

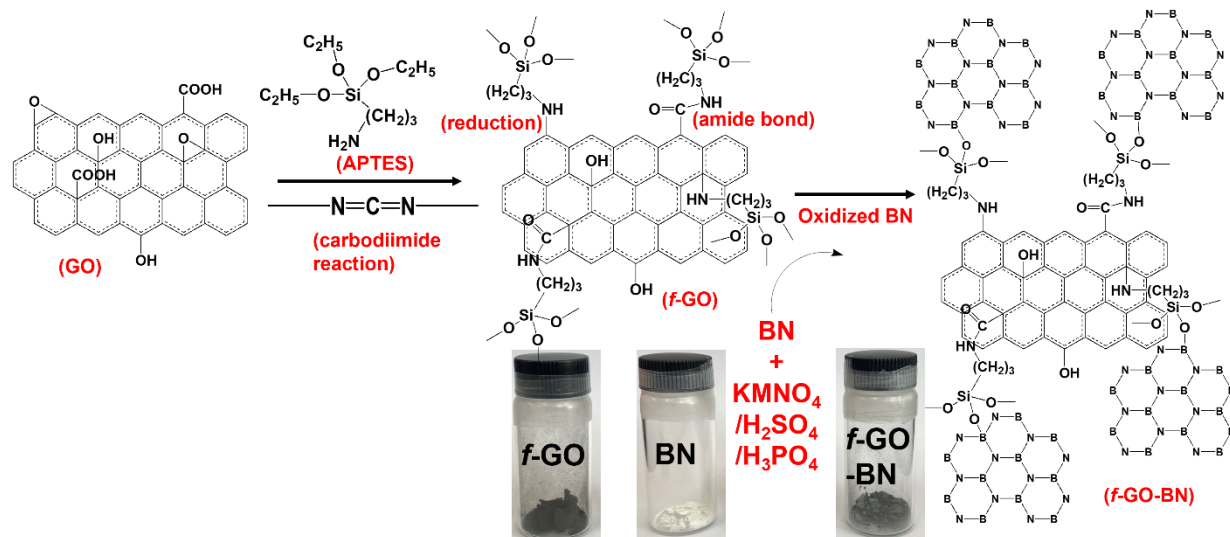
## **Fabrication of impermeable dense architecture containing covalently stitched graphene oxide/boron nitride hybrid nanofiller reinforced semi-interpenetrating network for hydrogen gas barrier application**

### **Electronic Supplementary Information**

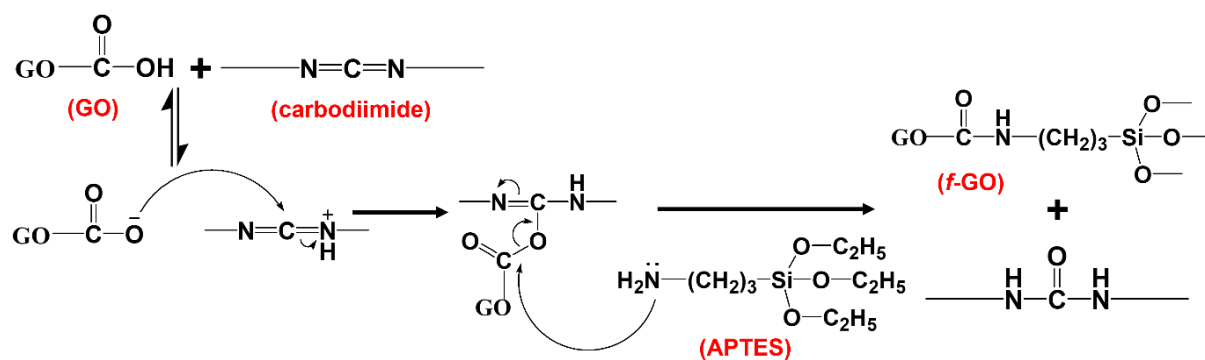
#### **Synthesis of oxidized BN**

BN was exfoliated in 1:1 (v/v) mixture of isopropanol and DI water by bath sonication for 4 h. After the sonication, the suspension was centrifuged and the supernatant was taken. The exfoliated BN was then collected by vacuum-assisted filtration. After that, it was oxidized by following the TOUR method, which is commonly applied to synthesize GO. To carry out the reaction, 300 mg of BN was mixed with 1.8 g of  $\text{KMnO}_4$  in a round bottom flux. 40 mL of mixed acid was prepared separately by mixing  $\text{H}_2\text{SO}_4$  (95%) and  $\text{H}_3\text{PO}_4$  (85%) in 9:1 (v/v) ratio and added slowly in the dry BN/ $\text{KMnO}_4$  mixture in an ice bath. The reaction was mildly exothermic, leading to a rise in temperature up to 35-40 °C. The reaction mixture was allowed to stir for 30 min in an ice bath. After that, the ice bath was removed and the stirring was continued for 12 h at 60 °C. Subsequently, the reaction was cooled down to room temperature, and the suspension was slowly added to 50 ml of ice-cold DI water under constant stirring followed by the addition of 3 mL of  $\text{H}_2\text{O}_2$  to cease the oxidation reaction. The stirring was continued for another 1 h. After the reaction, the suspension was washed with DI water, 30 % HCl and ethanol. Further, the washing was carried out with DI water until the pH of the medium reached above 3. After each washing, the sediment was collected by ultracentrifugation and finally dried under the vacuum.

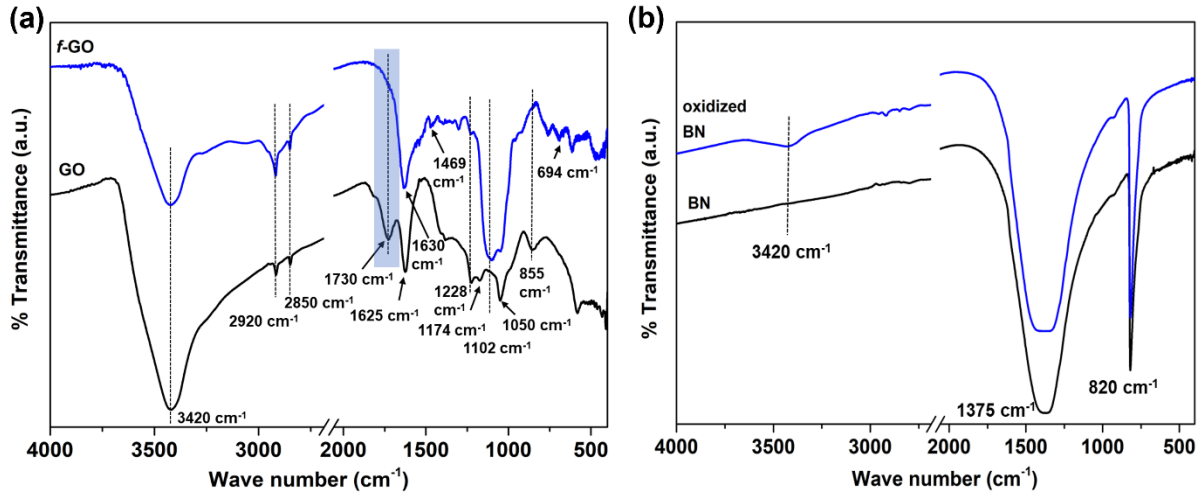
(a)



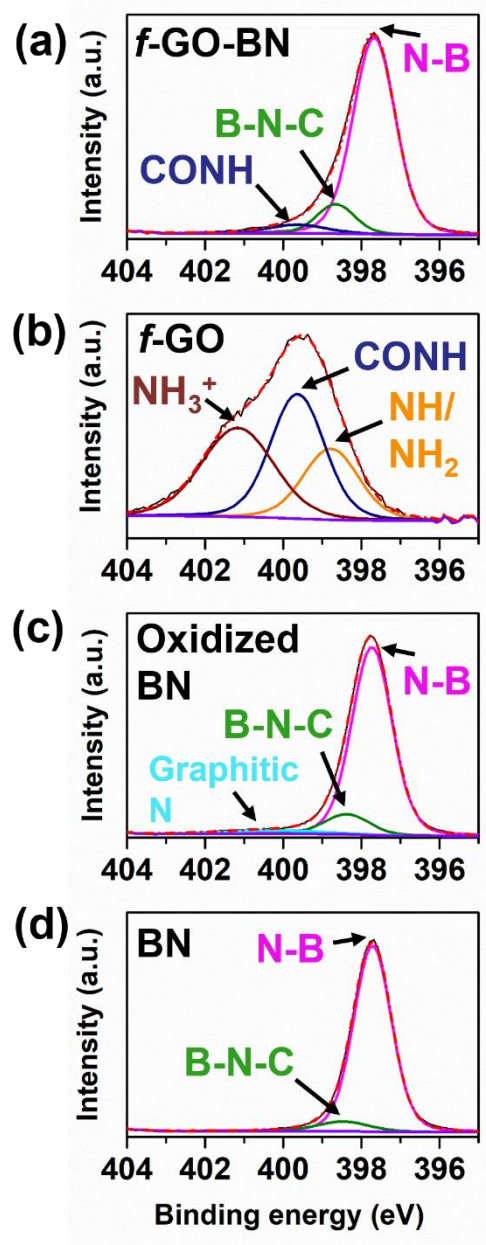
(b)



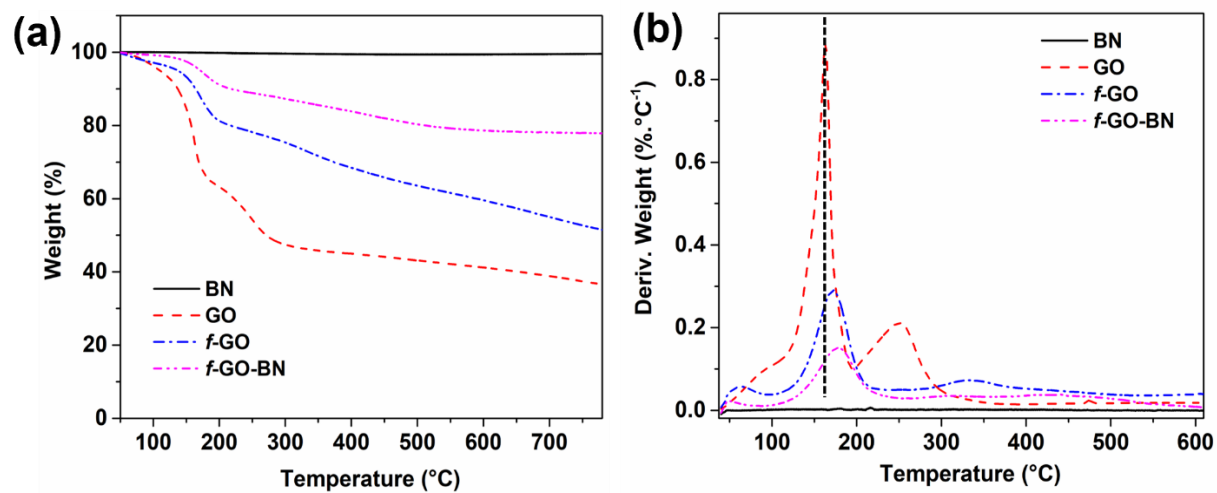
**Fig. S1** (a) Probable reaction mechanism involved in the formation of *f*-GO-BN, (b) carbodiimide reaction mechanism for the grafting of APTES onto GO.



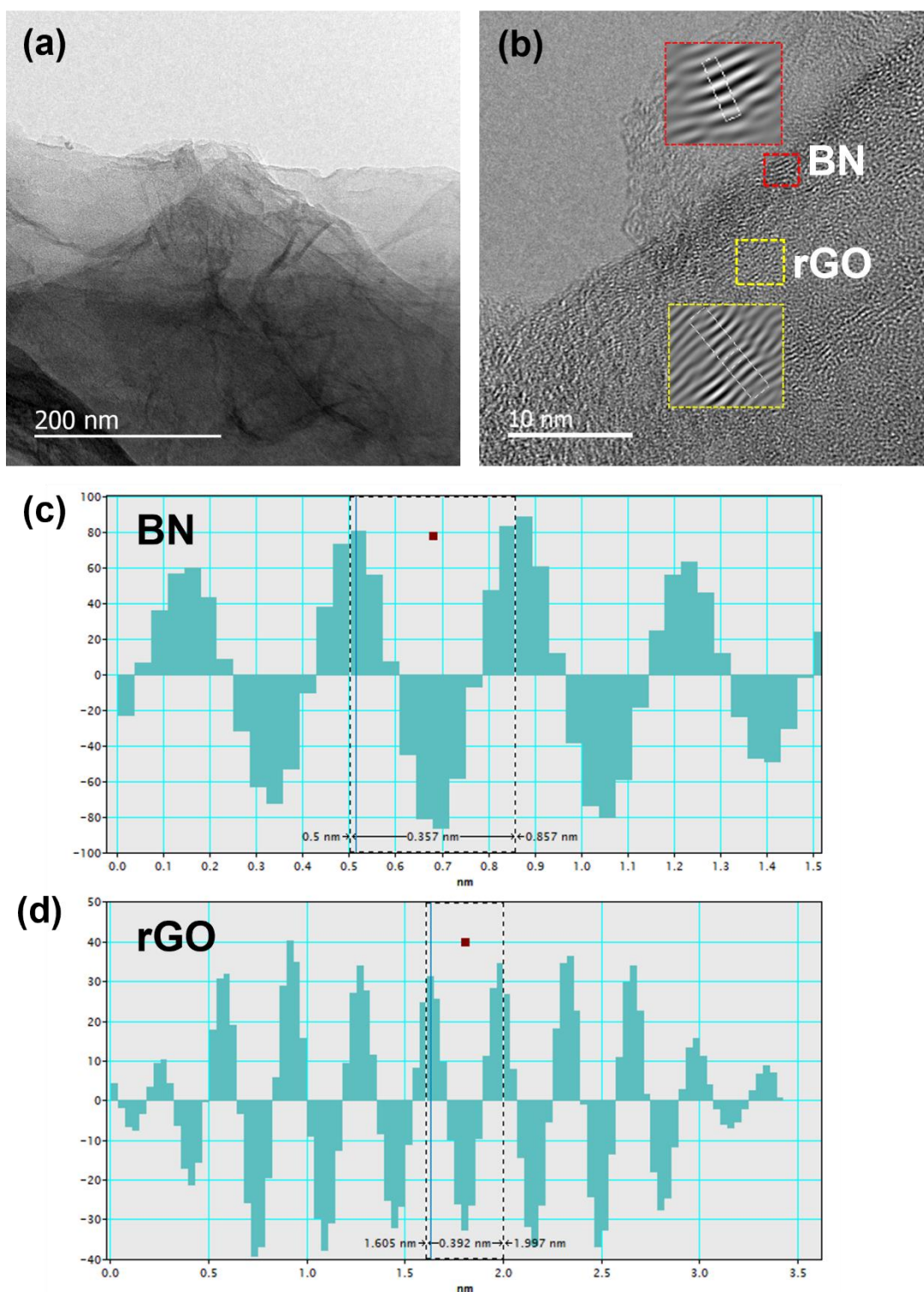
**Fig. S2** FTIR spectra of (a) GO and APTES functionalized GO (*f*-GO), and (b) BN before and after oxidation.



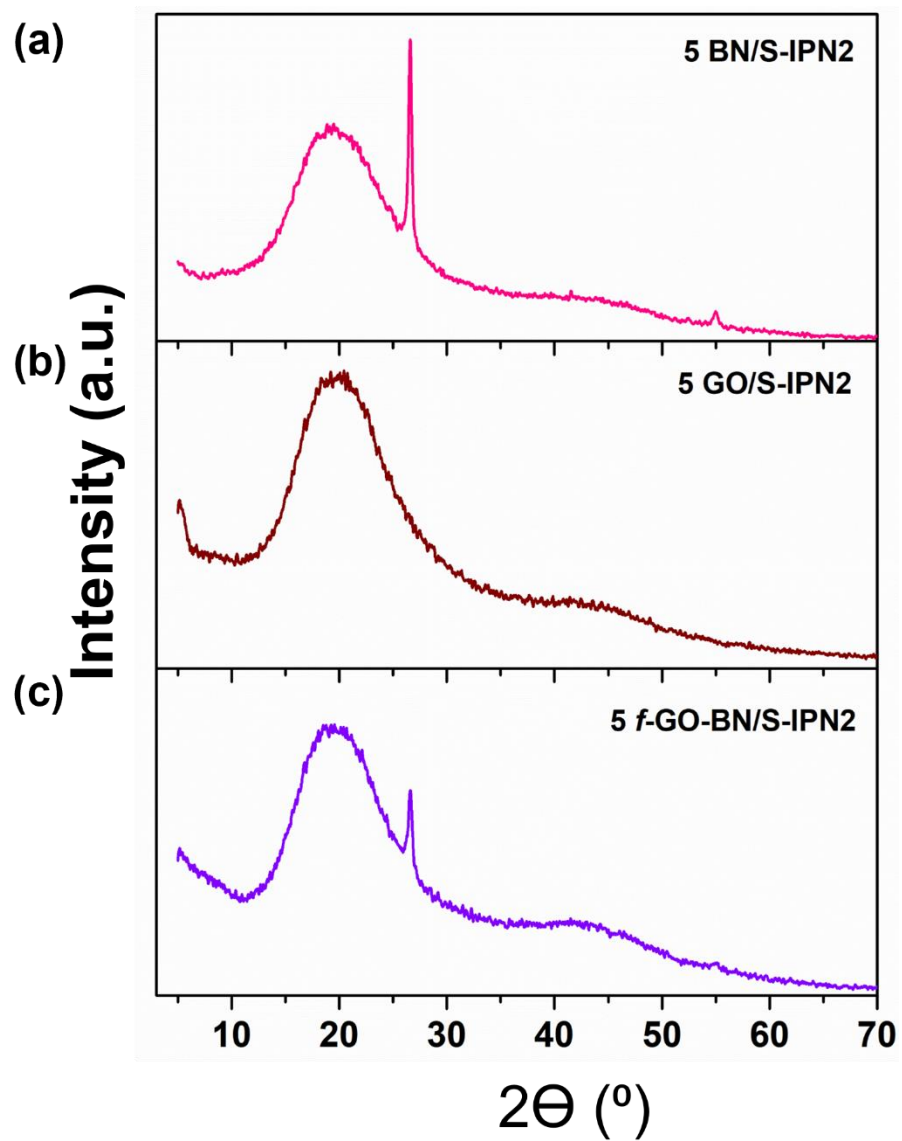
**Fig. S3** High resolution N1s spectra of (a) *f*-GO-BN, (b) *f*-GO, (c) oxidized BN and (d) BN.



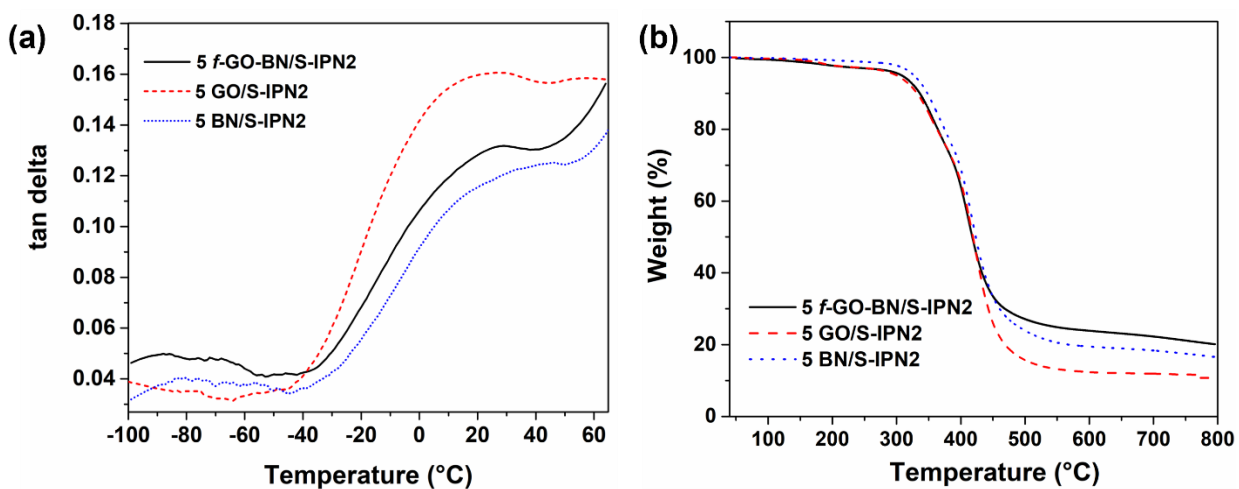
**Fig. S4** (a) and (b) TGA and DTGA curves of BN, GO, *f*-GO and *f*-GO-BN, respectively.



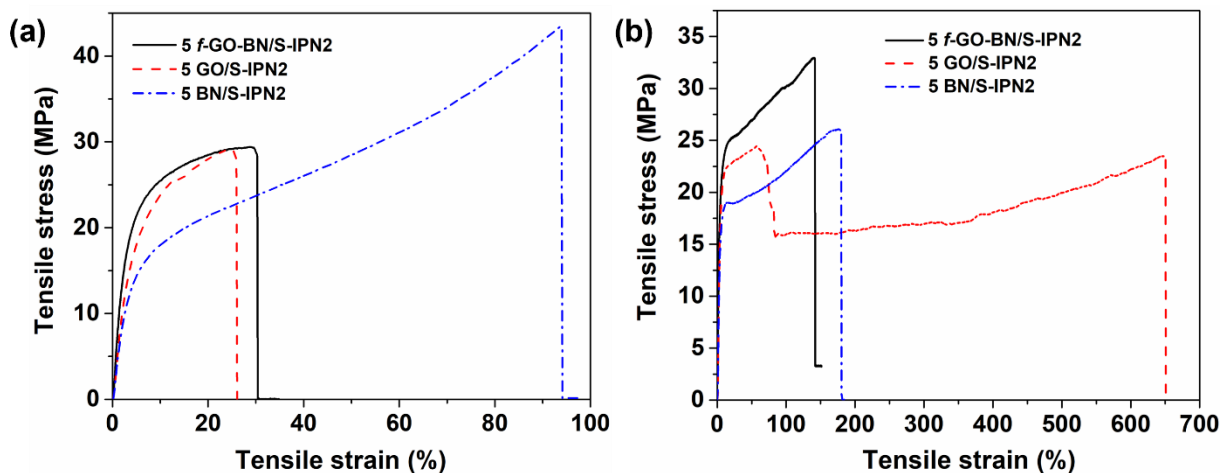
**Fig. S5** TEM image of *f*-GO-BN at (a) low magnification, (b) high magnification (inverse FFT images are given in the inset); d spacing image of (c) BN and (d) rGO, as shown in (b).



**Fig. S6** Wide-angle XRD pattern of (a) 5 BN/S-IPN2, (b) 5 GO/S-IPN2 and (c) 5 *f*-GO-BN/S-IPN2.

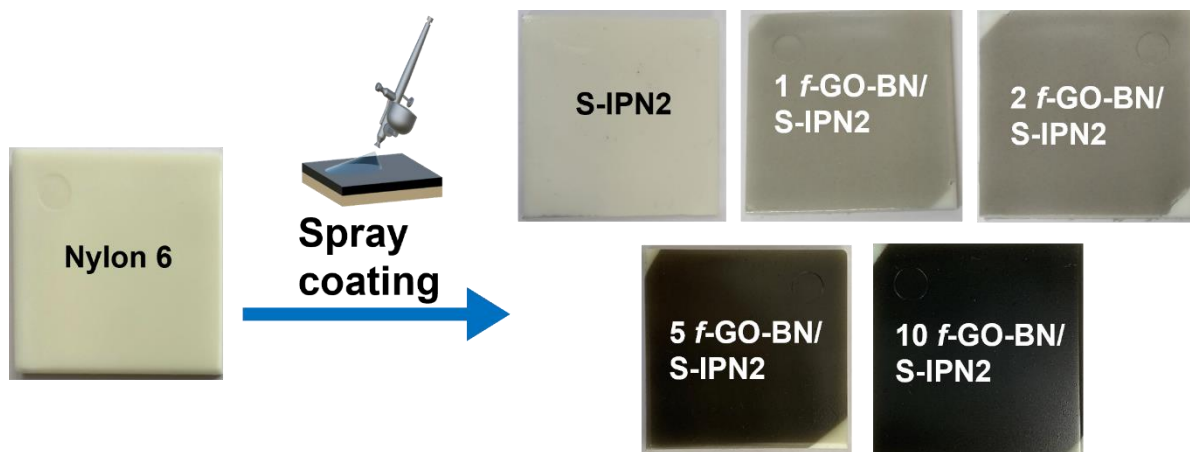


**Fig. S7** (a) tan delta and (b) TGA curve of 5 *f*-GO-BN/S-IPN2, 5 GO/S-IPN2 and 5 BN/S-IPN2.

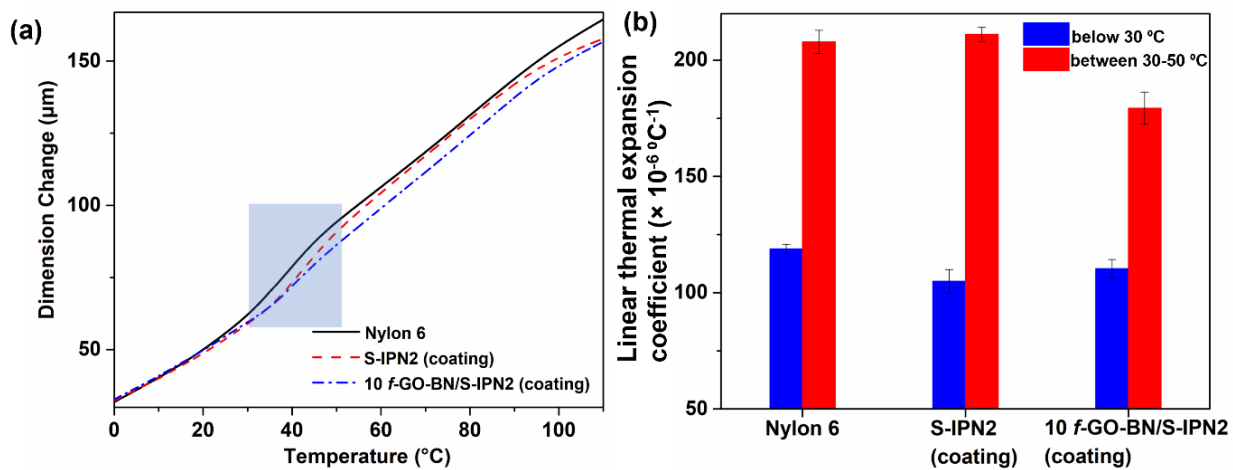


**Fig. S8** Tensile stress-strain curve of (a) solvent cast film and (b) spray-coated film of 5 *f*-GO-BN/S-IPN2, 5 GO/S-IPN2 and 5 BN/S-IPN2.

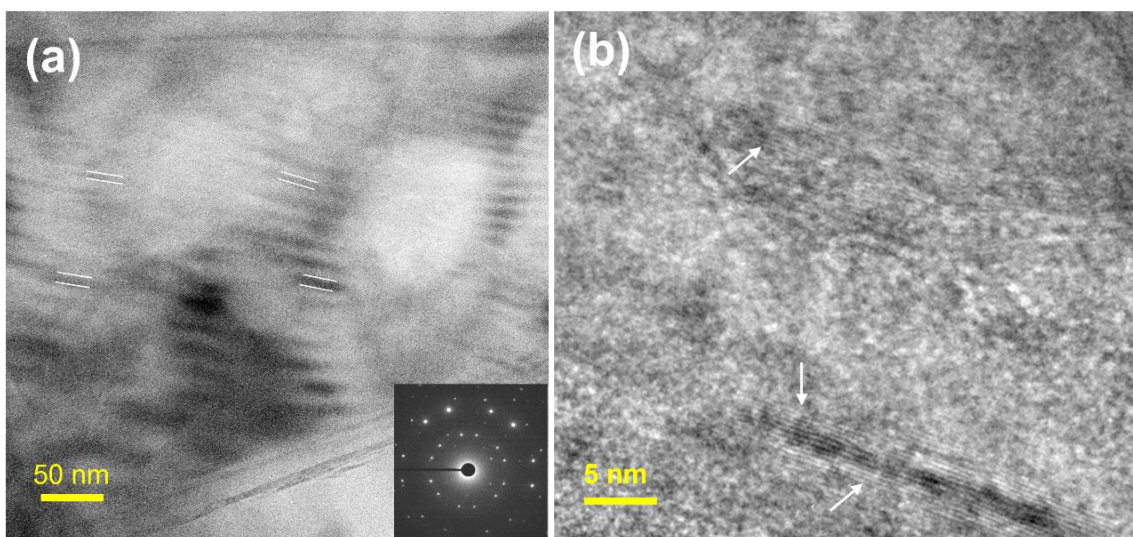




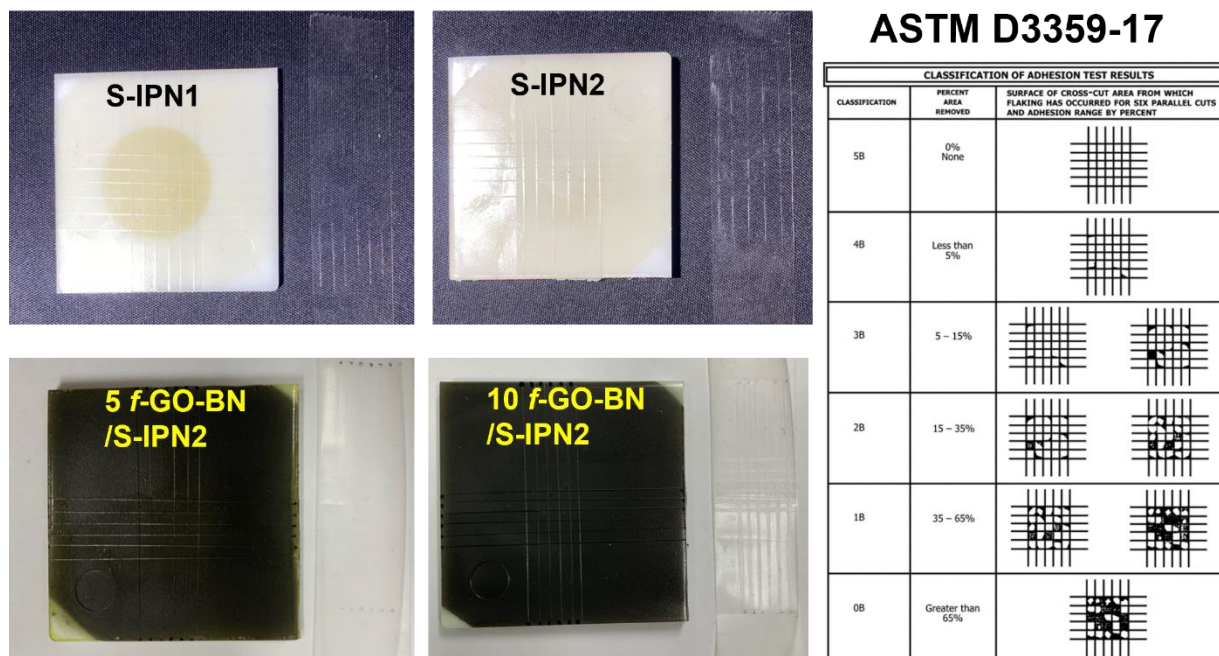
**Fig. S9** Photographic images of nylon 6 substrate before and after coating with S-IPN2, 1 *f*-GO-BN/S-IPN2, 2 *f*-GO-BN/S-IPN2, 5 *f*-GO-BN/S-IPN2 and 10 *f*-GO-BN/S-IPN2.



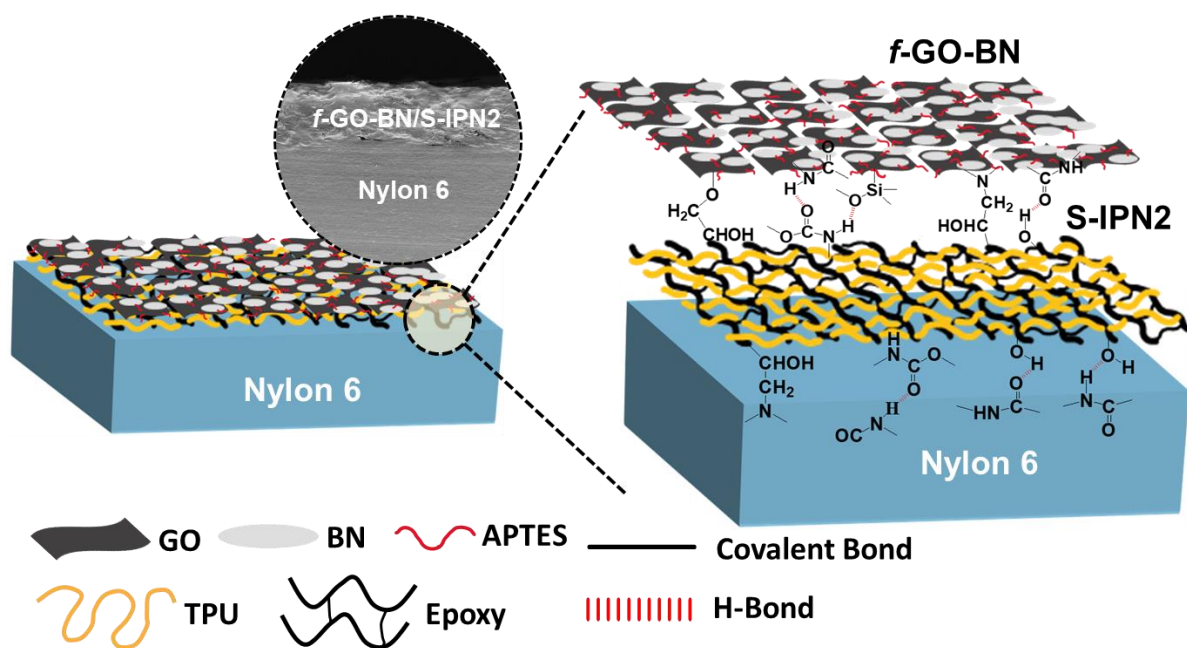
**Fig. S10** (a) and (b) Linear thermal expansion and thermal expansion coefficient of uncoated nylon 6, S-IPN2 and 10 *f*-GO-BN/S-IPN2 coated nylon 6, respectively.



**Fig. S11** (a) HR-TEM image of 10 *f*-GO-BN/S-IPN2 along with the SAED pattern, (b) high magnification HR-TEM image of 10 *f*-GO-BN/S-IPN2 showing the parallel fringes.



**Fig. S12** Analysis of the peel-off adhesion test of coated nylon 6 samples as per ASTM D3359-17. (For 5 *f*-GO-BN/S-IPN2 and 10 *f*-GO-BN/S-IPN2, the images were taken after the cryogenic adhesion test.)



**Fig. S13** Schematic representation of some of the possible interactions in the nanocomposite coating.

Table S1 Atomic compositions of GO, *f*-GO, BN, oxidized BN and *f*-GO-BN as obtained from the XPS.

Sample	Atomic %				
	C1s	O1s	B1s	N1s	Si2p
GO	62.47	37.53	-	-	-
<i>f</i> -GO	62.94	26.79	-	5.34	4.92
BN	3.51	1.51	52.53	42.45	-
Oxidized BN	8.74	3.01	48.36	39.7	-
<i>f</i> -GO-BN	28.93	9.78	32.34	27.37	1.58

Table S2. H<sub>2</sub>GTR values of this work and their comparison with reported graphene-based polymer coated films.

Films	Filler loading	Substrate/ Coating method	H <sub>2</sub> GTR <sup>a</sup>	% of decrease of H <sub>2</sub> GTR compared to substrate	Permeability coefficient <sup>b</sup>
PU/TET-mGO <sup>1</sup>	22 wt%	Nylon/ Spray coating	12.8	93	0.15
PU/RGO-HA <sup>2</sup>	43.3 wt%	Nylon/ Spray coating	32	82	0.40
MAPP/mGO-ODA <sup>3</sup>	60 wt%	Nylon/ Spray coating	10.6	94	0.26
EVOH/BA/GO <sup>4</sup>	5 wt%	Nylon/ Spray coating	6.9	96.1	~0.15
PVOH/GO <sup>5</sup>	80 wt%	PET/ Spray coating	5	96	0.56
PEI/GO <sup>6</sup>	-	PET/ Layer-by-layer assembly	145	55.5	-
PEI/RGO <sup>7</sup>	12.5 wt%	PET/ Vacuum filtration	~40.3	87	~6.2
PDDA/SPVDF-GO <sup>8</sup>	2 wt%	PET/ Layer-by-layer assembly	11.7	96	3.1
HPEI/rGO <sup>9</sup>	16.6 wt%	PET/ Spray coating	8.0	95	~0.96
PEI/GO <sup>10</sup>	-	PET/ Layer-by-layer assembly	158.1	41	~0.04
Chitosan/PAA/rGO <sup>11</sup>	-	PET/ Layer-by-layer assembly	102.3	44	-
EAA/MXene-GO <sup>12</sup>	10 wt%	Nylon/ Spray coating	2.01	89	~0.03
S-IPN2/ <i>f</i> -GO-BN (Present work)	5 and 10wt%	Nylon/ Spray coating	0.9 and 0.5	95 and 97	0.01 and 0.006

<sup>a</sup>H<sub>2</sub>GTR (cc.m<sup>-2</sup>.d<sup>-1</sup> .atm<sup>-1</sup>) of the film.

<sup>b</sup>H<sub>2</sub> gas permeability coefficient (cc.mm.m<sup>-2</sup>. d<sup>-1</sup> .atm<sup>-1</sup>) of the coating layer.

## References

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