

Supporting information

Increasing the interlaminar fracture toughness and thermal conductivity of carbon fiber/epoxy composites interleaved with carbon nanotube/polyimide composite films

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NEW CARBON MATERIALS

S1 Processing procedure of CNTs

S1.1 Purification of CNTs

Multi-walled carbon nanotubes and concentrated hydrochloric acid were placed in a 250 mL flask, and the mixture was under ultrasonic treatment for 2h. After that, the product is centrifuged several times and rinsed with distilled water the pH value is close to neutral. Finally put the CNTs in a vacuum oven at 80 °C for 2h to remove water.

S1.2 Acidification of CNTs

Purified CNTs 40mL of mixed acid (sulfuric acid (98%): nitric acid (60%)=3:1) were put in a 100mL flask and then under ultrasonic treatment at room temperature for 2h. After diluted with plenty of distilled water, the mixture was filtered with a polytetrafluoroethylene film (pore diameter: 0.22 μ m) filter until the pH value of the filtrate is about 6. Place the resulting carbon nanotubes in a vacuum oven and dry at 80 °C for 24 hours to obtain acidified carbon nanotubes.

S1.c Characterization of CNTs

The structure of CNTs before and after acidification treatment was characterized by FT-IR and XPS. As shown in **Figure S1(a)**, the FT-IR spectrum of acidified CNTs presented absorption peak at 1548 cm^{-1} (stretching vibration of C-C on the surface of CNTs), 1116 cm^{-1} (stretching vibration of the C-C skeleton). The absorption peaks at 2928 cm^{-1} and 2858 cm^{-1} were significantly enhanced compared with spectrum of purified CNTs, indicating that the graphitized structure of the CNTs' surface was destroyed. New absorption peaks appeared: 1617 cm^{-1} (symmetric stretching vibration absorption peak of C=O in the carboxyl group); 1734 cm^{-1} (asymmetric stretching vibration absorption peak of C=O in the carboxyl group); 3420 cm^{-1} (stretching vibration absorption peak of the O-H bond in the carboxyl group). The appearance of these characteristic peaks indicates that carboxyl functional groups were generated on the surface of CNTs after acidification treatment. As shown in Figure S1(b), in addition to the C1s peak of carbon atoms, there are also O1s peaks of oxygen atoms on the surface of acidified CNTs, indicating that oxygen containing functional groups are introduced into the surface of CNTs during the acidification process. The C1s peak of carbon atoms was finely scanned with high resolution. As shown in Figure S1(c), the XPS curve of the purified CNTs showed C SP² peak (284.2 eV), C SP³ peak (285.1 eV), and π - π^* peak (290.7 eV) generated by the spontaneous electron transition in the graphite sheet structure of the CNTs surface. As shown in Figure S1(d), new peaks appeared in the XPS curve of acidified CNTs, including the C1s peak (287.1 eV) of the

C=O, the C1s peak (288.9 eV) of the COOH group, and the C1s peak (286.2 eV) related to hydroxyl groups of the C-OH. These results indicate that carboxyl groups are generated on the surface of CNTs after acidification.

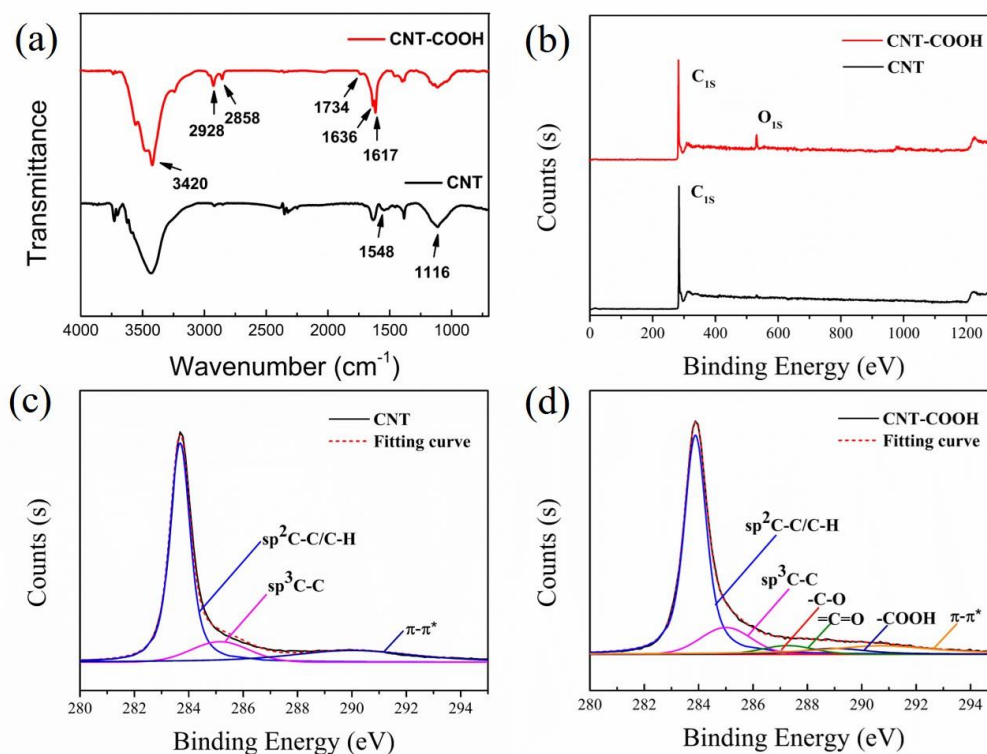


Fig. S1 FT-IR spectrum (a) of CNT and CNT-COOH, XPS survey scan of CNT and CNT-COOH (b), XPS spectrum of CNT (c) and CNT-COOH (d)

S2 SEM micrographs of BOH-2.0CNT film toughened laminate

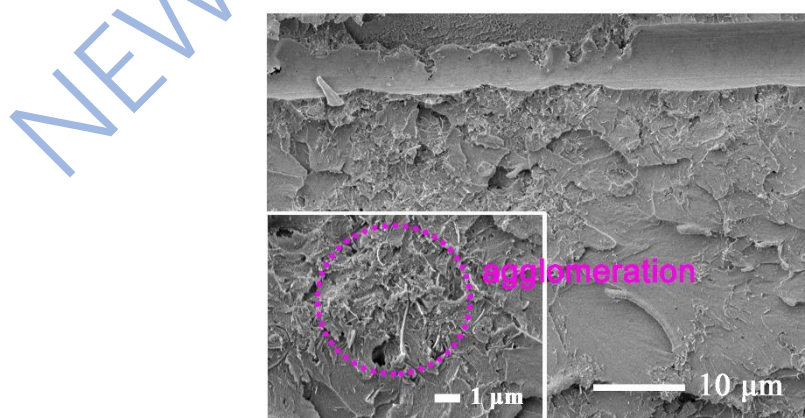


Fig. S2 SEM micrographs of the fracture surface of BOH-2.0CNT film toughened laminate under mode I loading.

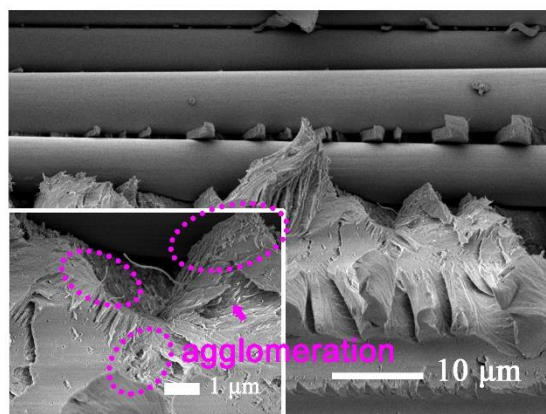


Fig. S3 SEM micrographs of the fracture surface of BOH-2.0CNT film toughened laminate under mode II loading.

S3 Comparison of the G_{IC} , G_{IIC} , and flexural strength

Table S1 Comparison of the G_{IC} , G_{IIC} and flexural strength.

Interleaf	$G_{IC-NL}/J \cdot m^{-2}$ (Gain)	$G_{IC-Prop}/J \cdot m^{-2}$ (Gain)	$G_{IIC}/J \cdot m^{-2}$ (Gain)	Flexural strength /MPa (Gain)	Ref
PEK-C nanofiber	241 (+65%)	376 (+82%)	-	-	[4]
PSF/CNC nanofibers	-	1120 (+29%)	2440 (+49%)	-	[5]
Sprayed MWCNT	996 (+39%)	748 (+22%)	-	-	[10]
CNT films	-	1380 (+100%)	-	-	[23]
Bucky paper	-	-	2400 (+104%)	800 (-10%)	[24]
CNT veils	-	440(+60%)	-	-	[25]
CNTs/epoxy films	-	-	452 (+126%)	-	[26]
GPS-CNTs	228 (+79%)	1201 (+95%)	1078 (+91%)	-	[29]
CNT sheets	300 (+57%)	690 (+30%)	-	1505 (+49%)	[33]
BOH-0.5%CNT	150(+107%)	505 (+260%)	2708 (+220%)	1530 (+8.3%)	This work

S4 Thermal conductivity of CNT/BOH film toughened laminates

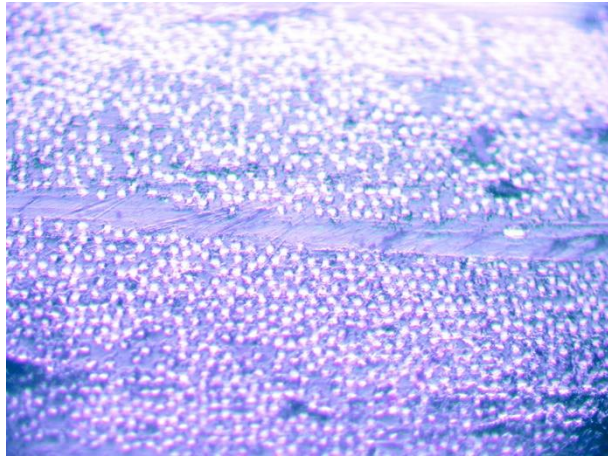


Fig. S4 Optical photo of cross section of BOH film toughened laminate

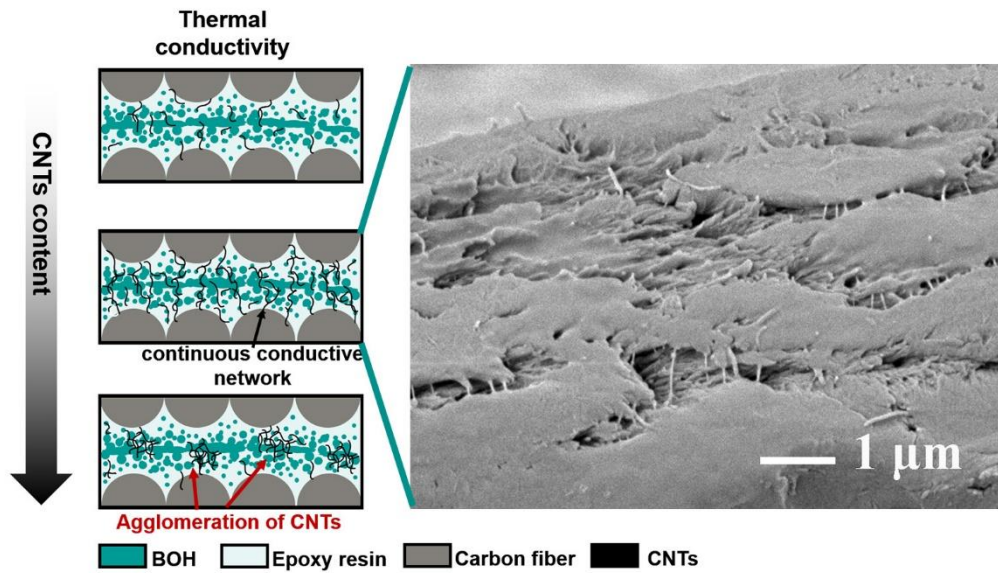


Fig. S5 Illustration of thermal conducting mechanism for CNT/BOH film toughened CF/EP laminates