Graphene oxide-mediated thermo-reversible bonds and in-situ grown nano-rods trigger ‘self-healable’ interfaces in carbon fiber laminates

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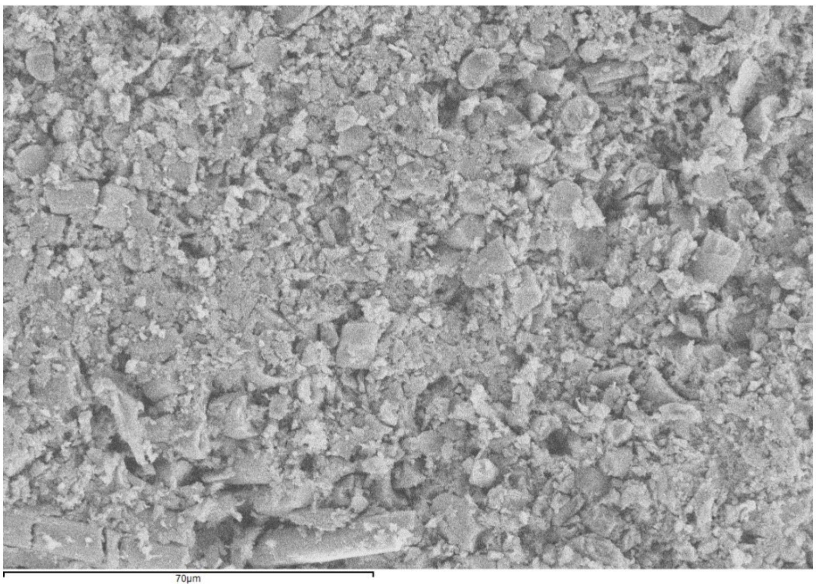
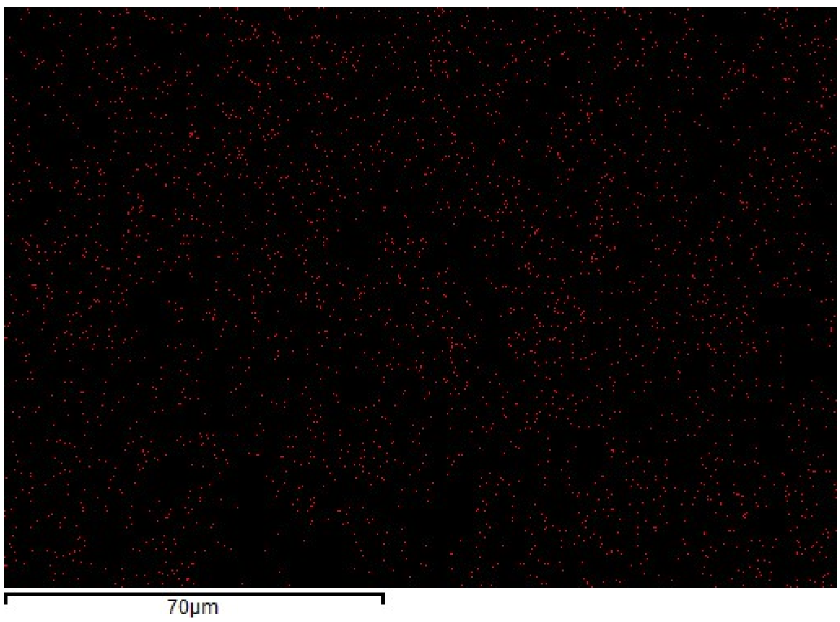
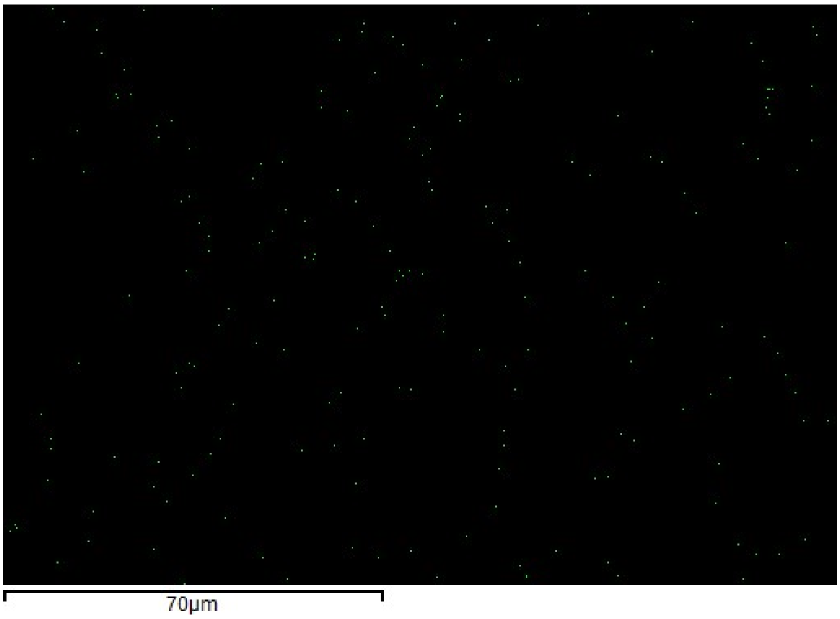
S1: EDX area mapped region of ZBCFRE with GO



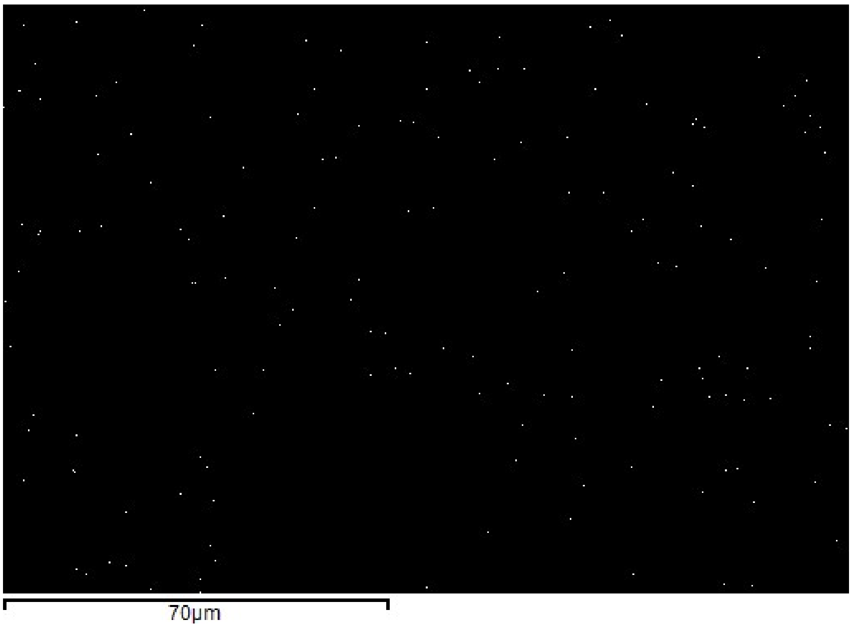
EDX area mapped region of ZBCFRP



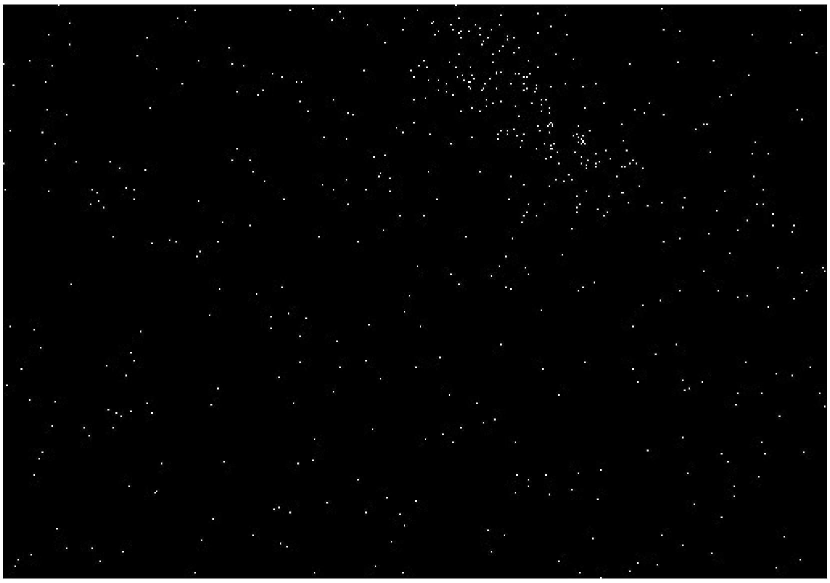
C1 Ka1 spectra



O Ka1 spectra



Si Ka1 spectra



Zn La1 spectra

Figure S1: EDX spectroscopic points representing the presence of various elements in the imaged ZBCFRE with GO.

S2: ZnO concentration optimisation with respect to seeding times

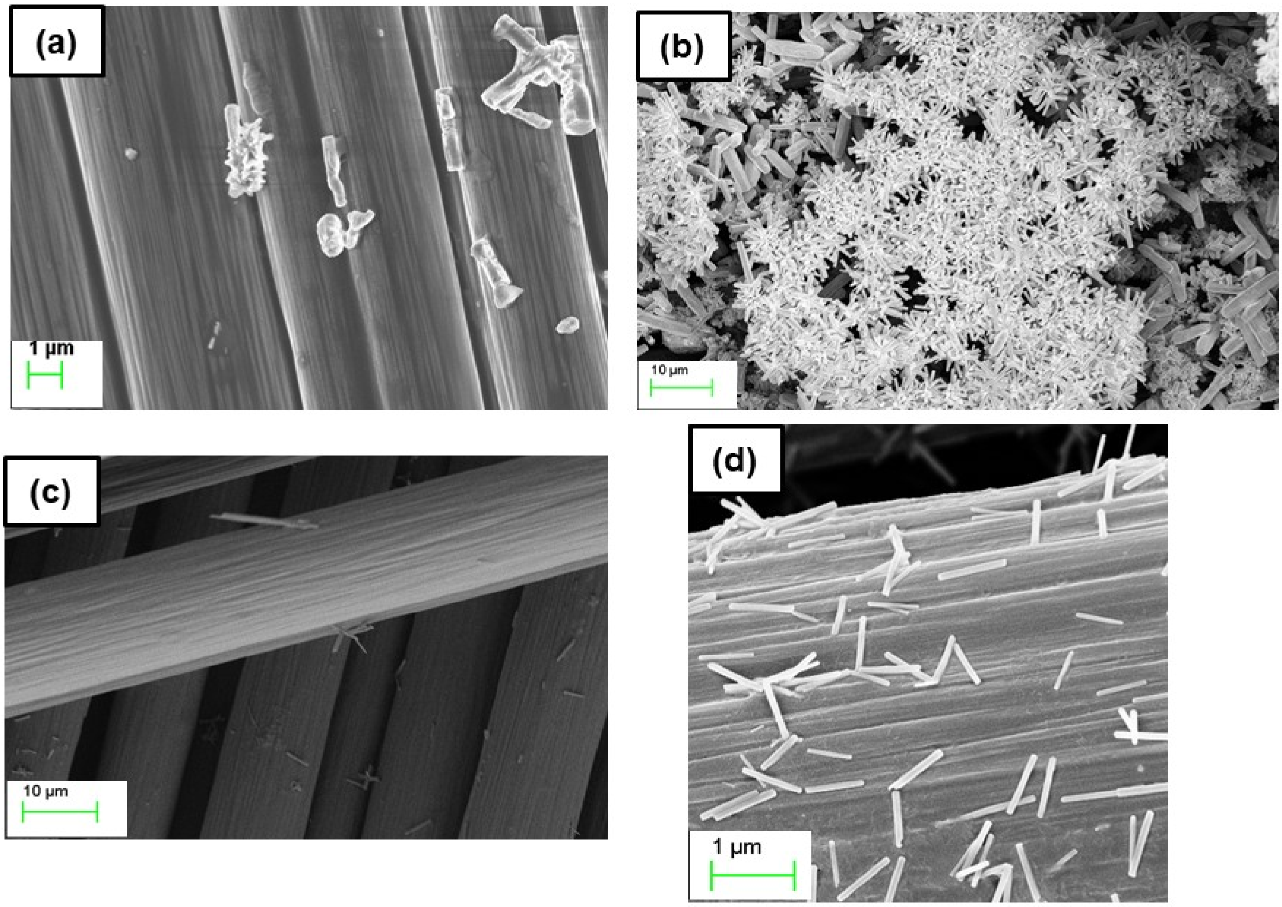


Figure S2: Growth of ZnO nano-rods on CF after (a) 5 min (b) 40 min and (C) 25 min of seed solution adsorption (d) Zoomed in image of (c) depicting the uniformity and distribution of ZnO nano-rods

S3: Characterisations of neat CF and ZnO-CF

The available literature on graphitic materials has confirmed a Raman band at ~1585 cm^{-1} related to the C-C bond vibrations and is present in all CFs. Also, the Raman band at ~1330 cm^{-1} is associated with the boundaries of the graphitic crystals and is related to the structural disorder effects. This band is visible in the case of poorly graphitized fibers. The ~1330 and ~1585 cm^{-1} are assigned as D- and G-bands, respectively.

Raman spectra performed on both the neat and ZnO-CF indicated the presence of both the D and G bands. It was observed that while neat CF showed an I_d/I_g value of 0.93, ZnO-CF had its value increased to 1.02. Higher I_d/I_g ratio in ZnO-CF indicated that the sp^2 bonds of the graphitized crystals were broken and had transitioned to sp^3 hybridization. Thus, the growth of ZnO over CFs via ionic treatment showed oxidation of the fiber surface to an extent and an increase in unsaturated active sites on the neat fibers that could lead to better interaction of the modified fibers with the matrix, thereby improving the matrix adhesion over the fibers.

AFM images of the neat CF and ZnO-CF showed that ZnO-CF had a rugged topography with a mean square roughness (Ra) of ZnO-CF around 20 times higher than that of neat CF. This uneven and higher surface area of the ZnO-CF justified their better wetting capability with the epoxy matrix through more points of contact between the matrix and the fiber, implying higher interfacial adhesion.

TGA analysis was conducted for both neat CF and ZnO-CF showed that initial mass of ZnO-CF was higher than neat CF for the exact dimensions by approximately 1% due to the growth of ZnO particles. However, ZnO-CF showed higher mass loss than that of neat CF. ZnO-CF showed a three-stepped degradation profile. The mass loss up to 100 °C can be attributed to the moisture loss. The mass loss up to about 300 °C can be attributed to the loss of volatilities situated around the ZnO nano-rods during the modification of CF. The third stage of degradation above 300 °C was indicative of the fiber degradation. Neat CFs did not show staged degradation, indicating that the fibers without any surface conditioning were almost inert in nature. ZnO-CF showed a mass loss of 3% higher than neat CF after 650 °C implying that the ZnO rod's interaction with the fibers post chemical modification reduced the inertness of the fibers allowing for higher susceptibility of atmospheric interaction. Figure S3 showed the raman, TGA and AFM characteristics of neat CF and ZnO-CF respectively.

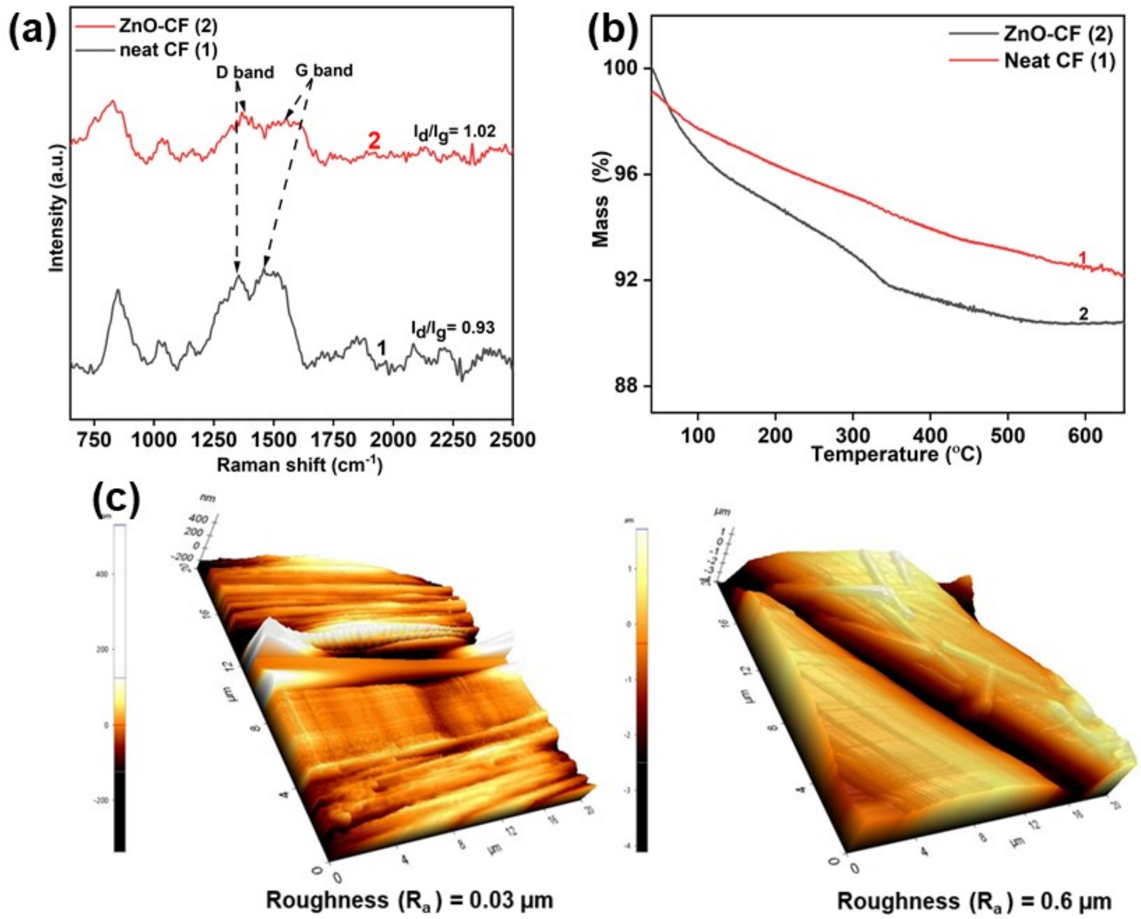


Figure S3: (a) Raman analysis (b) TGA analysis and (c) AFM analysis of neat CF and ZnO-CF

Table S1: Percentage improvement of mechanical property in the laminates with modified architectures

Material/ Property	**Neat CFRE (MPa)**	**ZCFRE with (absolute/increment)**		**ZBCFRE with (absolute/increment)**	
		0.2 wt. % GO (MPa)	**0.5 wt. % GO (MPa)**	**0.2 wt. % GO (MPa)**	**0.5 wt. % GO (MPa)**
FS	556 ± 18	652 ± 42 (17%)	805 ± 52 (45%)	706 ± 40 (27%)	815 ± 73 (46%)

ILSS	46 ±2	57 ± 4 (24%)	60 ± 5 (30%)	60 ± 3 (30%)	61 ± 6 (33%)