Supplementary material

The enlarged boundary SEM images of TFBT sample between the CF bundle and epoxy were added behind Figure S1(a) and (b). Firstly, the discontinuous boundary can be seen in Figure S1(c) marked by the red dashed line. Secondly, the high magnification image of boundary was shown in Figure S1(d) for determining the boundary, where the brittle carbon fiber was marked by the red circle and the flat epoxy resin was marked by the red rectangle. In one word, the boundary could be identified via zoom-in Figure S1(c) and (d). Finally, the thickness of CF bundle could be measured according to the boundary lines shown in Figure S1(c). Finally, the art image of Figure S1(b) is given in Figure S1(e), where the boundary between the black and grey zone clearly represents the boundary of carbon fiber and matrix (Tables S2–S6) (Figures S2–S4).

The thermal behavior and fiber volume were carried out by the thermogravimetric analysis (TGA8000, PerkinElmer Instruments) using TGA800 in a flowing nitrogen atmosphere from ambient temperature to 800 °C with a heating rate of 20 °C/min.

The fiber volume (V_f) of composite could be calculated as follows:

$$V_f = \frac{\frac{W_f}{\rho_f}}{\frac{W_f}{\rho_f} + \frac{1 - W_f}{\rho_m}} \tag{1}$$

where V_f is the fiber weight fraction obtained by the TGA result, ρ_f and ρ_m are the density of carbon fiber and resin matrix respectively. The volume fraction of composites is summarized in Table S1.

Table S1: The main experimental materials

Material	Specification	Purity	Supplier
Epoxy	RS-M135	Analytical purity	PRF composite materials
Hardener	RS-MH137	Analytical purity	PRF composite materials
p-CNTs	C139834-5g	>95%	Macklin
C-CNTs	C835676-5g	>95%	Aladdin
Gelatin	C41087-500g	>99%	Shanghai yuanye Bio-Technology Co., Ltd
NaOH	1310-73-2	Analytical purity	Sinopharm Group
Acetone	776-99-8	Analytical purity	Sinopharm Group
Ethanol	64-17-5	Analytical purity	Sinopharm Group
Filter membrane	2 μm	Analytical purity	Shanghai New Asia
Release agent	Easy-Lease200	Analytical purity	Easycomposites

Table S2: The IFNS of TFBT samples *via* fiber enhancement

Nanofiller	Concentration/ mg mL^{-1}	Time/min	IFNS/ MPa	Error/ MPa
C-CNTs	0.05	1	24.23	4.83
		2	24.85	4.52
		5	26.79	5.87
	0.1	1	28.28	3.48
		2	26.27	2.54
		5	26.94	4.52
g-CNTs	0.05	1	22.53	2.16
		2	26.91	2.21
		5	20.76	1.51
	0.1	1	26.13	2.66
		2	31.49	2.66
		5	28.62	2.29

Table S3: The IFNS of TFBT samples *via* matrix enhancement

Nanofiller	Loading content/wt%	IFNS/MPa	Error/MPa
Neat	—	22.44	2.10
p-CNTs	0.05	24.09	3.71
	0.1	26.20	3.58
	0.2	18.48	6.03
C-CNTs	0.05	27.18	4.58
	0.1	27.50	2.42
	0.2	26.61	5.82
g-CNTs	0.05	27.98	4.72
	0.1	29.50	2.22
	0.2	28.61	2.29

Table S4: The volume fraction of composites

Composite	V_f/%
Sample 1	52.16
Sample 2	55.04
Sample 3	52.68
Sample 4	54.16
Average	53.51

Table S5: Flexural properties various composites via matrix enhancement

Composite	Loading content/ wt%	Strength/ MPa	Error/ MPa	Modulus/ GPa	Error/GPa
Neat	—	679.23	51.70	52.78	5.04
CF/p-	0.05	661.94	26.86	50.36	4.04
CNTs@EP	0.1	675.93	15.53	58.98	2.51
	0.2	651.23	28.00	44.59	3.47
CF/C-	0.05	674.78	3.84	49.56	3.96
CNTs@EP	0.1	730.51	30.47	57.86	4.50
	0.2	703.04	53.16	57.52	5.25
CF/g-	0.05	783.20	30.33	55.25	3.68
CNTs@EP	0.1	813.43	39.98	67.83	4.55
	0.2	769.25	11.75	61.93	4.06

Table S6: Comparison of flexural properties between matrix enhancement and fiber enhancement composite

Enhancement	Composite	Parameters	Strength/MPa	Error/MPa	Modulus/GPa	Error/GPa
—	Neat	—	679.23	51.69	52.78	5.04
Matrix enhancement	CF/C-CNTs@EP	0.1wt %	730.50	30.48	57.87	4.45
	CF/g-CNTs@EP	0.1 wt%	813.43	39.99	67.83	4.55
Fiber enhancement	C-CNTs@CF/EP	0.1 mg/mL, 1 min	807.36	17.40	60.13	2.74
	g-CNTs@CF/EP	0.1 mg/mL, 2 min	828.02	30.51	66.14	1.07

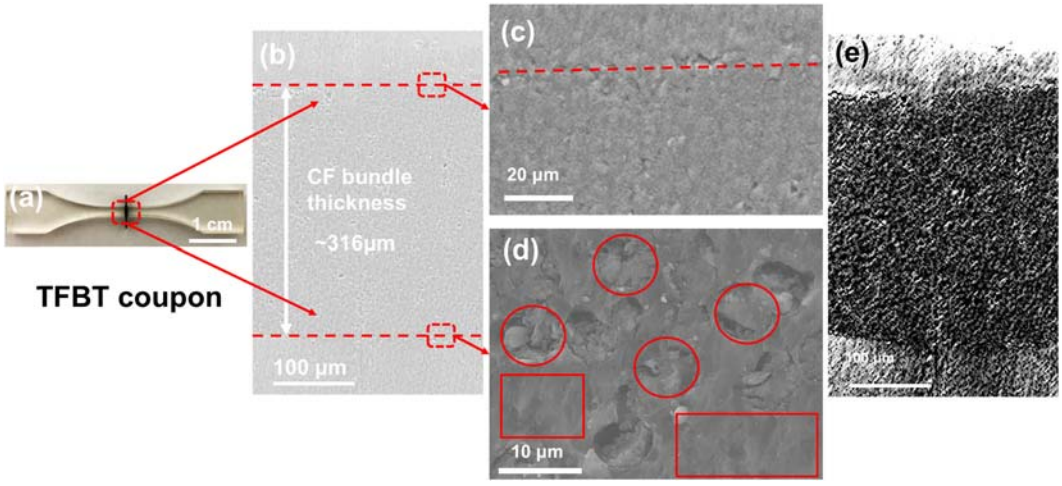

Figure S1: Optical image of TFBT sample (a), cross section SEM image (b), high resolution image of upper (c) and lower (d) boundary between CF bundle and epoxy resin. The art image (e) for image (b).

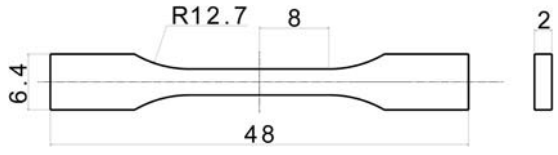

Figure S2: Sample dimensions of transverse fiber bundle tensile test based on ASTM D638, all dimensions are in mm.

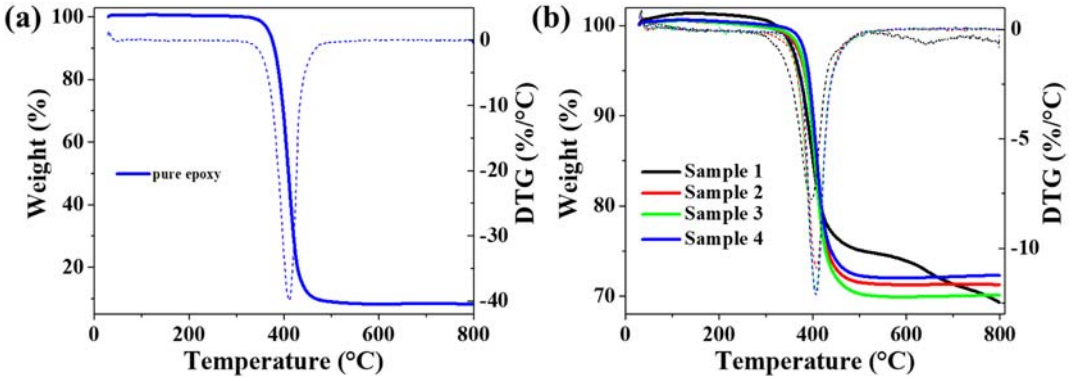

Figure S3: Thermal analysis of pure epoxy (a) and CFRP composite (b).

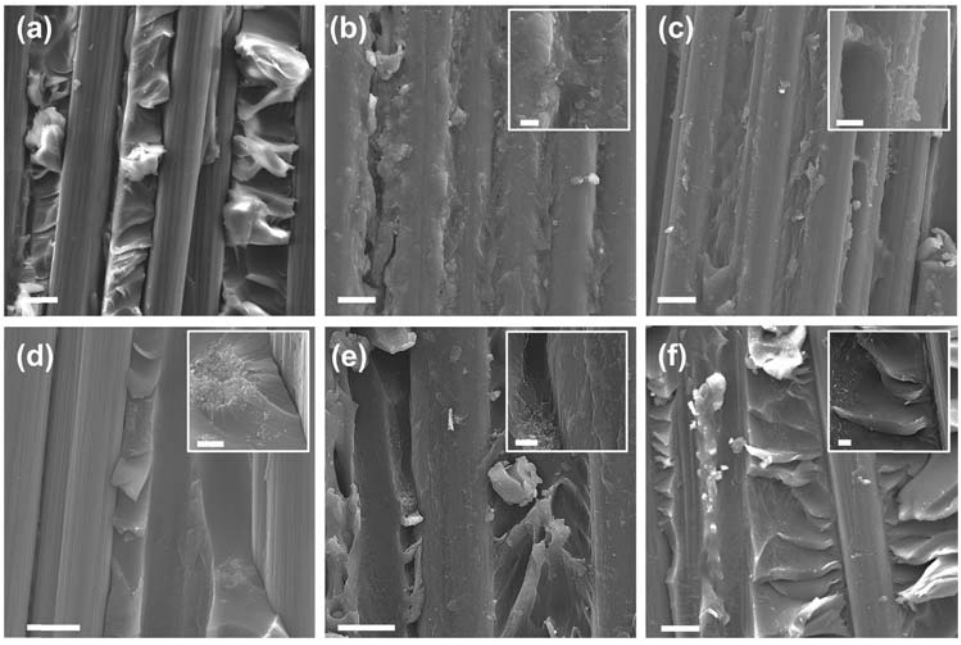

Figure S4: SEM images of various CFRP composites after three-point bending test via fiber enhancement (up) and matrix enhancement (down): pure CFRP (a); g-CNTs@CF/EP (b); C-CNTs@CF/EP (c); CF/p-CNTs@EP (d); CF/g-CNTs@EP (e); CF/C-CNTs@EP (f) composite. All the composites with the same nanofiller loading of 0.1 wt%. The scale bar of main and insert images are 5 and 1 μm, respectively.