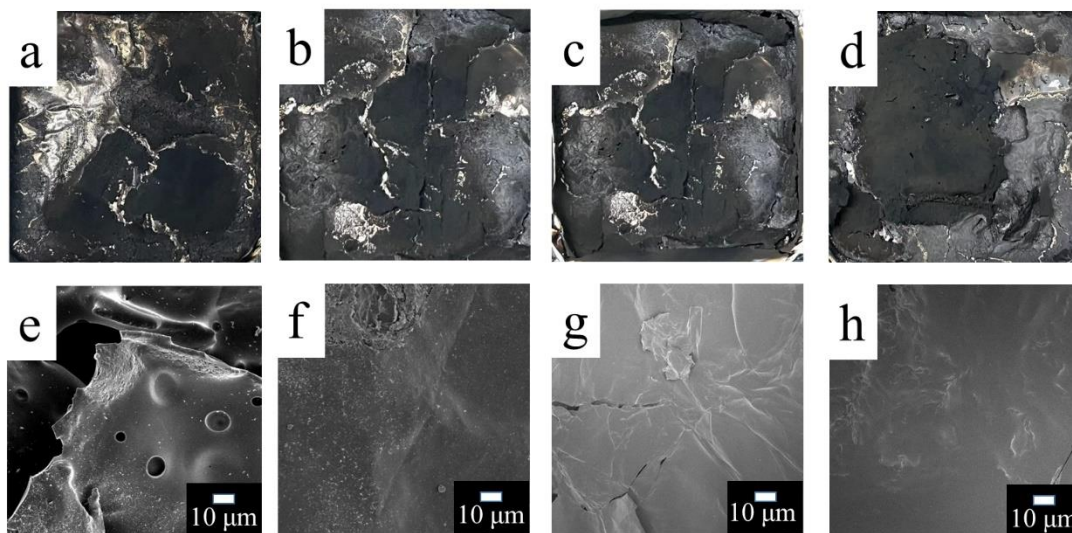


Figure S6. Digital images of char residues for (a) 0.5% DOPO-M-rGO/EP; (b) 1% DOPO-M-rGO/EP; (c) 1.5% DOPO-M-rGO/EP; (d) 2% DOPO-M-rGO/EP and SEM images of char residues for (e) 0.5% DOPO-M-rGO/EP; (f) 1% DOPO-M-rGO/EP; (g) 1.5% DOPO-M-rGO/EP; (h) 2% DOPO-M-rGO/EP after cone calorimeter test.

## 2. Supplementary Tables

Table S1. Performance comparison of the graphene-based flame retardants.

Flame retardant	Amount of flame retardant added (wt%)	Cone Calorimeter Test			UL-94 Rating	Ref.
		pHRR (%)	THR (%)	TSP (%) <sup>a</sup>		
DOPO-M-rGO	1.5	55	30	20	V-0	This paper
F-RGO-CuMoO <sub>4</sub>	2	60	44	62	V-1	S1
ZnPhP-PGO	3	41	17	6	V-0	S2
PPGO	3	42	22	24	-	S3
RGO@Ni(OH) <sub>2</sub>	2	32	9	8	V-2	S4
PN-rGO	5	31	29	51	V-1	S5
RGO-PANI/Ni(OH) <sub>2</sub>	3	40	11	36	NR	S6
rGO@ZnPb	5	39	31	34	V-0	S7
SBA-15-RGO-ZHS	3	56	27	38	V-1	S8
OapPOSS-rGO	2	49	37	-22	-	S9
ZIF8/rGO	2	65	34	42	V-1	S10

*a*: The negative number indicates an increased value.

Table S2. Performance comparison of the DOPO-based flame retardants.

Flame retardant	Amount of flame retardant added (wt%)	Cone Calorimeter Test			UL-94 Rating	Ref.
		pHRR (%)	THR (%)	TSP (%) <sup>a</sup>		
DOPO-M-rGO	1.5	55	30	20	V-0	This paper
DOPO-MMT	6	55	20	9	V-0	S11
30DOPO-P-KC	5	51	27	20	V-0	S12
DOPO/AHPP	5	59	45	14	V-0	S13
FR-CNTs	8	31	49	-15	V-0	S14
DOPO@ZIF-67	4	11	6	-53	V-0	S15
DOPO-CuPc	20	58	57	22	V-0	S16
AVD	10	61	17	15	V-0	S17
HPDAI	2	25	25	23	V-0	S18
DEN438/D/F	2.5	31	-6	49	NR	S19
PDTT	10	31	26	-32	V-0	S20

*a*: The negative number indicates an increased value.

Table S3. Performance comparison of the flame retardants based on DOPO-functionalized rGO.

Flame retardant	Amount of flame retardant added (wt%)	Cone Calorimeter Test			UL-94 Rating	Ref.
		pHRR (%)	THR (%)	TSP (%) <sup>a</sup>		
DOPO-M-rGO	1.5	55	30	20	V-0	This paper
GP-DOPO	2	27	32	31	V-2	S21
DOPO-VTES-GO	5	45	41	36	-	S22
DGO	10	38	46	-	V-0	S23
GNS/DOPO	5	67	34	-14	-	S24
OapPOSS-rGO	2	49	36	-22	-	S25
GO-DOPO-V	2	29	16	-	-	S26
DOPO-V-PA	3	22	9	-	-	S27
FRs-rGO	5	35	-	-	V-0	S28
Graphene-DOPO	3	39	25	30	V-1	S29
PD-rGO	4	43	30	32	V-1	S30
FRGO	3	34	14	30	V-1	S31
f-rGO	3	31	34	10	-	S32

<sup>a</sup>: The negative number indicates an increased value.

Table S4. LOI and UL-94 test results of the pure EP and the EP-based composites with 1.5% rGO, 1.5% DOPO-M, and different contents of DOPO-M-rGO as the additives.

Sample	LOI (%)	UL-94
pure EP	25	NR
1.5% rGO/EP	25	NR
1.5% DOPO-M/EP	25	NR
0.5% DOPO-M-rGO/EP	28	V-2
1% DOPO-M-rGO/EP	32	V-1
1.5% DOPO-M-rGO/EP	32	V-0
2% DOPO-M-rGO/EP	32	V-0

Table S5. Cone calorimeter data of the EP-based composites with different DOPO-M-rGO contents.

Sample	pHRR (kW/m <sup>2</sup> )	THR (MJ/m <sup>2</sup> )	TSP (m <sup>2</sup> /kg)
0.5% DOPO-M-rGO/EP	551	90	51
1% DOPO-M-rGO/EP	592	85	46
1.5% DOPO-M-rGO/EP	719	88	52
2% DOPO-M-rGO/EP	688	104	61

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