Manned deep-space exploration requires structural materials with performance beyond the current state-of-the-art carbon fiber (CF) reinforced plastics (CFRP). Carbon nanotube (CNT) composites offer the potential to surpass CFRP in both mechanical and transport properties, however fundamental challenges remain in achieving high CNT volume fraction, high alignment, and scalability. This research addresses these challenges through seven objectives: characterization of continuous CNT yarns; development of techniques to manufacture both unidirectional and quasi-isotropic laminates; evaluation of functionalization treatments; and assessment of transport properties. A graphitization technique for CNT yarn carbon/carbon (C/C) composite laminates is demonstrated, and modeling is used to establish microstructure–property relationships.

The CNT yarn laminates achieve ultra-high reinforcement volume fractions (~90 wt.%) and can incorporate CNT aerogel to reduce anisotropy. A 150 mm square [0/90/±45]s quasi-isotropic laminate was fabricated to demonstrate scalability. Among functionalization treatments, gamma-ray irradiation at 700 kGy yielded the highest specific modulus of 258 GPa g⁻¹ cm³, while the electron beam treatment achieved the highest specific strength of 2.293 GPa g⁻¹ cm³. Transport properties also exceeded CFRP, with in-plane thermal conductivity of 75.78 W m⁻¹ K⁻¹, and longitudinal electrical conductivity of 4,545 S cm⁻¹. These values translated to EMI shielding effectiveness exceeding 100 dB in the X-band. Graphitized C/C laminates exhibited a unique toughened graphite microstructure with a thermal anisotropy ratio of 167.

Overall, these results demonstrate the potential of CNT yarn laminates as intrinsically multifunctional structural materials for next-generation aerospace vehicles.