Supporting Information for

Optimizing Thermal and Dielectric Properties of Ethylene-tetrafluoroethylene (ETFE)/h-BN Composites *via* Interface Engineering: Activation of C-F Bonds on ETFE for Surface Grafting

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Instrument

The particle size of the raw polymer powder was measured using a HELOS (H3938) 8 OASISDRY/L R7 laser particle size analyzer. The chemical structure of graft copolymers was confirmed using Fourier Transform Infrared Spectroscopy (FT-IR) (Tensor 27 FT-IR, Bruker, Germany). Differential Scanning Calorimetry (DSC) results were obtained using Netzsch DSC 200 PC (Netzsch, Germany) with a heating rate of 10°C/min from 30°C to 180°C. The crystalline structure of samples was determined by X-ray Diffraction (XRD) analysis using Cu Kα radiation (λ = 1.542 Å, 2.2 kV), with a scanning rate of 4°/min. Thermogravimetric analysis was conducted under a nitrogen atmosphere using a TGA instrument (Mettler Toledo, Switzerland). The heating rate was 10°C/min, with a testing temperature range of 30-600°C (flow rate of 50 mL/min). The mechanical properties of the films were tested at room temperature using a universal testing machine. The in-plane thermal conductivity was measured using a TC 3000E thermal conductivity meter based on ASTM C 1113 (hot wire method). The through-plane thermal conductivity of the composites was determined using a LFA447 laser flash analyzer from NETZSCH. Samples were disc-shaped, with a diameter of 12.7 mm and a thickness of approximately 1 mm, with a thin graphite layer sprayed on both surfaces to enhance heat transfer. The thermal dissipation properties of the composites were characterized using a handheld infrared thermal imaging camera (THT46, HT Italia). The dielectric constant and dielectric loss of the sample films were

tested using a Novocontrol Concept 80 broadband dielectric spectrometer (Novocontrol, Germany).

Characterization

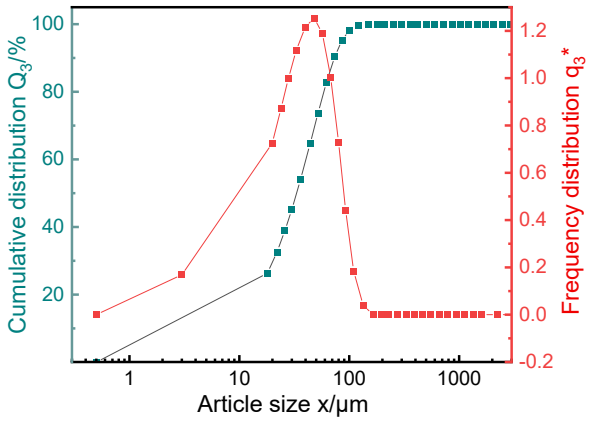

Figure S1. Particle size testing diagram.

Table S1. Cumulative distribution.

X_o/μm	Q_3/%	X_o/μm	Q_3/%	X_o/μm	Q_3/%
0.5	0	74	90.49	420	100
18	26.3	86	95.25	500	100
22	32.61	100	98.15	600	100
26	38.94	120	99.61	720	100
30	45.14	150	99.99	860	100
36	53.97	180	100	1020	100
44	64.59	210	100	1220	100
52	73.69	250	100	1460	100
62	82.78	300	100	1740	100

Table S2. Frequency distribution (LOG.).

X_m/μm	q_3 lg	X_m/μm	q_3 lg	X_m/μm	q_3 lg
0.5	0	67.73	1.004	388.84	0
3	0.169	79.77	0.729	458.26	0
19.9	0.724	92.74	0.442	547.72	0
23.92	0.872	109.54	0.184	657.27	0
27.93	0.998	134.16	0.039	786.89	0
32.86	1.116	164.32	0.001	936.59	0
39.8	1.216	194.42	0	1115.53	0
47.83	1.254	229.13	0	1334.62	0
56.78	1.19	273.86	0	1593.86	0

Table S3. h-BN contents in composites.

Sample	BN content (vol%)
ETFE@BN-10	10
ETFE@BN-20	20
ETFE@BN-30	30
ETFE-*g*-PGMA@BN-10	10
ETFE-*g*-PGMA@BN-20	20
ETFE-*g*-PGMA@BN-30	30

Table S4. Percentage of elemental content of ETFE and ETFE-*g*-PGMA.

Sample	Atomic percentage (%)			Elemental	
	C	O	F	F/C	O/C
ETFE	47	0	53	1.1	0
ETFE-*g*-PGMA	52	6	42	0.8	0.12

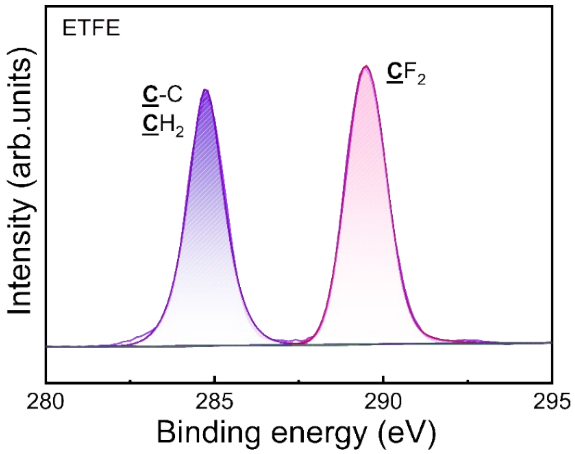

Figure S2. C1s spectra of ETFE.

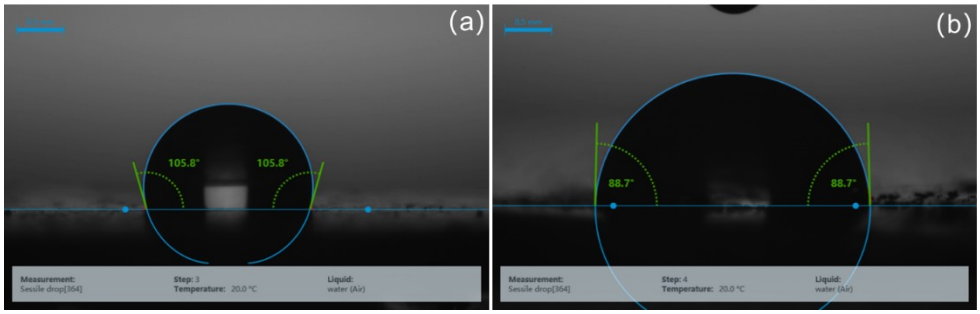

Figure S3. Contact angles on: (a) ETFE (contact angle = 105.8°) and (b) ETFE-*g*-PGMA (contact angle = 88.7°).

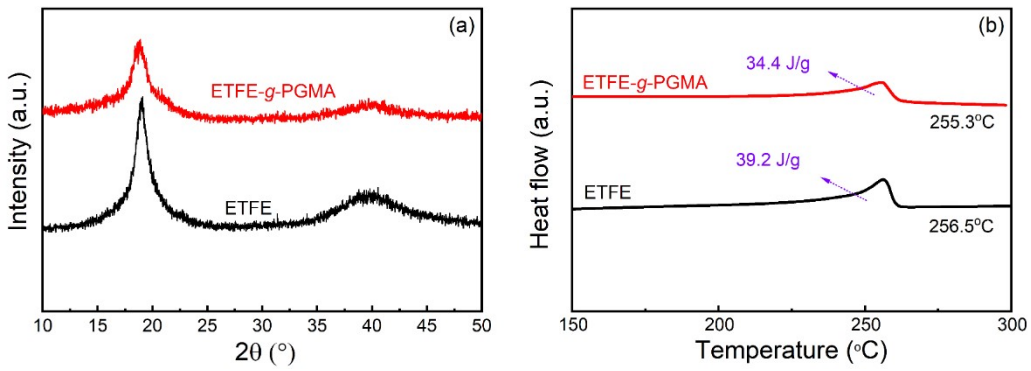

Figure S4. (a) XRD patterns and (b) DSC thermograms of pristine ETFE and ETFE-*g*-PGMA.

Table S5. Tensile Strength and Modulus of the Samples.

Entry	Tensile Strength (MPa)	Young's Modulus (MPa)
ETFE	24.3±1.3	4.6±0.3
ETFE-g-PGMA	22.4±1.5	5.5±0.4
ETFE@BN-30	20.2±1.1	16.4±0.6
ETFE-g-PGMA@BN-30	19.4±0.9	18.5±0.7

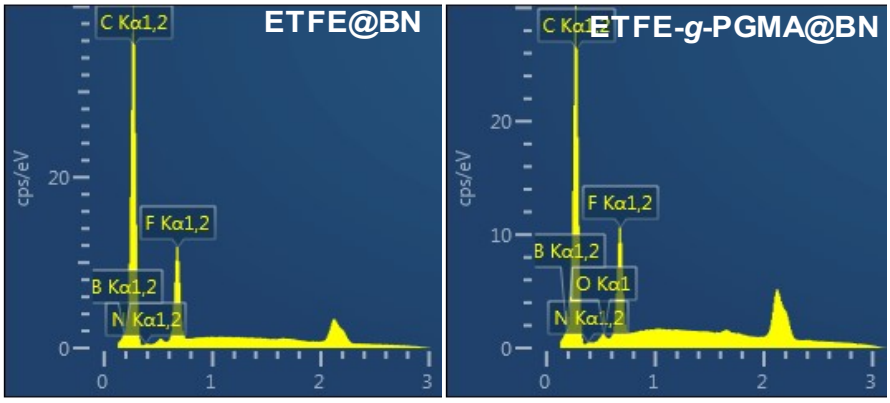

Figure S5. EDX spectra of ETFE@BN and ETFE-*g*-PGMA@BN composites.

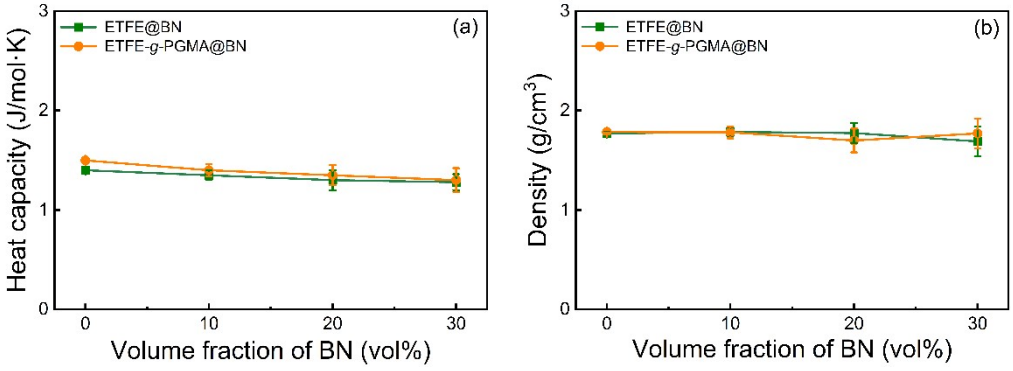

Figure S6. (a) Heat capacity determined by DSC as a function of h-BN filler content. (b) Measured density as a function of h-BN filler content, with an average ranging from 1.7 to 1.8 g/cm^3.