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Interface engineering of carbon fiber composites using CNT: A review

Renuka Sahu[1] | Sathiskumar A. Ponnusami[2] | Christian Weimer[3] | Dineshkumar Harursampath[1]

[1]Department of Aerospace Engineering, Indian Institute of Science, Bangalore, India

[2]Department of Mechanical Engineering and Aeronautics, University of London, London, UK

[3]Central Research and Technology (CRT), Airbus Defence and Space Gmbh, Taufkirchen, Germany

Correspondence
Sathiskumar A. Ponnusami, Department of Mechanical Engineering and Aeronautics, University of London, London, UK.
Email: sathiskumar.ponnusami@city.ac.uk

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Abstract

This paper aims to explore the potential of carbon nanotubes (CNTs) in enhancing the structural capability and multifunctionality of carbon fiber composites in aerospace applications, primarily by focusing on interfacial applications. The conventional method of dispersing CNTs in a matrix is not fully efficient in exploiting the mechanical and multifunctional performance of CNTs. Hence, the use of CNTs at the interface or as a coating on the surface of carbon fibers has been suggested as a means of achieving multifunctionality, in addition to enhanced mechanical performance. The paper presents an overview of the various processes for growing CNTs on carbon fiber surfaces and examines the effects of CNT geometry and growth parameters on the properties of grafted fibers and their composites. Furthermore, it discusses the potential improvements in thermal and electrical conductivity achievable by incorporating CNTs at the interface, as well as the benefits of using CNTs as a sizing layer for carbon fibers, including enhanced fracture toughness and resistance to delamination.

Highlights

- Comprehensive study of interface engineering in carbon fibers and Carbon Fibre Reinforced Plastic (CFRPs) using carbon nanotubes (CNTs).
- Improved transverse mechanical properties and overall thermal and electrical properties.
- Multifunctional applications possible with the use of CNT.
- Both experimental and numerical studies reviewed.

KEYWORDS

carbon fiber, CNT, effective properties, electrical and thermal properties, fuzzy fiber, interface

1 | INTRODUCTION

Carbon nanotubes (CNTs) have recently become one of the most sought-after materials due to their superior mechanical,[1–4] electrical,[5,6] electronic,[4,7] and thermal properties,[3,4,8] as well as their lightweight[9–11] and structural superiority.[12] CNTs are versatile enough to be used in a variety of industries, including electronics,[11,13] medicine,[14–16] aerospace,[13,17,18] defense,[19] chemical industry, and waste treatment.[20,21] CNTs and their nanostructures have seen a lot of development and scientific exploration since their discovery in 1991. The order of all three dimensions, length, radius, and thickness, differ due to the size, shape, and inherent unique geometry of CNT. The elastic properties of CNT are found to be dependent on its geometrical parameters, particularly thickness.[22,23] Additionally, it has been hypothesized that the CNT's length affects its thermal characteristics.[24] Numerous studies have been conducted on double and multi-walled CNTs (DWCNTs and MWCNTs respectively) where interatomic interaction come into play.[25] Researchers have employed various approaches, such as molecular dynamics (MD), molecular structural mechanics (MSM), atomistic finite element methods (FEM), and even continuum approaches that utilize non-local theories, to study the interatomic effects influencing the overall behavior of CNTs.[26–30]

Experimental studies have shown that CNT has a Young's modulus of up to 1.2 TPa.[31] Shen and Li[32] determined the Young's modulus, shear modulus, transverse elastic modulus, and Poisson's ratio by examining the transversely isotropic properties of CNT. Due to these superior property values, the use of CNT as reinforcement, nanostructures, and other chemical components is of great interest. One application of CNT is in composites. There are two methods for adding CNTs to a composite: direct reinforcement, in which CNTs are dispersed throughout the matrix,[33–36] and grafting onto the fiber, in which CNTs are grown on the outer surface of the fiber. Such fibers are known as fuzzy fibers, and a matrix is used to cure them[37] to obtain fuzzy fiber reinforced composites (FFRC). Aligned CNT composites have also been studied, and it has been found that experimentally controlled aligned CNT reinforcement provides greater property enhancement than random CNT dispersion and orientation in an epoxy matrix.[38] Marconnet et al.[39] examined aligned CNT-reinforced epoxy nanocomposites to determine the impact of CNT volume fraction on the thermal conductivity of the whole nanocomposite. They found that at a volume fraction of 16.7%, the axial thermal conductivity increases by a factor of 18.5. The waviness of CNTs was also considered in a different study on aligned polymer nanocomposites.[40,41] For a system of aligned CNTs composed of MWCNTs with an outer diameter of around 8 nm, the waviness ratio significantly affected the elastic modulus with an increasing waviness ratio causing up to three-fold decrease in the elastic modulus. The reduction of effective elastic moduli up to 100 times has been observed due to CNT waviness.[42]

Several works investigate growing CNTs onto the fibers, be it metal fibers like aluminium[43–45] or ceramic,[46] carbon[47] or glass fibers.[48] Some work also studied growing nanofiber on carbon fibers.[44,49] In the case of metal fibers, a significant improvement has been observed in the mechanical properties of the reinforced fiber and the composite. Whereas for glass and carbon fibers, a decrease in the tensile strength has been noted when treated with CNT. This reduction in tensile strength has been attributed to the creation of surface voids and defects upon removing the protective fiber sizing, which is done in order to graft CNTs efficiently onto the fiber surface. However, this reduction is not significant, and in many cases, using certain processing techniques like electrophoretic deposition (EPD) or electrospray technique do not cause reduction in carbon fiber tensile strength. For glass fibers as well, when CNTs were grafted onto the outer surface, the electrical properties of the composite along with the fracture toughness were significantly improved.[50]

Grafting CNT onto the surface of carbon fibers poses significant challenges,[51] with the primary concern being achieving the required CNT growth density and type on the carbon fiber surface. Several promising methods, including chemical vapor deposition (CVD), EPD, and in-situ CNT growth[52] have been investigated. However, scaling these techniques for industrial purposes presents challenges since the goal is to develop fuzzy fibers for commercial applications. To achieve this, an industry-level preparation methodology is necessary. Some cases have been discussed in subsequent sections to shed light on this issue.

It has been shown in literature that interface engineering of conventional fiber matrix composites also helps to enhance the failure strength, debonding and changing the fracture mode of the composite. Delamination can be avoided by using FFRCs. Studies also suggest the use of CNTs between the laminates to get better interlaminar shear strength (ILSS) and strength properties of the laminated composites.[52,53] Villoria et al.[54] used CNTs at the laminate interface to increase interlaminar toughness. There has been evidence of changes in fiber-reinforced composites' failure behavior going from debonding to changing to fiber failure upon the integration of CNTs at interface of composite laminae. Thakre et al.[55] introduced SWCNTs in the interlaminar region of 3-D woven carbon fiber epoxy composite to improve the interlaminar fracture behavior. Mode-I delamination behavior was investigated. It helped in understanding the differences in mechanism of delamination when functionalized and unfunctionalized (pristine) nanotubes are added to the woven carbon fiber-epoxy matrix composite laminates. Jakubinek et al.[17] investigated the use of CNT as structural and conductive adhesives for aerospace applications. These works add only to the support of the argument of using CNTs in the interface of the composites. The effect of grafting strength and density on the mechanical properties of the composite lamina developed was also studied.[56,57]

The current review paper discusses various characterization techniques, experimental and computational, to estimate the mechanical, thermal, and electrical properties of FFRCs. The paper is organized as follows: Section 1 gives the introduction. Section 2 highlights the various methods used for developing fuzzy fibers. Experimental studies focusing on FFRCs are discussed in Section 3. Section 4 gives insights into the numerical modeling techniques used for the property determination of FFRCs. Finally, Section 5 gives the gap analysis and Section 6 the conclusion.

2 | METHODS TO DEVELOP FUZZY FIBERS

The most challenging aspect of utilizing CNTs is their synthesis, as the dimensions of CNTs lie close to the atomic scale, the processing is highly demanding. Developing CNTs requires strict control and monitoring of processing parameters, which are also capital-intensive. These factors have been major obstacles to the commercial growth of CNT-based products and technologies. Unfortunately, there has been no significant development in large-scale, industry-based production of bulk CNTs. Although there have been some suggestions for production-level adoption based on laboratory-developed methods,[58–60] these are still being studied. Current processing methods used in literature to develop fuzzy fibers can be broadly summarized as follows.

2.1 | Chemical vapor deposition

Chemical vapor deposition (CVD) is the most widely used processing technique to synthesize individual CNTs or grow CNT forest onto a substrate[61–64] (Figure 1). It is a chemical reaction-based process of material deposition/generation in the form of crystals, film or powder and works by modifying the bonding between the chemical constituents. The bond developed in a CVD process is due to physical adsorption and is of weak type that is, van der Waals (vdW) bond.[65] In CVD, acetylene gas or

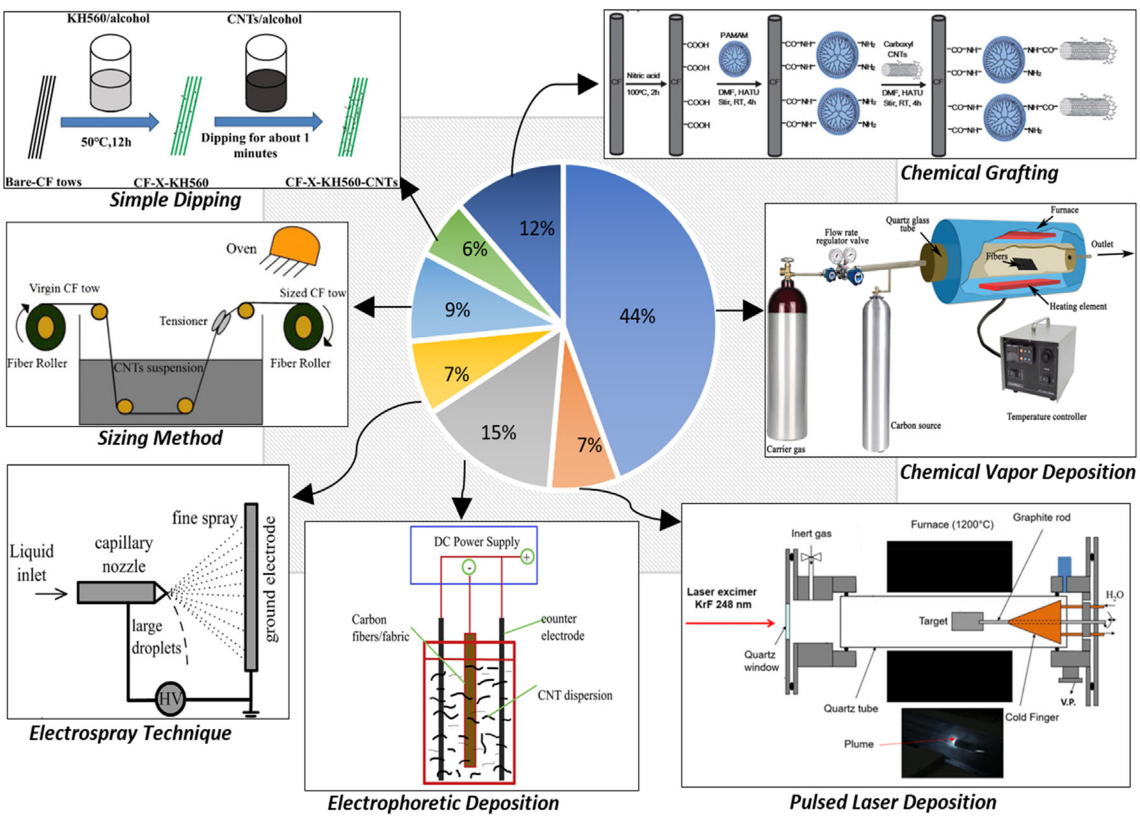

FIGURE 1 Graphical representation of distribution of various production techniques used in literature along with schematic representation of each technique.[65,69,73,85,89]

any other hydrocarbon is usually used, and the decomposition reaction occurs in the presence of a heated substrate and catalysts (usually iron or nickel) inside a pressurized chamber. Silicon wafer is used as a substrate onto which individual carbon starts depositing. Reaction parameters such as temperature, reactive gases used, and type of catalyst influence the material type and distribution.[66] It has been shown that the size of catalysts affects the size of CNTs deposited onto the substrate.[61,63] Smaller catalyst sizes give uniform, finer CNT deposition with fewer CNT walls. It is also possible to go for selective deposition, localized treatment, and deposition onto substrates with different geometry using CVD. The CVD processes are carried out at a temperature range of 80–1020°C.[64,67–69] Modifying carbon fiber surface using treatments like modification with carboxylic acid groups for better adhesion of CNT onto the surface can also be done.[70] CVD is known to produce uniform and dense CNT growth in the range $2–6 \times 10^9$ cm^2,[62] length ~10–30 μm to few mm, width ~50–120 nm[63] and diameter ranging from 5 to 30 nm. CVD offers the advantage of highly pure, uniform, and conformal coatings on a wide range and size of materials. However, there are certain disadvantages associated such as complex equipment and setup and slower deposition rates. Moreover, higher temperature associated with the process can lead to damage/diminishing of inherent properties of the substrate.

2.2 | Pulsed laser deposition

Pulsed laser deposition (PLD) is a physical vapor deposition process wherein a high-power laser source is used to produce vapors of the material to be deposited as opposed to the chemical reaction in the CVD process (Figure 1). This technique has been used to synthesize nanotubes[71] and nanopowders.[72] PLD gives the advantage of producing fuzzy fibers at lower temperatures compared to CVD. PLD temperature range is about 25–600°C,[73] which helps prevent thermal damage to carbon fibers. In addition, there is minimal damage to the quality of CNTs produced.[74] MWCNTs with around 5–10 tube walls and an inner diameter in a range of 2.5–4 nm have been developed using PLD. The length of these MWCNTs was about 100 nm.[75] Since no additional catalysts are needed, the deposition done using PLD is high quality, and the chances of contamination are less. However, they have the disadvantage of huge equipment size and energy consumption.[76]

2.3 | Electrophoretic deposition

Electrophoretic deposition (EPD) process is based on the movement and deposition of charged particles under an electric field onto a conductive electrode to develop thin or thick films and coatings (Figure 1). EPD process can be considered to comprise of two steps: one involving the movement of charged particles upon application of electric field and the other the deposition of the particles onto the electrode.[69] Due to CNT's electrical responsiveness, EPD is the easiest and most effective process to produce fuzzy fibers. The advantages of the EPD process are the short formation time, simple apparatus, and the ability to produce uniform coatings on complex geometries. Furthermore, EPD offers easy control over film thickness and morphology by simply adjusting depositing time and applied potential.[77] It is useful and convenient method for the generation of 'multi-scale' coating on carbon fiber such as MWCNTs.[78] The use of an annular electrode is also suggested to improve the deposition of CNTs.[79] High purity CNTs of around 95%–99% can be obtained via EPD.[80] However, the choice of substrate is limited in it to be electrically conductive. Also, there is drawback of poor adhesion and post processing is needed to improve adhesion.

2.4 | Electrospray technique

The electrospray method also uses an electric field to achieve a suspension of materials to be deposited onto any surface/substrate (Figure 1). It produces particles of uniform size and has no problem with agglomeration.[69] As with EPD, the electrospray technique is also very cost-effective and could be used to mass produce fuzzy fibers and their composites. Applied voltage, spray nozzle size and distance between the spray nozzle and depositing surface are some parameters affecting the CNT deposition.[69] The electrospray method has produced carbon nanofibre-based electrodes for supercapacitors.[81] It is also possible to develop a forest of MWCNTs using coaxial electrospray to achieve a material with very high electrical conductivity.[82] The tensile strength of carbon fiber is preserved in this process as opposed to CVD.[83] Highly graphitized CNT of diameter ~10–20 nm could be produced without any unnecessary oxidation via the electrospray process.[84] The electrospray technique is limited by the low efficiency of the material deposited to material used and sensitivity to operating parameters such as nozzle geometry, voltage and flow rate.

2.5 | Sizing method

The sizing method is used to prominently deposit a thin film on filaments such as carbon or glass fiber to protect the filament surface from damage during manufacturing.

Sizing materials provide adhesion between the fiber and matrix and ensure better processibility.[78] The sizing method has been adopted for CNTs as well to create fuzzy fibers. Fuzzy fibers can be produced by using mechanical roller-based tows of carbon fiber and immersing them into the liquid suspension containing dispersed CNTs[85] (Figure 1). The sizing process is simple and does not require higher temperatures or heavy equipment.[86] Deposited CNTs have been found to have a length of 0.2–2 μm and a diameter of 20–40 nm.[87] Functionalization of CNT is important for the sizing process as it enhances the binding between the carbon fiber and CNT.[87,88] The process is susceptible to varying deposition with change in environmental parameters. Also, the bonding between CNT and carbon fiber might not be as strong as that achieved using CVD or other methods.

2.6 | Simple dipping

The simple dipping or dip coating process uses a coupling agent to bridge the bond between CNTs and carbon fiber or any other material to be coated. Alcohol suspension of CNTs which has been ultrasonicated to achieve good CNT dispersion, is then used as the dipping medium onto which carbon fibers are immersed[89] (Figure 1). Dip coating techniques have also been used to deposit CNT onto glass fiber[90] and carbon fabric for wearable applications.[91] This is the simplest process for creating fuzzy fibers making it cost effective and a relatively faster method. However, there are problems with uniform deposition and proper attachment of CNT to the fiber surface.

2.7 | Chemical grafting

Chemical grafting uses chemical methods such as radical polymerization or condensation reactions to modify the surface by functional groups or polymers onto a substrate surface, chemically bonded to the surface through covalent bonds. It is used to improve the surface properties of materials, such as adhesion, wettability, biocompatibility, and chemical reactivity. It is evident that with this method, functionalized CNTs are grafted onto the carbon fiber surface,[92] with functional groups such as carboxylic acid[93] and polymeric groups.[65] The usual procedure involves desizing carbon fibers and then dipping them in a polymer functional group solution. Further, functionalized CNTs are introduced in a modified carbon fiber solution to complete the grafting process.[94] Usually, MWCNTs are grafted using this method, and the specifications of CNTs are 30–100 nm in diameter and length around 0.5–20 μm.[94,95] The use of coupling agents to obtain high-density grafting of CNTs is also seen.[96] Chemical grafting provides stronger interactions than physical bonds formed in sizing or other processes and allows precise controllability. However, it can be cost extensive, and only a limited thickness of grafted layers can be achieved, with incomplete homogenous coverage in some cases.[97] Also, the CNT-grafted is not pure and is functionalized.

3 | EXPERIMENTAL TECHNIQUES

This section discusses the various experiments performed on fuzzy fibers and FFRCs to study their behavior when subjected to different external stimuli. The section is subdivided into three parts: mechanical, thermal, and electrical, to showcase effectively the broad range of functionality exhibited by fuzzy fibers.

3.1 | Mechanical properties

Zhi-hui et al.[64] used a modified CVD technique for synthesizing CNT growth on carbon fiber to reduce interdiffusion between the carbon fiber and transition metal catalyst, thereby increasing the catalyst's lifetime. It was also found to give a better mechanical characterization of grafted carbon fibers than normal thermal CVD. The fracture load was reduced by 35% compared to raw fibers for the modified thermal CVD grafted carbon fibers. This reduction was 79% for the case of carbon fibers grafted using a normal thermal CVD process. Peng et al.[65] used a newer way of grafting CNTs onto carbon fiber surface wherein a chemical bond was created between the two materials as opposed to vdW bonding obtained with CVD. Firstly, the carbon surface was coated with Poly (amido amine) (PAMAM) dendrimer, and CNT was further deposited. The FFRCs produced this way had a 111% increase in the interfacial shear strength (IFSS) compared to acid-treated carbon fiber composites.

Zhao et al.[98] compared the mechanical behavior of two kinds of CNT reinforced nanocomposites: composite with randomly dispersed CNT and FFRCs. The fuzzy fiber was developed using EPD, whereas the dispersion of CNT in the matrix was carried out using the ultrasonic dispersion technique. The advantage of using EPD was that there was no significant reduction of the tensile modulus of carbon fibers after grafting CNTs (as in Figure 2). It was also found that compared to carbon fiber composite, the FFRC showed an increase of 24.42% in tensile strength, whereas, for CNT dispersed composite, this increase was only 10.41% (Figure 2). Similarly, the

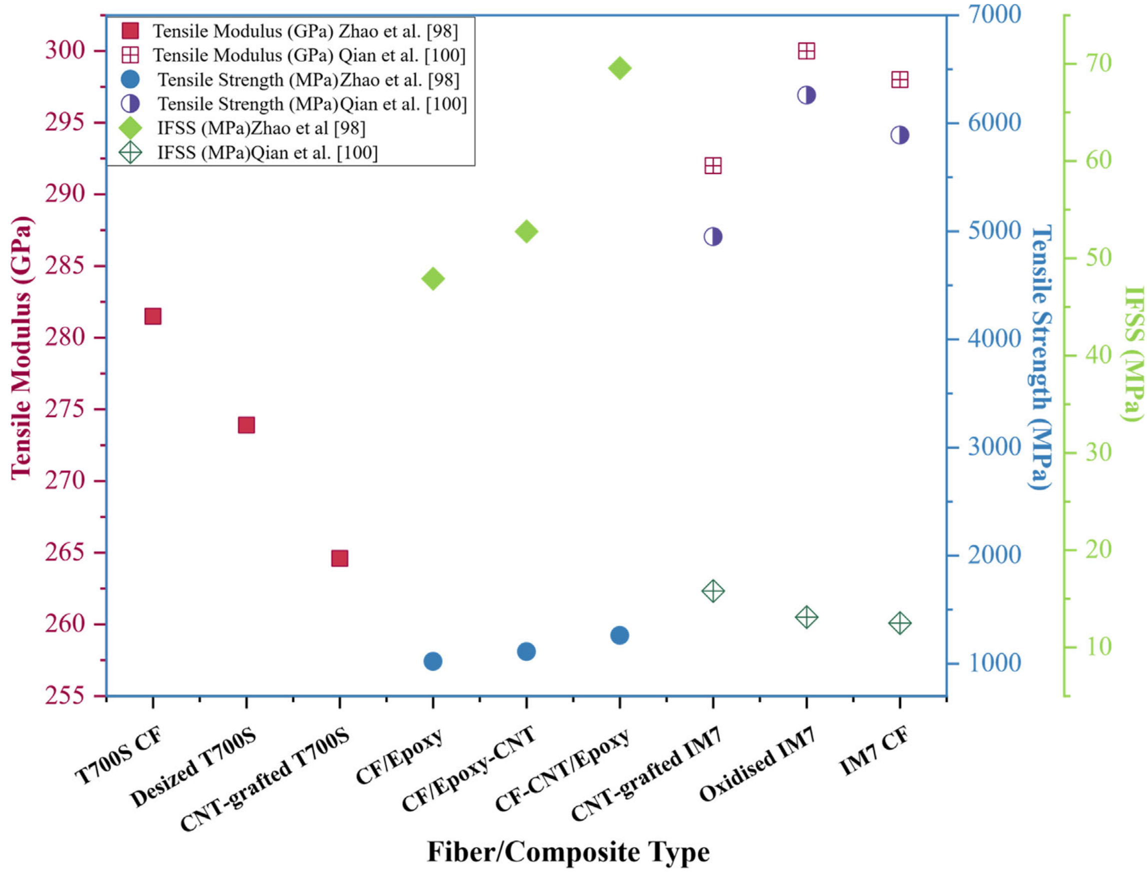

FIGURE 2 Mechanical properties of different composites using normal and carbon nanotube (CNT)-grafted carbon fiber (CF stands for carbon fiber).

flexural strength was improved by 18.43% for the FFRC, and 10.22% for CNT dispersed composite. The ILSS was also the highest (80 MPa) for the FFRC. Zhang et al.[99] grafted CNTs onto carbon fiber surface to improve strength, toughness, and the interfacial interaction in the CFRP. CNTs acted as a transition layer to moderate the large difference between the elastic modulus. It was shown that the composites containing these modified carbon fibers showed an increase of 66.3% in the Young's modulus from the normal CFRP. Also, tensile strength and elongation at break improved by 40.5% and 41.9% respectively. Qian et al.[100] used the CVD method to generate CNT-grafted carbon fiber. The importance of wetting the fibers with polymer matrix and the effect of CNT in the wetting behavior were considered. Grafting with CNT increased Brunauer–Emmett–Teller (BET) surface area (which is used to characterize the porosity of material/ physical adsorption capability) by three times, but there was a reduction in tensile strength of carbon fibers by 15%. In the presence of CNT, the wettability of carbon fiber was moderately improved, indicating better adhesion and bonding with the matrix. A single fiber fragmentation test was performed to determine the IFSS, and it was found that CNT-grafted carbon fiber has higher IFSS by about 1.264 times the as-received carbon fibers (Figure 2.) Also, there are changes in the tensile modulus of the carbon fiber after undergoing desizing, oxidation treatment and grafting of CNTs. The tensile modulus of CNT-grafted carbon fibers is found to be slightly reduced compared to commercial carbon fibers as used in Reference [98] (T700S carbon fiber) and Reference [100] (IM7 carbon fiber). The chemical procedure used to create fuzzy fibers was EPD in Reference [98] and CVD in Reference [100] showing that there is higher reduction in tensile modulus for CVD process. Figure 3. shows the fracture surface for the case of normal carbon fiber composite and CNT-grafted carbon fiber composite.

Bedi et al.[101] examined the fiber wettability for CNT-grafted and unsized carbon fibers. They used experimental characterization for contact angle measurement and for single fiber pull out tests. The significant contribution for their work was emphasizing the impact of choice of polymer matrix in improving the IFSS, wettability, and mechanical properties of the carbon fiber/polymer composites. It was found that the improvement in composite's properties was significant with the use of epoxy matrix

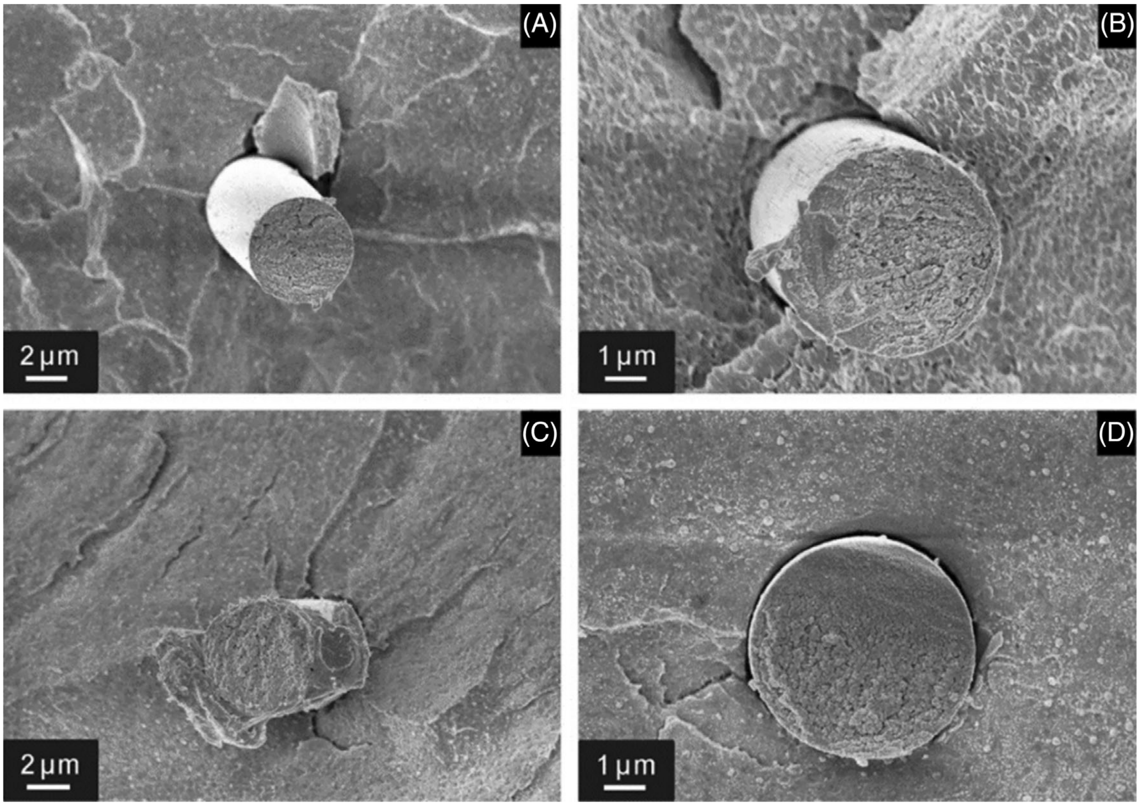

FIGURE 3 Fracture surface images of carbon fiber/Epoxy composite (top two images) and carbon fiber grafted with carbon nanotube (CNT) (bottom two images).[100]

but not for the case of polyester matrix. Zhang et al.[102] found that for CNT-grafted carbon fiber composite with Polyphthalazinone ether ketone (PEEK), the IFSS improved by 55.52%. Li et al.[103] generated CNT-reinforced FFRCs and using the polymeric functional coating, poly(styrene-alt-dipotassium maleate) (K-PSMA) could retain the tensile properties of the carbon fibers. This coating adsorbs on the surface of carbon fiber and helps in the ion exchange process with the iron catalyst used in fabrication. This is possible because of potassium ions on the surface of carbon fiber. Hence, the fiber was not desized, and the interfacial fiber matrix properties remained intact even after grafting with CNT. CVD was used for CNT grafting, and vacuum-assisted resin transfer molding (VARTM) was the fabrication method for FFRC generation. It was found that the IFSS of FFRC was in the range of the IFSS of normal carbon fiber composite. Aziz et al.[104] used floating CVD method for growing CNTs onto carbon fiber and studied the characterizing factors such as growth temperature time and growth conditions in CVD chamber for their effect on the surface morphology of the modified carbon fiber. It was found that for a reaction temperature of 700°C and reaction time of 30 min, the resulting growth of CNT coating showed 45% increase in IFSS over the untreated carbon fiber.

Liu et al.[105] used the sizing method to grow MWCNTs onto the surface of carbon fibers, for which the CNTs were hybridized using an amine monomer. A uniform CNT distribution and a 69.6% increase in the IFSS for FFRC were achieved. Similarly, Laachachi et al.[97] used oxidation treatment to grow CNTs onto carbon fiber using the chemical processing method. Both CNTs and carbon fibers were functionalized separately to grow carboxylic acid groups and amine functions onto them. These functional groups then interact among themselves to form anchoring bonds which lead to the adhesion of CNTs onto the carbon fiber surface. Moise et al.[73] used PLD for grafting SWCNT grown on the carbon fiber surface. PLD technique was unique in that it helps prevent the mechanical characteristics of the carbon fiber by allowing the use of lower temperatures for CNT growth. The effect of temperature on the SWCNT growth was prominent, where lower temperatures showed fine and even deposition of SWCNT with smaller lengths and higher temperatures led to deposition of longer CNTs and formation of nodules and clusters. Micro-droplet pull-out test was used to evaluate the effect of SWCNTs on interfacial properties of carbon-epoxy composite. Also, IFSS improvement by 20% was registered when CNTs were deposited at a temperature of 290°C.

Zhang et al.[106] utilized thermal CVD to grow high-density MWCNTs directly on two different polyacrylonitrile (PAN)-based carbon fibers (T650 and IM-7). The influence of different atmospheric conditions under the CVD growth process on the tensile properties of single fiber was studied. They registered that under high temperature and vacuum conditions, the tensile property of fuzzy fiber degraded significantly. This reduction was less when the CVD process occurred in the presence of argon. Also, with an increase in temperature, the growth density of CNTs was increased. Further increasing feed rate and temperature (from 750 to 800°C), the uniform and aligned deposition changed to non-uniform and random. Li et al.[107] used a simple electrospray technique to deposit short MWCNTs onto the carbon fiber surface. No degradation in carbon fiber tensile properties and improvement in fiber-matrix wettability and BET surface area was found. A significant increase in surface roughness, which leads to better fiber matrix bonding, and increased friction, was also registered. Fractographic analysis showed that adding CNT led to a decrease in crack size and fiber pull-out length. Thus, concluding that CNT grafting gave better interfacial properties to the carbon fiber-polymer matrix composite. Shan et al.[108] used a modified spraying technique assisted with spraying E20 epoxy resin in surface of carbon fiber to graft MWCNTS onto the carbon fiber. It was found that depositing CNTs with E20 increases the mode I fracture toughness by 24% in the average propagation and mode II fracture toughness by 11% in the propagation and ILSS by 12%, while it preserves the in-plane mechanical properties. It can be emphasized that spraying E20 is effective for fixing the CNTs on the carbon fiber surface under resin flushing, and has no negative effect on the processing quality of laminates, including thickness and defects.

Using thermal CVD with catalytic decomposition of acetylene, Sharma et al.[109] showed that CNT-grafted carbon fiber composite had 69% higher tensile strength than composites made of normal carbon fiber. Two mechanisms to explain the effect on tensile strength of carbon fiber by CNT were suggested. One was that CNTs improved the bonding at the interfacial level. The other was that aligning CNT along fiber axis direction contributed to surface roughness, thereby increasing it, and resulting in mechanical anchoring of CNTs with the polymer matrix. Kamae et al.[110] used a novel method of producing fuzzy fibers where CNTs are treated with cationic polymers and are then coated onto carbon fibers by immersion into a CNT/water suspension. Good dispersion is achieved by repulsive force between positively charged CNTs, and uniform coating of the CNTs is achieved by attractive forces between positively charged CNTs and negatively charged carbon fibers. CNT used in this study were MWCNTs with a purity of 90%, and an average diameter of 10 nm. Significant improvement in the IFSS was found. For unsized fiber, IFSS increased from 30 to 35 MPa for MWNT-coated carbon fiber, whereas for sized fiber, IFSS increased from 78 to 81 MPa.

Zhang et al.[111] summarized various methods for growing CNTs onto carbon fiber surfaces and highlighted effect of CNT type, distribution and growing technique on the failure and delamination behavior of the composite laminates. It was found that CNTs deposited using the CVD method, surface assembly and interlayer insertion techniques led to enhancement of the Mode I fracture toughness of the laminates. CNTs incorporated with prepreg modification and resin mixing improved Mode II fracture toughness. Also, the vertically oriented CNT along the thickness direction was suggested to be the best among all different orientations to cause a significant improvement in composite mechanical properties. Feng et al.[112] used injection CVD (ICVD) for CNT grafting as it is known to cause less damage to carbon fiber surface and suggested that it can be used for large-scale production of CNT-grafted carbon fibers. The microstructure of the pyrocarbon (PyC) matrix and compression and shearing tests on the laminate composite were studied. It was found that the presence of CNTs led to an increase in the in-plane and out-of-plane compressive strength by 115% and 32%, respectively. Similarly, the in-plane and out-of-plane compressive modulus was increased by 46% and 11%, respectively and ILSS was enhanced by 108%. CNTs could also change the damage initiation and propagation behavior pertaining to reducing the stress concentration at the fiber/matrix interface.[113] Fiber/matrix debonding was avoided using CNT, but IFSS was also reduced in the presence of CNT. The reduction of 35.7% in IFSS was attributed to the stress concentration at the CNT/matrix interface, poor wettability of grafted fibers and degradation of carbon fiber in the process of CNT grafting. This showed that fracture phenomena and redistribution of stresses need to be analyzed at a microscopic level to understand the CNT/fiber/matrix bonding phenomena fully. A way to determine and develop the optimum CNT distribution and grafting is needed to develop high-performing fuzzy fibers. Figure 4. shows the detailed fracture surface and failure mechanism for CNT-grafted carbon fiber composite.

Yao et al.[114] used the CVD grafting technique and analyzed the out-of-plane and dielectric properties of the FFRC. An increase of 30.73% in the IFSS and 32.29% in the ILSS of the fuzzy fiber compared to the desized carbon fiber composite was found. The dielectric permittivity was improved upon grafting CNT as they help create nanocapacitor structures, improving electrical conductivity by

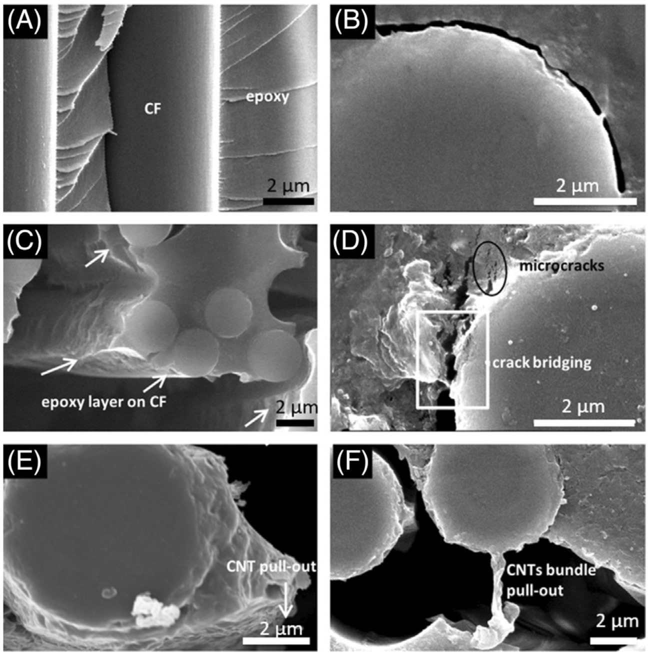

FIGURE 4 Fracture surface for different composites. (A,B) Fiber-matrix debonding in normal carbon fiber epoxy matrix composite, (C–F) Surface image of carbon nanotube (CNT)-grafted carbon fiber epoxy matrix composite showing the different failure phenomena of crack bridging, micro cracks and CNT pull-out.[113]

reducing the mutual contact between different carbon fibers. The out-of-plane conductivity was found to increase significantly.

Wang et al.[115] studied the grafting force between CNT and carbon fiber and investigated the effect of wettability on the IFSS using the microdroplet and contact angle tests. Pull-out tests were carried out for various fuzzy fiber samples revealing two main modes of failure: either detaching of CNT from the fiber surface or fracture of CNT. A strong grafting force of 5 N was found, and the IFSS was improved by 30%. Jin et al.[116] coated SWCNT onto a carbon fiber surface using a simple surface coating technique and generated their composite. Two carbon fiber types were used; high modulus and low modulus. The study suggested an increase of 70.66% in the IFSS for composites having high modulus carbon fiber. Using Raman spectroscopy, the IFSS for low modulus carbon fiber composite was determined for the first time and was found to be 51.1 MPa in the presence of SWCNTs. Qian et al.,[117] using the CVD method of CNT grafting, found that the IFSS increased by 60% during the fiber pull-out test. The dependence of IFSS on the CNT embedded length was also studied, showing that IFSS remains practically unchanged with length indicating ductile type failure. Push-out tests were also performed, but there was no significant improvement in the IFSS for the fiber push-out test. Qin et al.[118] developed a scalable process to grow CNTs in situ on the surface of continuous carbon fibers and achieve their uniform distribution. Open CVD apparatus with pre-treatment devices were used to achieve continuous growth of CNT. For the catalyst precursor concentration of 0.05 mol/L, CNTs/carbon fiber reinforcements with the best growth state and the highest tensile strength were obtained. An improvement of 95.43% more than the original carbon fibers was obtained for the IFSS of the composite. Li et al.[119] registered an increase of 29.7% for the IFSS of CNT-grafted epoxy composite and suggested use of chemical treatment for better adhesion of CNT to carbon fiber. They also studied the torsional strength and torsional modulus of normal carbon fiber and CNT-grafted carbon fibers with various surface treatments. Figure 5. shows the range of IFSS values obtained in literature for various composites of CNT-grafted carbon fibers.

Khan et al.[120] grew CNT onto a carbon fiber fabric surface using CVD. Two different growth types were used; in Type 1, the CNTs were grown only on the top surface of the lamina, whereas in Type 2, both the top and bottom surface of the lamina were covered with CNTs. Results from the short beam shear test, mode II fracture test, three-point bend test and flexural test to characterize the composites manufactured using grafted CNT and ungrafted carbon fiber fabric indicated that ILSS increased by 32% for Type 1 composite lamina and 102% for Type 2. The mode II fracture toughness was enhanced by 53% with the use of CNTs. Wu et al.[121] used polydopamine (PDA)-based carbon coating to achieve a uniform distribution and growth of CNT onto the carbon fiber surface. PDA coating was found to help keep the fiber surface damage free. The failure mechanism of the composite, ILSS and flexural strength was determined. It was found that for CNT-grafted fiber without PDA, crack propagation occurs along with the CNT/resin interface and the interface between carbon fiber and resin due to the uneven distribution of CNT. However, using PDA leads to CNT's stronger and uniform attachment to carbon fiber, and hence crack propagation occurs only at CNT/resin interface.

Boroujeni et al.[122] investigated the impact of using MWCNTs on the interlaminar fracture toughness of hybrid carbon fiber reinforced composites. For this, graphitic structure by design (GSD), which is a low temperature synthesis technique was adopted and MWCNTs, in two different morphologies-uniform and patterned, were grown on carbon fabrics. It was found that composites interlaminar fracture toughness was enhanced by 22% and 32% for uniform and patterned growth morphologies, respectively. Fractography analysis revealed that the MWCNTs played a role in interlaminar crack stoppage and deflection, leading to improved interlaminar fracture

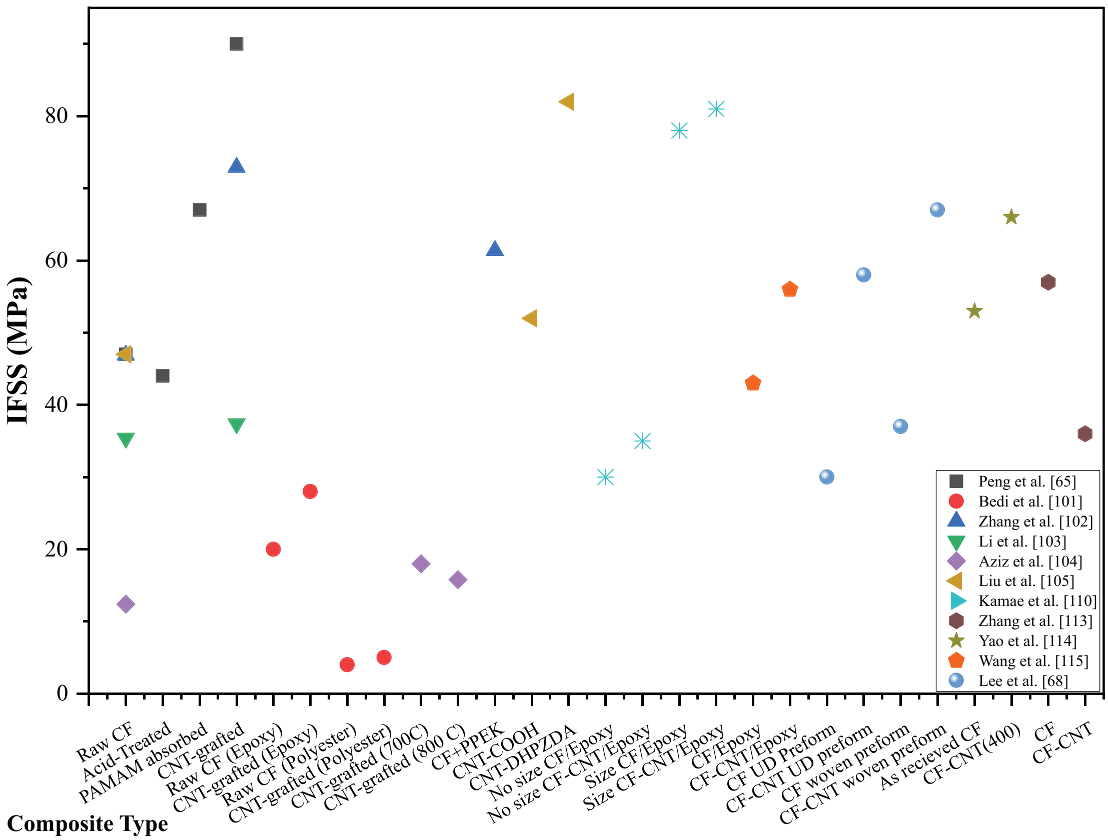

FIGURE 5 Interfacial shear strength (IFSS) graph for different composite types (here CF: carbon fiber).

toughness. Impact testing was done on carbon fiber composites developed using six different types of carbon fiber surface treatment.[123] Uniformly and patterned grafted CNTs were grown on the carbon fiber surface with patterned grafted fibers showing greater improvement on the in-plane performance and impact energy dissipation, with minimal delamination area. Nasirmanesh et al.[124] used CNT interleave sheets in between CFRP plies and found that for low velocity impact tests, the planar damage area was higher for laminates with CNT sheet whereas the external dent area and depth were higher for normal laminates without CNT sheet. Bedi et al.[125] examined the tailoring of CFRP composite by grafting CNT onto carbon fibers. They observed that microstructure of the interface plays an important role and effect the IFSS and fracture toughness. Moreover, the IFSS and fracture toughness were also shown to have power-law type rate dependence on the applied loading and are not constants. Table 1. gives a highlight of various property enhancement recorded in literature particularly in the elastic properties of fuzzy fibers and FFRCs. Further, the nonlinear mechanical behavior of fracture, failure and delamination is summarized in Table 2. The arrows in the tables indicate the overall increase/decrease in the fuzzy fiber/FFRC properties when compared to carbon fiber and CFRP respectively.

3.2 | Thermal properties

Gong et al.[126] used chemical vapor infiltration to produce aligned CNT-grafted CFRP laminates and investigated the thermal properties. The thermal diffusivity improves by 3–5 times with the use of aligned CNTs, and even for a lower density growth of CNTs, the thermal conductivity of the composite was 72.24 W/m K, which is around 12.31% higher than that for normal carbon fiber composites. Jie et al.[127] deposited CNTs onto pyrolytic carbon, developed on carbon fiber's surface after heat treatment using the CVD process. Analyzing the composites of these fuzzy fibers, the transverse thermal conductivity was five times higher than for conventional composites. This improvement was only 2.5 times the original composite for the axial direction. Naito et al.[128] grafted CNTs onto PAN and pitch-based fibers and studied their respective thermal behavior. The grating was carried out using the CVD process, and improvement was observed for both fiber types. For PAN-based carbon fiber (T1000GB),

TABLE 1 Elastic properties of fuzzy fiber and fuzzy fiber reinforced composites (FFRCs) from experimental studies.

Ref.	CNT	Production	Properties (Fuzzy Fiber)	Properties (FFRC)
[98]	MW	EPD	Tensile Modulus: 262.6 GPa ↑	Tensile strength: 1200 MPa ↑ Flexural strength: 1078.84 MPa ↑ Flexural modulus: 57.47 GPa ↑
[100]	–	CVD	Tensile strength: 1450–1840 MPa ↓ Tensile modulus: 240–246 GPa	–
[103]	–	CVD	Tensile modulus: 200–203 GPa	Tensile modulus: 128–130 GPa
[106]	MW	CVD	Tensile strength: 1000–3500 MPa ↓ Tensile modulus: 150–220 GPa	–
[107]	MW	Electrospray	Tensile strength: 4–4.3 GPa Fiber Wettability	–
[109]	MW	CVD	–	Tensile strength: 280–580 MPa ↑
[110]	MW	Dipping	–	Shear modulus ↑
[112]	–	CVD	–	In-plane compressive modulus: 6.92–9.22 GPa ↑ In-plane compressive strength: 110–122 MPa ↑ Out-of-plane compressive modulus: 3.37–3.67 GPa ↑ Out-of-plane compressive strength: 200–210 MPa ↑
[114]	–	CVD	Tensile strength: 3.7–4.1 GPa	
[118]	–	CVD	–	Tensile strength: 3.8–4.1 GPa
[121]	–	CVD	–	Tensile strength: 3.1–3.4 GPa
[68]	–	CVD	–	Tensile strength: 4000–4100 MPa
[141]	MW	CVD	–	Flexural strength: 360–520 MPa ↑ Flexural modulus: 210–590 MPa ↑

the increase was from 12.6 to 18.6 W/mK and for pitch-based fibers (K13D), it was from 745.5 to 956.6 W/mK. Hu et al.[129] developed CNT growth on the carbon fiber/silicon carbide matrix composite using pyrolysis and polymer impregnation. The CNT growth was developed in the inner part of the carbon preform, and space between carbon fiber bundles of 1–2 μm was filled with CNT. Analyzing the thermal properties of the composite, an increment of 24% for parallel thermal conductivity and 52% for perpendicular thermal conductivity was found for a 4% CNT volume fraction.

Zakaria et al.[84] employed the EPD method for woven carbon fiber composites. MWCNTs of length 10–30 μm and diameter of 5–30 nm were deposited on the woven carbon fiber and later treated with epoxy using the VARTM process. Different electro spraying modes such as micro dripping, cone jet, multi-jet and rim jet were studied for different voltages ranging from 2 to 20 kV. The optimal CNT deposition voltage was found to be 15 kV. The thermal conductivity of the resulting composite was 1 W/mK demonstrating a 35% increase compared to normal composite. Also, the ILSS was improved by 25% and tensile modulus by 37%. Li et al.[130] could achieve a coefficient of thermal expansion (CTE) of in range 3.2×10^{-6} to 4.2×10^{-6} K^{-1} upon reinforcing the carbon fiber felt with CNT using EPD process. Apart from this, PyC was also infiltrated into the composite. The different microstructure of these materials, helped reduce the CTE from that of normal composite and enhanced the compressive strength and modulus by 37% and 19% respectively. Shin et al.[131] deposited different CNT types: CNT mat, CNT buckypaper and direct CNT growth. For each of these reinforcement types, the out-of-plane thermal and electrical conductivity of carbon fiber composites was found out. CNT weight fractions in range 1–3.7 wt% were used. The out-of-plane thermal conductivity for the composite increased from 0.5131 to 0.6293 W/mK, whereas for the case of CNT deposition on fabric, this increase was more going from 0.5034 to 1.3864 W/mK. Vertically aligned CNTs were used to improve the thermal conductivity in the through thickness direction of carbon fiber/SiC composites.[132] CNTs of length 13–60 μm and high density were perfectly bonded in presence of SiO_2 coating. Hence a thermal conductivity increases from 7.94 to 16.8 W/mK was seen for the composite with improved heat transfer in through thickness

TABLE 2 Fracture, damage, and failure properties of fuzzy fiber and fuzzy fiber reinforced composites (FFRCs) from experimental studies.

Ref.	CNT	Production	Properties (fuzzy fiber)	Properties (FFRC)
[64]	–	CVD	Fracture Load: 32–156 N ↓	–
[65]	–	Grafting	–	IFSS: 90 MPa ↑
[73]	SW	PLD	IFSS: 14.6–49.3 MPa ↑	–
[98]	MW	EPD	–	ILSS: 80 MPa ↑
[100]	–	CVD	–	IFSS: 110–120 MPa ↑
[103]	–	CVD	Interfacial Strength: 33–41 MPa	Interfacial strength: 25–70 MPa
[105]	MW	Sizing	–	IFSS: 50–80 MPa ↑
[107]	MW	Electrospray	–	IFSS: 40–60 MPa ↑ Fracture/crack size ↓
[109]	MW	CVD	–	Debonding
[110]	MW	Dipping	–	IFSS: 31–78 MPa ↑
[111]	–	CVD	–	ILSS: 22–80 MPa Fracture toughness: 0.5–2.4 kJ/m^2 ↑
[112]	–	CVD	–	ILSS: 17.8–21.4 MPa ↑
[113]	–	CVD	–	IFSS: 50 MPa ↑ Damage
[114]	–	CVD	–	IFSS: 55–68 MPa ↑ ILSS: 70–82 MPa ↑
[115]	–	CVD	Breakage Strength: 46–76 GPa Fracture	IFSS: 55 MPa ↑ Fracture
[116]	SW	Coating	–	IFSS: 38.3–58.3 MPa ↑
[118]	–	CVD	–	IFSS: 90 MPa ↑
[120]	–	CVD	–	Fracture toughness: 2200 J/m^2 ↑ ILSS: 14 MPa ↑
[121]	–	CVD	–	IFSS: 48–78 MPa ↑ ILSS: 80–90 MPa ↑ Failure
[125]		CVD	IFSS	IFSS Fracture toughness
[68]	–	CVD	ILSS: 50 MPa	IFSS: 55–67 MPa ↑ ILSS: 51–54 MPa

direction. Similar trends in increase of composite thermal conductivity in the range of 12%–21% was observed[133,134] upon grafting with CNTs. Figure 6. shows the thermal conductivity of various CNT-grafted carbon fiber composites along with the normal carbon fiber composites. The percentage increment has also been plotted.

3.3 | Electrical properties

Yao et al.[135] investigated the effect of orientation and distribution patterns onto the electrical properties of the CNT reinforced composite laminates. Using CVD synthesized vertically aligned CNT forest, which was grown onto iron substrate, the web of horizontally oriented CNTs was generated and was then embedded between two layers of glass fiber prepregs (serving as insulators) and their electrical characteristics were studied. The results demonstrate that the conductivity of the specimens was influenced by their aspect ratio, with the variation in web orientation. This relationship was bounded by two theoretical curves, representing aspect ratios approaching zero and infinity. Yang et al.[136] developed fibers based of the renewable biodegradable polylactide

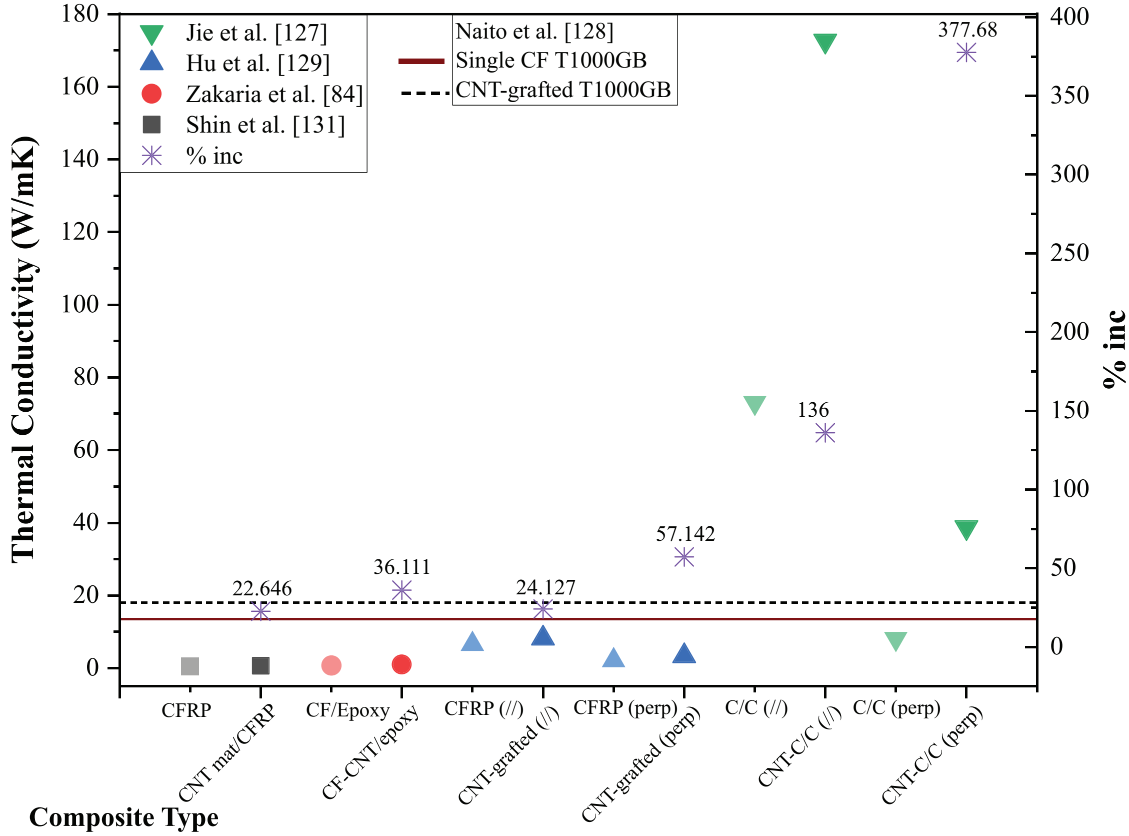

FIGURE 6 Thermal conductivity plot for various composites along with percentage increase for the case of carbon nanotube (CNT)-grafted fiber composites (//—indicates thermal conductivity along the fiber axis, perp-indicates thermal conductivity in direction perpendicular to the fiber axis).

and further reinforced CNTs using electrospun technique and identified the electrical conductivity behavior based on the variation in loading of the CNT. It was found that at high loading levels CNTs are distributed along the fiber axis in huge bundles, thereby giving a nine-fold increase in conductivity from 0 to 1 wt%.

Pozegic et al.[137] developed fuzzy fiber networks that enhanced the electrical conductivity of the composite overall. They used a low-temperature growth technique for grafting CNT, the photo thermal chemical vapor deposition (PTCVD), which uses optical radiation for heating and includes a water-cooled substrate. Further, VARTM was used to manufacture the FFRC with better wettability than conventionally sized carbon fiber composites. A reduction of only 9.7% in the tensile properties was registered. Moreover, the electrical conductivity of the FFRC was enhanced by 330% in the in-plane direction and 510% in the out-of-plane direction, indicating the multifunctionality of FFRCs.

Lee et al.[68] grafted two different length types of CNT- short & thin and long & thick to determine the effect on the FFRC's mechanical, thermal, and electrical properties. Unidirectional and woven composites were considered. A difference in tensile strength of short CNT-grafted composite, and long CNT-grafted composite was observed for both unidirectional and woven composites. The long CNT composite exhibited higher tensile strength, which was 15.38% higher than the shorter CNT composite for the unidirectional case and 18.11% for the woven case. It was also found that the long CNTs suppressed the splitting crack initiation. The electrical and thermal conductivities were also improved in the presence of long CNT. Yue et al.[58] grafted continuously grown CNT onto carbon fiber using CVD, the composite of which can be used for electromagnetic wave absorption. Not much difference in the tensile strength of the desized, electrolyzed, and fuzzy carbon fiber was seen. Reflection loss for different thickness values of CNT-grafted carbon fiber was calculated using the transmission line theory. The CNT-grafted carbon fiber composite was found to have outstanding electromagnetic wave absorption capabilities, particularly in the Ku band. Such behavior in Ku band frequency range has also been demonstrated for CNT-based vinyl ester composites.[138] The 3-D structure developed in the presence of CNT on carbon

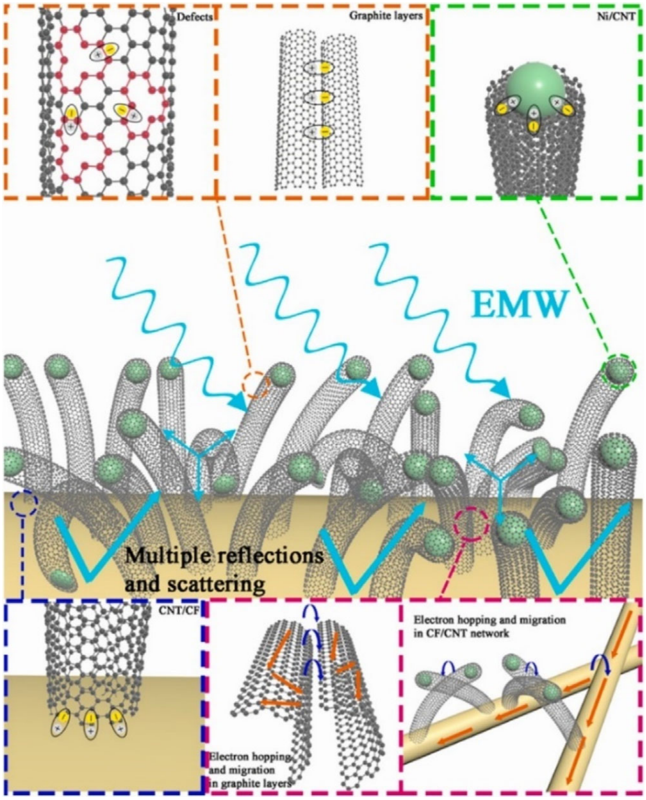

FIGURE 7 Schematic representation of electromagnetic wave absorption and reflection by carbon nanotube (CNT).[58]

fiber surface leads to creating an electrically conductive network with strong attenuation capability. This work supports the argument of replacing currently used polymeric sizing with CNTs to get electrically advanced, multifunctionally applicable composite structures. Figure 7. shows a representation of mechanism of electromagnetic wave interaction with CNTs.

Duongthipthewa et al.[139] investigated the electrical conductivity of CNT-grafted carbon fiber composites. They used the floating catalyst CVD method on carbon fiber coated with Nickel catalyst to develop fuzzy carbon fiber. Further, FFRCs were manufactured using the autoclave curing, wet lay-up technique. Also, graphene nano-platelets (GNPs) were dispersed on the fuzzy fiber surface to form synergistic physical interactions between two different low-dimensional carbon-based nanostructures. It was found that both mechanical properties and functional conductivity have a synergistic enhancement. By including GNPs in the composite, the electrical conductivity was increased by approximately 40%, 300%, and 190% along the fiber, surface and through-thickness direction compared to the FFRCs. Mechanical properties, including flexural strength, work of fracture, impact, and ILSS were also enhanced. Patton et al.[140] investigated using FFRCs for sensor applications by studying the FFRC's electromechanical response. Very small localized compressive loads were applied, and resistance displacement was applied. It was found that hysteresis in resistance was observed for CNT/SiO2/carbon fiber. This hysteresis loss was attributed to the CNT entanglement and vdW forces between CNTs, leading to a reduction in conduction paths.

Gurkan et al.[50] grafted CNT onto glass fibers using the thermal CVD method. Further, the glass fuzzy fiber composite was developed using a vacuum infusion process. For these glass fuzzy fiber composites also, the CNTs led to an enhancement in the fracture toughness by about 119%. Performing electrical conductivity tests, it was estimated that grafting of CNT led to an improvement in electrical conductivity of the composite by 7–8 times. Samsur et al.[141] studied woven composite made up of CNT-grafted carbon fibers. They also investigated the electrical and flexural mechanical properties of the composite. The significant finding was the reduction of electrical resistivity of composite from 25 to 0.2 Ω m when a CNT weight fraction of 3.3% was used. An increase in flexural strength by 34% for a CNT weight fraction of 1.65% and an increase in flexural modulus by 126% for the same weight fraction were also found. Singh et al.[142] studied composite materials' electromagnetic interference shielding effectiveness upon grafting carbon fibers with CNT. The multiscale composite prepared was found to be effective for the absorption of microwaves in the X band. They also studied the electrical conductivity of the FFRCs. By increasing the MWCNT volume fraction from 0.5% to 3%, the electrical conductivity increased by about 46%. ILSS of the composite was also investigated, showing an increase of 117%.

Pozegic et al.[59] suggested CNT reinforced carbon fiber as an alternative to polymer fiber sizings used in the aerospace industry. They used high-quality CNTs grown at a high density in the presence of an aluminium interlayer, which reduces the diffusion of catalyst and has previously been shown to minimize diffusion of the catalyst in the carbon fiber substrate. The FFRC was fabricated using VARTM. The mechanical properties of the carbon fiber were found to remain intact irrespective of CNT growth. Electrical conductivity tests were carried out on FFRCs, and it was shown that a maximum increase in electrical conductivity was registered along the thickness direction, which was about 450%. There was a 300% increase along the surface direction, and along the volume direction, it was 230% (Figure 8). Apart from this, the thermal conductivity was improved by 107% along the thickness direction after fiber normalization. Electrical property analysis of CFRP laminates with aligned nanotubes grown directly at interfaces was carried out by Russello et al.[143] The direct synthesis of

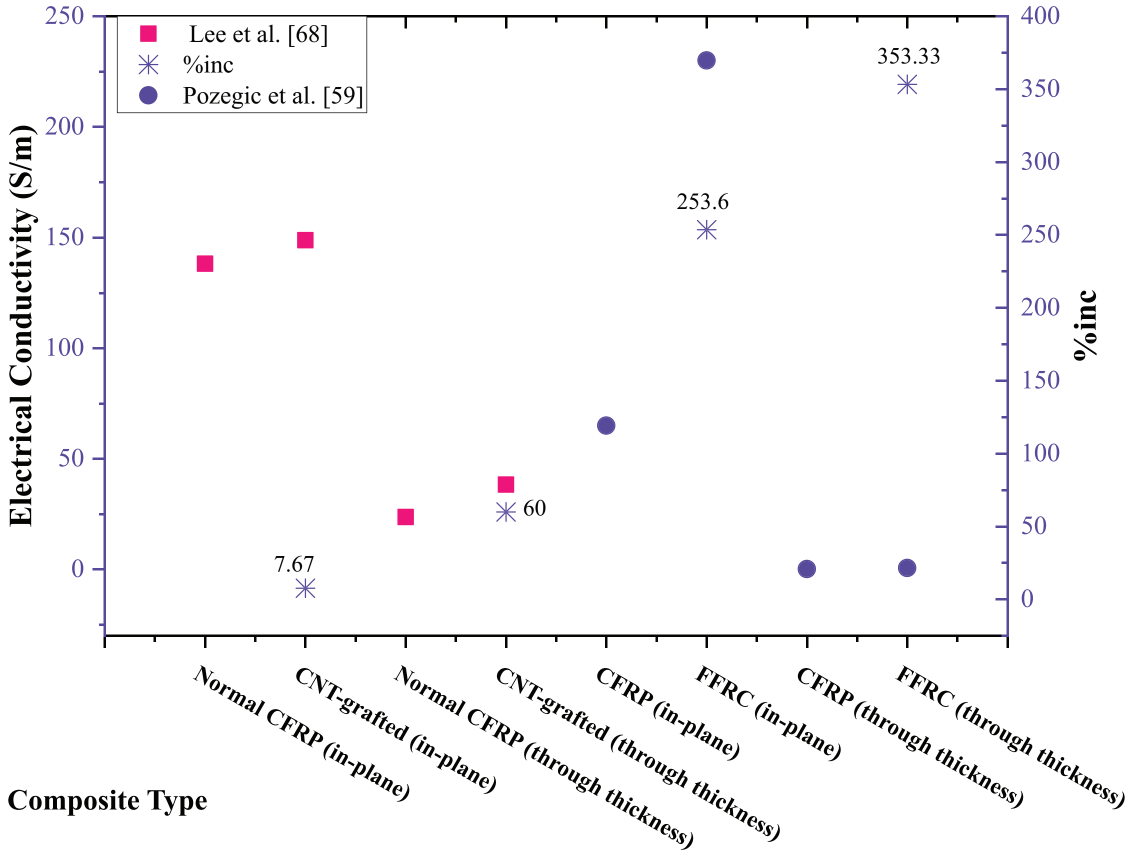

FIGURE 8 Electrical conductivity plot for various composites along with percentage increase for the case of carbon nanotube (CNT)-grafted fiber composites.

TABLE 3 Experimental studies on the electrical and thermal properties of fuzzy fiber reinforced composites (FFRCs).

Ref.	CNT	Production	Properties (FFRC)
[114]	–	–	Electrical conductivity ↑
			Dielectric properties
[68]	–	–	Electrical conductivity: 38–150 S/m ↑
			Thermal conductivity: 0.945–2.78 W/m K ↑
[140]	MW	CVD	Electrical resistance
[141]	MW	CVD	Electrical resistivity: 0.2 Ω m
[59]	MW	CVD	Thermal conductivity: 0.9–1.5 W/m K ↑
			Electrical conductivity: 0.9–2500 S/m ↑
[143]	–	CVD	Electrical Conductance: 50–220 mS ↑

CNTs was done using CVD process, achieving a CNT content in the range of 10–30 wt%. The transverse conductance of the laminates was improved by about 2800% depending upon the reaction time and type of CNT growth. For inhomogeneous CNT deposition, the increase was from 6 to 50 mS, whereas upon increasing the reaction time, the conductance further improves to 110 mS (for 10 min) and 220 mS (for 12 min). The transverse conductivity improvement is however not reflected for longitudinal case thereby reducing the electrical anisotropy of the material.

Gong et al.[144] deposited CNTs onto carbon fiber prepregs using by dry spray deposition and used these composites for electromagnetic shielding. It was found that electromagnetic interference shielding effectiveness was dependent on CNT loading and varied linearly with it. Moreover, the prime mode of shielding was adsorption whereas reflection mode remained independent of CNT content. Table 3. summarizes the findings of experimental studies on the thermal and electrical properties of FFRCs.

4 | MODELING METHODS

There have also been a lot of numerical studies focusing on the modeling of fuzzy fiber and its composites. The modeling methods face a major challenge in effectively capturing the geometry of the fuzzy fiber. Many different techniques such as FEM, MD, micromechanics, and homogenization theories have been used to characterize FFRCs.

4.1 | Mechanical properties

Romanov et al.[145] performed a multiscale modeling analysis using a finite element approach to determine the effect of CNTs grown on fiber surface on the interfacial stress at the fiber/matrix interface. They generated a square 3-D unit cell consisting of carbon fiber, and each CNT was modeled individually as wavy objects. The waviness of CNT was achieved by describing the centreline using a four-point spline. Carbon fiber volume fraction was taken as 60%, and CNT forests with 0.27% weight of CNT compared to carbon fibers and 1.33% weight per cent was grown. Subjecting the unit cell to transverse tensile loading, the principal stress at the matrix fiber interface was determined. It was found that the presence of CNT growth on the carbon fiber surface reduced the stress concentrations at the interface significantly. This is shown in Figure 9.

Kundalwal et al.[146] studied the effective elastic properties of FFRCs using analytical that is, mechanics of materials (MOM) and Mori-Tanaka (MT) models. They considered radially grown CNTs on the carbon fiber surface and divided the model into two parts wherein the CNT and polymer part of the FFRC were taken as polymer nanocomposite (PMNC). The carbon fiber was then taken to be reinforced in this PMNC to create the FFRC. The transverse elastic properties were found to be significantly improved in the presence of CNTs. In another work, they used the finite element (FE) model and Method of Cells (MOC) to determine the effective elastic properties of the FFRCs.[147] The comparisons of both the work and all the methods used are as given in Figure 10.

As evident from the above works, and the popularity of the FEM method, use of representative volume elements (RVEs) seems to be an efficient method of modeling the FFRCs.[148] The use of laminate theories and natural element methods (NEM) have also been characterized to help simulate the 3-D FFRCs.[149] Rafiee et al.[150] considered radially aligned CNT on carbon fiber surface to develop fuzzy fiber. They developed an analytical model for studying Young's modulus of the polymer matrix reinforced with unidirectional fuzzy fiber, which is the matrix impregnated CNT coated fiber. Further, with the use of stochastic modeling, the effect of volume fraction and curvature of CNT was considered. The overall modeling was done at four scales: nano, micro, meso and macro. At nano and micro-scale, FEM was used to characterize individual CNT and vdW interactions. Further, micromechanical theories of MT and Halpin-Tsai (HT) were used for mechanical characterization at meso and macro scales. An improvement of 65.3% in transverse modulus of the composite compared to normal carbon fiber composite was found at a CNT volume fraction of 2% only.

Analyzing the importance of the off-axis angle of carbon fiber in FFRCs, simplified unit cell (SUC) micromechanical model was used for mechanical characterization.[151] It was found that in the presence of CNT, the elastic properties decreased first for off-axis angle increase from 0° to 45°, but on increasing from 45° to 90°, the elastic properties increased, which is not the case for normal CFRC. There the elastic properties decrease continuously. Also, the

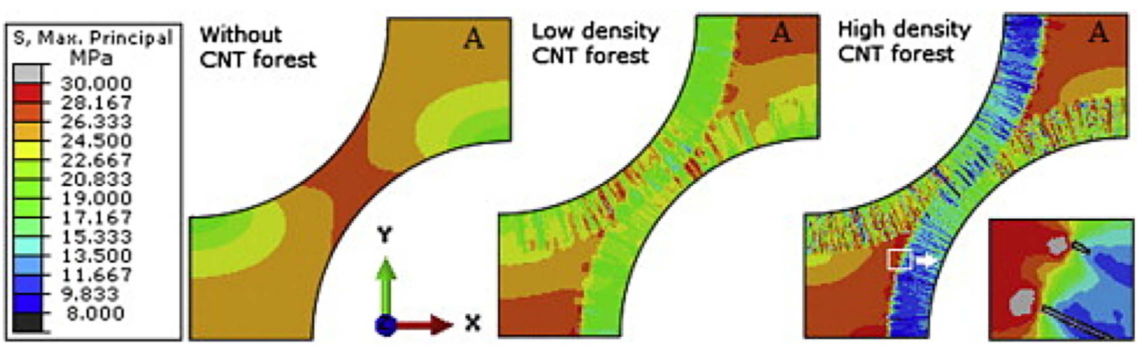

FIGURE 9 Depiction of reduced stress concentration at carbon fiber/polymer matrix interface on using carbon nanotubes (CNTs).[145]

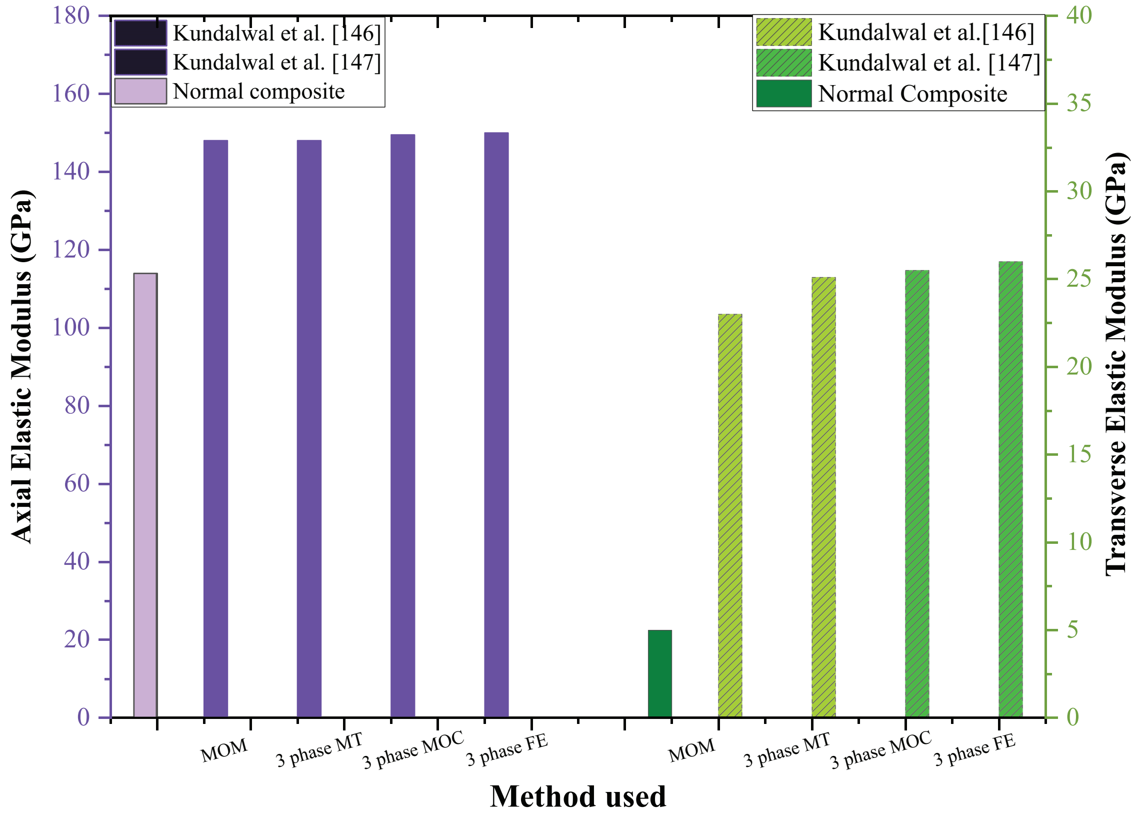

FIGURE 10 Comparison of axial and transverse elastic modulus obtained from various computational models.

transversely elastic properties of the FFRC were found to be in excellent agreement with the experimental results. The effect of the CNT volume fraction on the off-axis elastic modulus was positive, showing an increase in modulus when the CNT volume fraction is increased. Also, the Poisson's ratio was increased with increasing CNT volume fraction.

Subramanian et al.[152] used MD simulations to investigate the mechanical properties of the interface of FFRCs. They modeled the surface of carbon fiber as stacks of graphene layers that had voids. Atomic modeling of PSMA (polymeric coating) was introduced for the first time and was carried out in PACKMOL software using Merck Molecular Force Field. Subjecting the FFRC unit cell to virtual deformation along the x, y and z directions, the tensile and transverse stress–strain deformation curves were obtained. They found out that the longitudinal modulus of the fuzzy fiber/matrix interface was 17.32 GPa, whereas the out-of-plane modulus was 10.4 GPa.

Almousa et al.[153] used MD simulations to develop the microstructure of carbon fiber, polypropylene matrix and SWCNTs. They also used the PACKMOL software package to construct the fuzzy fiber nanoengineered polymer composite. A uniaxial tensile test was applied to get the stress–strain diagram and evaluate the mechanical properties of the composite. Also, the interface strength that is, the strength to peel off the polypropylene from carbon fiber, was calculated in the presence of CNT and without CNT. The results indicated that peeling strength increased to 7.59 MPa from 7.14 MPa in the presence of 1% CNTs, and for 3% CNTs, this value was 8.20 MPa.

Pawlik et al.[154] used computational micromechanics to study and predict the elastic properties of FFRCs. They generated RVEs with radially grown CNTs on the surface of the carbon fiber having hexagonal fiber arrays. The interface between CNT and matrix was thus taken to be transversely isotropic. The CNT modeled were considered to be straight and defect-free. Taking perfect bonding assumption, the element orientation command in ANSYS was used for analysis. Results revealed that there was two times increase in the transverse modulus for a carbon fiber volume fraction of 25%. This increment is dependent on the CNT volume fraction and the type of manufacturing process used. Longitudinal modulus for same carbon fiber volume fraction showed moderate difference with only a 5% increase. Thus, showing that CNT has more effect on the transverse modulus than the longitudinal modulus.

Malekimoghdam et al.[155] used the multiscale modeling approach and studied the bending of sandwich and laminated composite composed of CNT coated carbon fibers. The effective properties of the fuzzy fiber and the subsequent interface scale containing CNT, coating and matrix was obtained using Eshelby MT method and equivalent continuum fiber theory. Both random and aligned orientations of CNT around the carbon fiber was considered here. CCA was then used to get the equivalent elastic properties of the composite. Further, refined zigzag theory was used for stress analysis of simply supported composite laminates. The results revealed significant reduction in maximum transverse displacements and the transverse shear stresses in the core medium in the FFRC laminate beam compared to normal CFRP beam. Rao et al.[156] used the bridging mechanics model by Huang to estimate the effective elastic properties of CNT-grafted carbon fiber composites. They carried out two-step homogenization where micro scale involved modeling of CNT/polymer interface, and mesoscale modeling was composed of three phases that is, fiber, fuzzy interface, and matrix. Using a three-phase bridging model, the effective properties were determined. The effect of change in CNT waviness, length, and density on the composite's properties was studied. CNT's waviness directly impacted the longitudinal modulus, whereas increasing waviness decreased the transverse modulus. Similarly, for a constant CNT volume fraction, increasing CNT length causes an increase in shear and transverse moduli but a decrease in longitudinal moduli. Figure 11. highlights the elastic modulus obtained from different computational methods. Also, a brief summary of elastic properties of fuzzy fibers and FFRCs studied using various computational methods as described above can be found in Table 4.

4.2 | Thermal properties

Ray et al.[157] developed a micromechanical model to analyze the thermomechanical shear lag behavior of FFRCS. They considered wavy CNTs radially grafted along the outer surface of carbon fiber and the temperature dependence of the thermoelastic properties of the CNTs was considered. The MT method was used to estimate the effective thermoelastic properties. It was seen that the load transfer characteristic for the FFRC containing

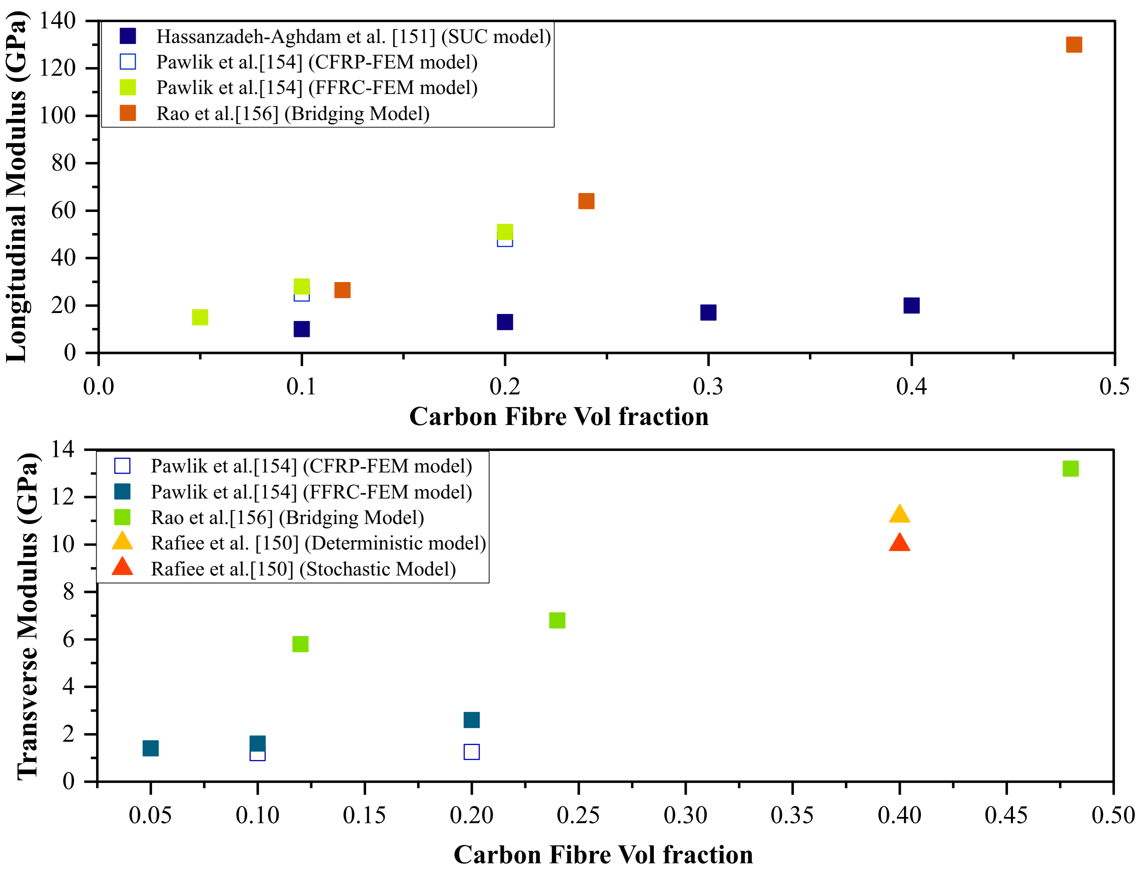

FIGURE 11 Comparison of longitudinal and transverse elastic modulus computed using different modeling techniques at different carbon fiber volume fractions.

TABLE 4 Mechanical properties of fuzzy fiber and fuzzy fiber reinforced composites (FFRCs) from computational studies.

Ref.	CNT	Method	Properties (fuzzy fiber)	Properties (FFRC)
[146]	SW	Micromechanics	Elastic properties C_{11}: 30–220 GPa ↑ C_{22}: 10–27.5 GPa ↑ C_{23}: 6–14 GPa ↑	Elastic properties C_{11}: 9–12 GPa ↑ C_{22}: 28–52 GPa ↑ C_{23}: 14–22 GPa ↑
[147]	SW	FEA, MOC	–	Elastic properties C_{11}: 30–220 GPa ↑ C_{23}: 6–14 GPa ↑ C_{55}: 4–14 GPa ↑
[150]	SW	FEA, Stochastic multiscale modeling	Tensile modulus: 649.37 GPa Shear modulus: 5.13 GPa Transverse modulus: 11.27 GPa ↑	Transverse Modulus: 11.2 GPa ↑
[151]	SW	Micromechanics	–	Elastic modulus: 8–80 GPa ↑ Poisson's ratio: 0.01–0.75
[152]	–	MD	–	Longitudinal modulus Transverse modulus ↑
[153]	SW	MD	Elastic moduli: 129.6–277 GPa Tensile strength: 9.9–26.7 GPa	Interfacial energy: 2.5–23 eV ↑
[154]	–	FEA, micromechanics	–	Longitudinal modulus: 10–65 GPa Transverse modulus: 1.2–3.5 GPa ↑ Shear modulus (out-of-plane): 3–11 GPa ↑ Shear Modulus (in-plane): 1.1–3.4 GPa ↑
[156]	–	Bridging model, multiscale modeling	–	Longitudinal modulus: 50–200 GPa Transverse Modulus: 3–26 GPa Shear Modulus: 2–15 GPa
[167]	–	FEA	–	Young's modulus: 3.3–47.25 GPa ↑ Poisson's ratio: 0.25–0.3
[169]	–	FEA, multiscale modeling	–	Longitudinal modulus: 6–18 GPa In-plane bulk modulus: 3.220–3.238 GPa In-plane shear modulus: 1.29–1.335 GPa
[175]	MW	FEA	–	Longitudinal modulus: 104–105.6 GPa Transverse modulus: 7.4–8.4 GPa

wavy CNTs significantly improved for lower values of temperature variation. Kundalwal et al.[158] also analyzed the effective thermoelastic properties of the FFRCs with radially grown CNTs. The approach used here was the generalized MOC and the MT method. They found that the axial CTE of the FFRCs slightly increases for the lower values of the carbon fiber volume fraction. In contrast, the transverse CTE of the FFRCs significantly decreases over those of the composite without CNTs (see Figure 12). In another approach by the same author, the effective thermal conductivities of an FFRC have been determined by using the effective medium approach (EMA) along with the composite cylinder assemblage (CCA) approach.[159] They found out that for CNT volume fractions 6.88% and 4.27% in the FFRC, the transverse thermal conductivities of the composite were improved by approximately 1040% and 400% respectively, compared to those of the composite without CNTs. It was also found that effective thermal conductivities of the FFRC increase with increase in carbon fiber volume fraction and the temperature. Moreover, the effective thermal conductivities of the FFRC are unaffected by the CNT/polymer matrix interfacial thermal resistance. In yet another work by Kundalwal and Ray,[160] the effect of CNT waviness on the thermal

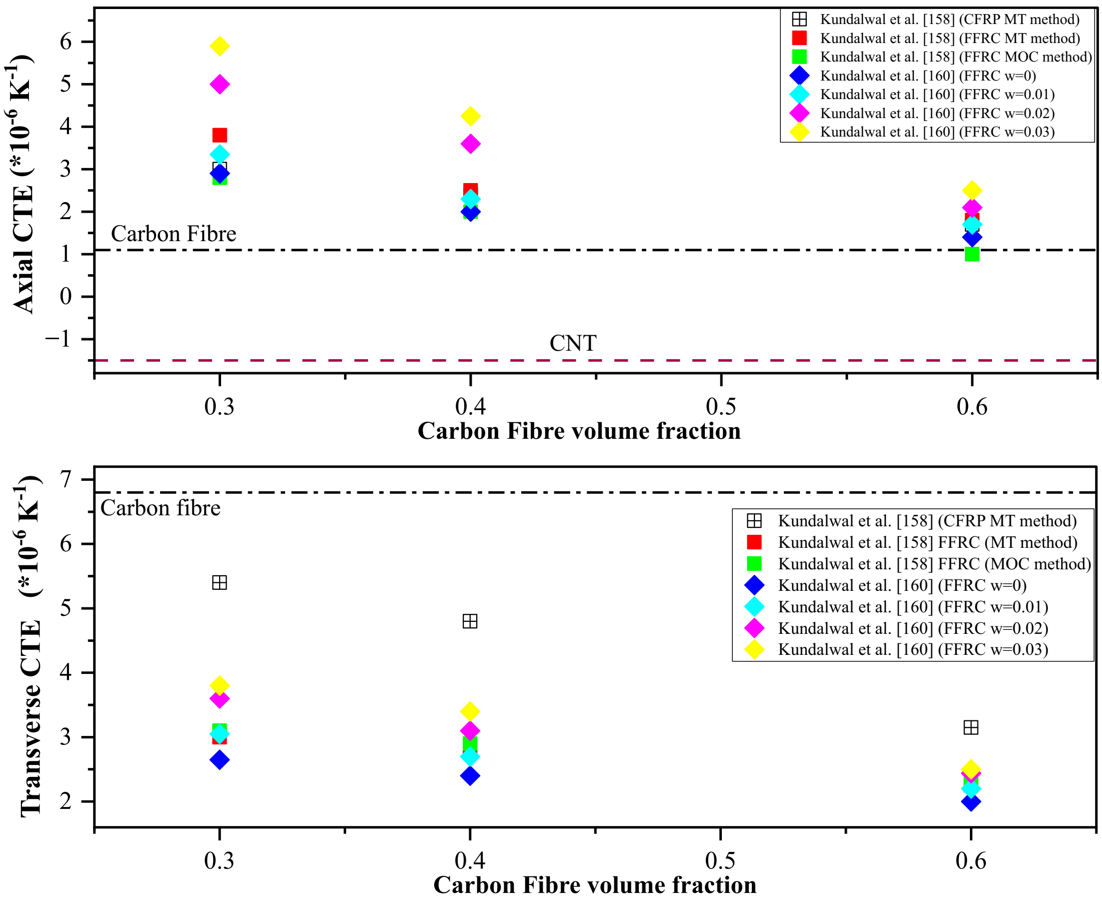

FIGURE 12 Axial and transverse coefficient of thermal expansion (CTE) for different composites at various carbon fiber volume fractions.

properties of FFRCs was studied. Using MOC, the CNT's waviness was characterized using sinusoidal functions. A unique feature adopted in this study was that they considered the sinusoidal CNT wave amplitude to be either parallel or transverse to the carbon fiber axis. The main finding was that having the amplitude of sinusoidal CNTs parallel to the axis of carbon fiber increased the effective CTE in both radial and axial directions (see Figure 12. for CTE values for different waviness factor, w).

Tian et al.[161] used a semi-analytical approach and locally exact homogenization theory (LEHT) to predict the thermal behavior of FFRCs. They studied two different RVEs: square array and hexagonal array. Lower-level unit cells were modeled, taking the unit cell depth to be equal to CNT length. The CNTs grown on carbon fiber that is embedded in the matrix were treated as an orthotropic interlayer. The unit cell at the upper level consisted of fiber, matrix and interlayer, the properties of which were generated from the lower-level unit cells. Validation of the model with FEA was done using DC2D4 elements. Their work suggested significant findings that the transverse thermal conductivity also increased on increasing the length of CNT. Moreover, there is little effect on axial thermal conductivity with the addition of CNTs. It was also found that fluctuation of heat flux between interlayer and fiber interface may lead to damage and microscopic cracks along the transverse direction.

Using multiscale micromechanics, Hassanzadeh-Aghdam et al.[162] modeled polymer nanocomposites with randomly grown CNT on carbon fiber for their thermal properties. Using a modified semi-empirical HT model and further coupled it with an analytical micromechanical extended simplified unit cell (ESUC) model, the overall thermal conductivity of CNT grown carbon fiber composite was determined. The influences of random dispersion, waviness, length, diameter, volume fraction of CNTs and the CNT/polymer interfacial thermal resistance and the carbon fiber cross-section shape parameters were also investigated. It was found that CNT coating has no effect on the longitudinal thermal conductivity of carbon fiber reinforced hybrid nanocomposites but the transverse thermal conductivities are significantly enhanced compared to the conventional fibrous composites without the CNTs coating. An improvement in the nanocomposite transverse thermal conducting

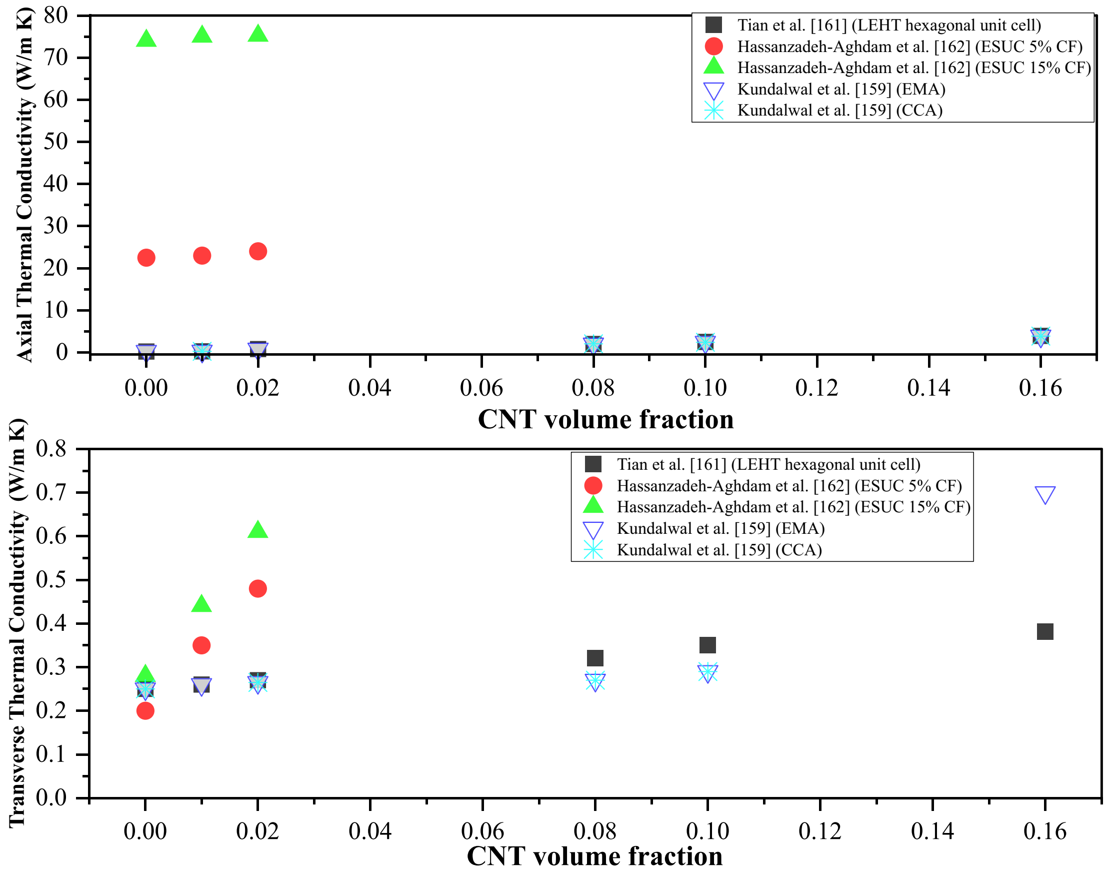

FIGURE 13 Axial and transverse thermal conductivity variation with carbon nanotube (CNT) volume fraction in fuzzy fiber reinforced composites (FFRCs) estimated using different computational methods.

behavior can be observed by (i) increasing the CNTs volume fraction and length, (ii) using straight CNTs and (iii) forming a perfect bonding interface. Figure 13. highlight the improvement in axial and transverse thermal conductivities.

In another work by Kundalwal,[163] micromechanics theory was used for thermal analysis. They investigated the effect of CNT waviness and the existence of interface between polymer and CNT, and its effect on the composite's thermal behavior. The results reveal that (i) the interface between a CNT and the surrounding polymer matrix plays a crucial role in the modeling of the thermoelastic properties of the CNT-based composite, (ii) planar orientation of CNT waviness has a significant influence on the effective CTEs of the hybrid nano-tailored composite, and (iii) for the particular planar orientation of CNT waviness and the value of CNT wave frequency, the effective CTEs of the hybrid nano-tailored composite become zero, making the nanocomposite a "super-insulator".

Kundalwal et al.[164] also did a case study on a heat exchanger made up of fuzzy carbon fiber made by grafting CNT. The model consisted of sinusoidal wavy CNTs grown radially onto a hollow carbon fiber surface. EMA and MOC approach were used for determining the effective thermal conductivity of the fuzzy fiber heat exchanger. It was found that the effective thermal conductivity was unaffected by the interfacial thermal resistance. Also, the transverse thermal conductivity decreases with an increase in temperature, irrespective of the CNT volume fraction and plane of waviness. For a CNT volume fraction of 0.1 and fiber diameter 60 μm, and temperature 300 K increment of 650% in transverse thermal conductivity was found. At a temperature of 400 K, this increment was 300%. Table 5 summarizes the thermal properties of FFRCs evaluated using different modeling techniques.

4.3 | Electrical properties

Park et al.[165] used EPD method to develop MWCNT-grafted carbon fibers. These fuzzy fibers were then used to develop FFRCs and micromechanical models were

TABLE 5 Thermal properties of fuzzy fiber reinforced composites (FFRCs) from computational studies.

Ref.	CNT	Modeling method	Properties (FFRC)	Range
[158]	–	MOC, MT	CTE (Longitudinal)	$0.02–1.1 \times 10^{-5}$ K^{-1}
			CTE (Transverse)	$0.1–7.1 \times 10^{-5}$ K^{-1}
[159]	–	CCA	Effective thermal conductivity	
			Axial ↑	50–800 W/m K
			Transverse ↑	0.2–4.2 W/m K
[160]	–	MOC	CTE (Axial) ↑	$1.2–5.8 \times 10^{-6}$ K^{-1}
			CTE (Transverse) ↑	$2–3.7 \times 10^{-5}$ K^{-1}
			CNT waviness effect	Negative values for coefficient of thermal expansion for some waviness factor
[161]	–	LEHT, FEA, micromechanics	Effective thermal conductivity	
			Axial ↑	0.5–4.1 W/m K
			Transverse ↑	0.22–0.38 W/m K
[162]	–	Semi-empirical HT model	Effective thermal conductivity	
			Axial	30–300 W/m K
			Transverse ↑	0.5–2.2 W/m K
[163]	–	Micromechanics	CTE (Axial) ↑	$1.0–4.3 \times 10^{-6}$ K^{-1}
			CTE (Transverse) ↑	$0.1–1.2 \times 10^{-6}$ K^{-1}
[164]	–	MOC, EMA	Thermal conductivity	
			Axial ↑	10–220 W/m K
			Transverse ↑	100–1400 W/m K
[167]	–	FEA	Thermal Conductivity ↑	5–37 mW/mm K
[168]	MW	FEA	Thermal lightning protection	–

used to estimate the effective electrical properties of the FFRC. It was shown that the FFRC had 78% higher electrical conductivity at 60% weight fraction of the MWCNT than the normal composite without CNT-grafted fibers. These micromechanical results were well validated with the experimental observations. Sha et al.[166] grafted vertical graphene on carbon fibers and improvement in electrical conductivity of such composites was seen. Apart from graphene, silver nanowires were also reinforced in the matrix. The through-thickness electrical conductivity of the composite was increased by 38 times and the in-plane conductivity was enhanced by 39%. FE models showcasing the current flow in the modified carbon fiber composite and normal composites was developed. Garouge et al.[167] used the FE model and micromechanics theories (MT, Self-Consistent theories) to determine and validate the electrical, thermal, and mechanical behavior of FFRC nanosensors. The behavior of composite material was evaluated by three models: full model (FM), half-model (DM) and quarter model (QM). They recorded a significant increase in electrical conductivity and the hardness of the composite with increasing CNT volume fraction. Heat dissipation of composite was also increased

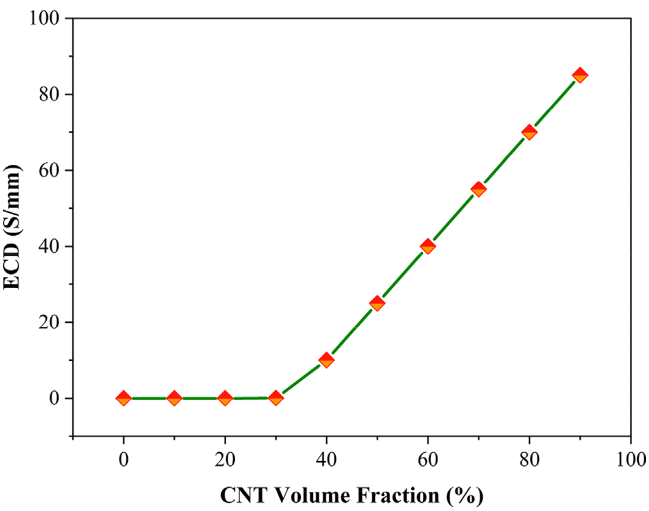

FIGURE 14 Variation of equivalent circulating density (ECD) with carbon nanotube (CNT) volume fraction of fuzzy fiber reinforced composite (FFRC).[167]

in the presence of CNTs. However, a decrease in thermal conductivity was observed with a higher volume fraction, which may be due to the agglomeration of CNTs. Thus, interface interaction and agglomeration must also be considered while modeling FFRCs. Figure 14. shows the electrical parameter that is, equivalent circulating density (ECD) variation with CNT volume fraction. The equivalent circulating density gives the electric current per unit area, leaving the considered surface. It is highlighted by Figure 14. the improvement in the ECD with increasing CNT volume fraction, indicating higher current in the system.

Duongthipthewa et al.[168] developed a system for protecting carbon fiber reinforced composites from lightning strikes using FFRC as the outer ply. They developed a numerical model and varied the composite parameters to understand how various thermal and electrical properties affect lightning damage. By integrating a highly conductive fuzzy fiber carbon-based protection layer into the topmost composite layer, the depth and area of damage caused by lightning can be limited by reducing thermal damage penetration through the underlying composites (Figure 15). This work opens the possibility of using a lightweight and effective anti-lightning strike layer made of fuzzy fibers to protect underlying composites. The use of CNTs helps increase thermal conductivity along both the in-plane and out-of-plane directions, which in turn helps reduce the area of thermal damage and delamination damage in the thickness direction compared to composites without a lightning protection system. This approach has significant potential for future aerospace applications.

Ren et al.[169] characterized the piezoresistive response of FFRC using the 3-D multiscale piezoresistive model and was validated with the single tow piezoresistive fragmentation. For randomly oriented and densely packed CNT, an approximately linear piezoresistive effect is observed within the fuzzy fiber tow region, with gauge factors on an average of 0.14. It was also shown that keeping the CNT volume fraction constant and increasing the interface thickness reduced the effective axial piezoresistivity of the composite. It was also reported that only for CNT volume fraction of 90%, in the case of cylindrically orthotropic interface, is a significant influence on overall piezoresistivity with the addition of CNT seen.

Suresh Kumar et al.[170] used an FFRC facing over a doubly curved sandwich shell for vibration control of the composite shell. The geometrically nonlinear vibration of these sandwich shells was analyzed. The waviness plane CNTs were taken to be coplanar with the carbon fiber plane. The smart doubly curved sandwich shells model was developed using 3-D FE with active constrained layer damping (ACLD) patches. Material properties of the base composite and CNT-reinforced fuzzy fiber composites were determined for different CNT volume fractions indicating improvement with CNTs. Wavy CNTs were found to help more with piezoresistive performance than straight CNTs. 45° alignment of piezoelectric fiber with ACLD patches causes maximum performance of the patches. Chaurasia et al.[171] developed a multiscale computational micromechanics-based approach to study the effect of applied strains on the effective macroscale piezoresistivity of CNT-polymer and fuzzy fiber-polymer nanocomposites. They used FE modeling and electromechanical cohesive zones to model the CNT-polymer interface. The electromechanical cohesive zones allowed for the consideration of interfacial damage in the application of strains.

4.4 | Failure

Gogoi et al.[172] used MD to model the CNT-grafted carbon fiber composite. Two types of CNT grafting were considered: pristine and amine functionalized CNT. Also, effect of defect of CNT on the failure properties of the fiber matrix interface was analyzed. Pull out simulations were generated and it was shown that a 106%increase in IFSS was observed for carbon fibers coated with defect induced amine functionalized CNTs. Such studies involving functionalization and defect in CNTs provide further insight into the parameters of grafted CNT affecting the overall composite behavior at macroscale. MD simulations were also used to analyze the effect of pull-out speed for determining the dynamic fracture toughness of

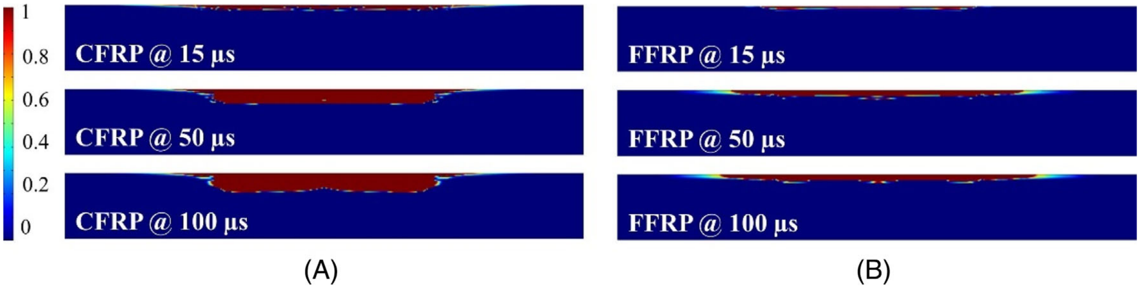

FIGURE 15 Thermal damage depth due to lightning strike modeled in (A) CFRP and (B) fuzzy fiber reinforced composite ply.[168]

nanocomposites.[173] The pull-out speed was found to inversely affect the maximum pull-out force. Xu et al.[174] developed an analytical model to determine the different modes of interlaminar failure of FFRCs. The CNTs were modeled as unidirectional, straight and without defects. The CNT pullout was modeled, and it was found that debonding is initiated when the interfacial shear stress exceeds the IFSS and interface failure occurs, causing increasing displacement. Further, CNT gets completely pulled out of the matrix, and debonding is completed. It was found that the CNT pullout length was half the length of the CNT at its debonding from the carbon fiber surface.

Malekimoghadam et al.[175] modeled a 3-D concurrent multiscale FE model considering debonding damage in FFRCs, covering from nano-to macro-scale. Using mixed-

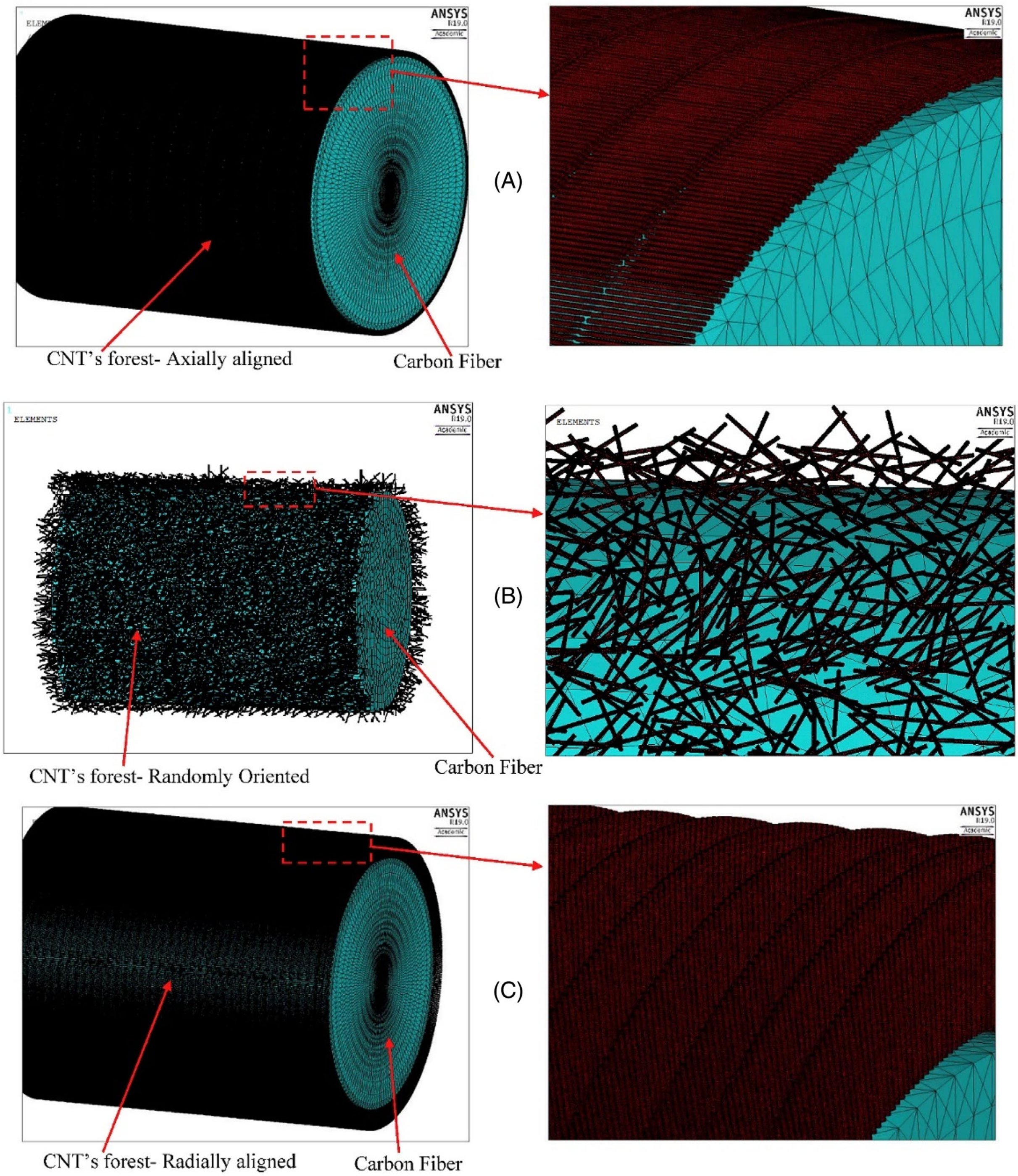

FIGURE 16 FE model of carbon nanotube (CNT)-grafted carbon fiber with (A) axially aligned, (B) randomly oriented, and (C) radially aligned CNTs.[175]

mode traction-separation law, CZM was modeled to analyze the debonding damage between CNTs and the surrounding matrix. MWCNTs were modeled as a transversely isotropic shell structure. Three different configurations of grown CNTs on the fiber surface, encompassing radially, axially, and randomly oriented CNTs, were considered (Figure 16). It was found that the addition of even 2% of CNTs caused a reduction in radial stress by 21.3% and interfacial shear stress by 19.2%. CZM has also been utilized to study interactive properties between CNT and polymer matrix, wherein the interface is modeled using CZM and vdW interaction is modeled using bilinear constitutive law.[176] FE based multiscale model has also been used to model fracture mechanical properties of polymer in presence of CNT. The interface region was simulated considering non bonded interactions and importance on considering CNT lattice structure while modeling was highlighted.[177] Significance of interface modeling has been highlighted for fracture behavior for various other cases as well and FEM has been used to model the interface between matrix and reinforcement.[178,179] Impact behavior has also been shown to be improved upon incorporating CNTs.[180] Using FEM based multi-scale model, non-bonded interactions at CNT matrix interface were captured and it was found that due to presence of CNT the deflection post impact was reduced by 2.5 times when compared to neat resin.[180]

Zhou et al.[181] used the FE model to simulate the FFRC. Also, short beam shearing tests were carried out, and SEM was used to observe the damage evolution. The delamination cracks were propagated along the fiber axes in the fiber interfaces for normal composites. Whereas upon the addition of CNTs, the fiber/matrix interface debonding is improved. The delamination was initiated at the interface between matrix and fiber sizing. An increase in fracture toughness of 36–53% was registered with the use of CNTs in fiber sizing. Thus, showcasing that not only the linear elastic properties are improved, but nonlinear composite behavior such as fracture, delamination and debonding are also significantly affected with the addition of CNTs. Moreover, they provide the multifunctional advantage of improved electrical and thermal properties. All these are summarized in Table 6.

4.5 | Stochastic modeling

Stochastic modeling is an essential methodology used to estimate the impact of random parameters associated with the composite and its constituents through probabilistic analysis. Numerous studies have delved into stochastic modeling of CNT-based composites, wherein a multiscale model was developed to consider various random parameters. These parameters include the length of CNTs,[182,183] random dispersion within the matrix, volume fraction,[182–184] and even factors such as CNT curvature,[183,185] which accounts for the waviness of CNTs and their effects on the properties of the composite material. Rafiee et al.[186] employed a full-range multiscale stochastic modeling approach to graphene-reinforced polymers accounting for all length scales from nano to macro and incorporating randomness due to graphene size, volume fraction, orientation, wrinkle, and

TABLE 6 Electrical and failure characterization of fuzzy fiber reinforced composites (FFRCs) from computational studies.

Ref.	CNT	Modeling Method	Properties (FFRC)
[145]	–	FEA, multiscale modeling	Interface stress: 15–20 MPa, % ↑
			Stress concentration at fiber/matrix interface↓
[167]	–	FEA	ECD: 0.0085–85 S/mm
[169]	–	FEA, multiscale modeling	Piezoresistivity ↑
[171]	MW	FEA, MD, CZM	Piezoresistivity
			Axial resistivity
[174]	–	Analytical modeling	Interlaminar fracture
			Toughness: 500–950 J/m^2
[175]	MW	FEA	IFSS: 53 MPa
			Interfacial radial stress
			Debonding
[181]	MW	FEA	ILSS: 50–65 MPa
			Fracture
			Delamination

formation of agglomerated particles, which are the process-induced uncertainties. In another work, nanoclay polymer composites were studied using stochastic modeling accounting for random parameters such as volume fraction of nanoclay morphologies, number of clay platelets, size and location of inclusions, spatial orientations of particles, and non-uniform dispersion of nanoclay.[187] Rafiee and Mahdavi[188] extended stochastic modeling to explore CNT-polymer interactions, considering random parameters such as the positions and covalent bonding between the polymer and CNTs. Rafiee and Eskandariyun[189] also introduced a novel approach that combines concurrent and hierarchical multiscale modeling, providing an alternative to computationally intensive stochastic modeling methods. Their results exhibited higher accuracy compared to stochastic approaches, suggesting the potential utility of this method as an alternative. While a handful of research articles include results from stochastic modeling of CNT-grafted carbon fibers, there remains a significant gap in this field, especially concerning fuzzy fibers and their composites. Rafiee et al.[190] claim their work is pioneering in addressing stochastic modeling of fuzzy fibers where CNTs are randomly grown on carbon fibers. They also predicted the mechanical properties of isolated fuzzy fibers through a top-down scanning approach, implementing a multiscale stochastic approach that factors in the inhomogeneous dispersion and non-straight shape of CNTs as random parameters.

Stein et al.[40,41] underscored the significance of stochastic modeling of aligned CNTs, incorporating CNT waviness as a random parameter. Meanwhile, Na et al.[191] employed statistical methods to predict multiple fracture number which represents the probability of fiber breakage propagation due to stress concentrations in the presence of broken neighboring fibers in a CNT-grafted carbon fiber composite. Additionally, they estimated IFSS, tensile stresses, and stress concentration factors. The IFSS in the CNT-grafted composite exhibited a remarkable 36.2% increase compared to conventional CFRP, leading to an enhanced stress concentration factor due to improved load transfer at the fiber-matrix interface. Moreover, the multiple fracture number decreased from 18 in CFRP to 12 in the CNT-grafted composite, indicating a reduced likelihood of fiber breakage propagation with the presence of CNT grafting on the carbon fiber.

In another study by Rafiee and Ghorbanhosseini,[192] a hierarchical multiscale modeling procedure was developed to predict the mechanical properties of carbon fibers coated with CNTs. This procedure considered both radially aligned and randomly oriented CNT-reinforced polymers around the carbon fiber, accounting for vdW interactions between CNTs and the polymer matrix. CNT curvature and volume fractions were treated as random parameters in both cases, with an additional consideration for the random variable of CNT orientation in randomly oriented CNT grafted carbon fiber reinforced polymer composites. Mechanical properties of composite laminates composed of these fuzzy fibers were estimated at the macroscale, showing a strong correlation with experimental results, especially for the randomly oriented CNT configuration. Addressing the non-uniformity of CNT volume fractions and non-straight shapes, Rafiee et al.[150] developed a stochastic model based on micromechanical theories such as the MT, CCA, and HT approaches for multiscale modeling of fuzzy fiber-reinforced composites. Neglecting the influence of random parameters in the model was found to lead to an overestimation of effective mechanical properties.

5 | GAP ANALYSIS

The incorporation of CNTs through grafting or coating onto fibers presents an enticing opportunity to augment the functionalities of existing materials, thereby reducing structural redundancy, and leading to benefits such as weight reduction, lower energy consumption, and increased production efficiency across diverse applications. Nonetheless, the manufacturing and processing of CNTs, fuzzy fibers, and their composites involve energy-intensive procedures necessitating heavy machinery operating at high temperatures resulting in environmental concerns and substantial costs. Furthermore, apprehensions persist over the release of harmful gas emissions, toxic byproducts, and solid nano waste into the environment.[193] Developing mechanisms for the repair and recycling of CNTs and their structures is imperative for the commercialization of CNT-based products. Currently, a comprehensive technological model for this purpose is lacking, demanding innovative solutions for production, handling, standardization, repair, and recycling of CNT-based structures. Established conventional procedures must give way to more inclusive approaches. With the amplifying global environmental concerns, it is crucial to ensure that the production and utilization of CNTs and their derivatives align with sustainability goals, rather than contributing to environmental burdens.

Realizing practical applications for CNTs necessitates substantial scientific progress. Particularly, the efficiency of property enhancement from the nanoscale to the micro/macro scale must be practically achievable. Despite theoretical predictions of significant property enhancements, practical outcomes have yet to match these projections. Bridging this research gap is a pivotal step for further advancement. There also exists substantial scope to enhance the bonding between CNTs and carbon fibers by refining deposition parameters, encompassing factors such as deposition density, uniformity, and controlling

CNT agglomeration, length, and diameter. Enhanced controllability is pivotal in grafting and growing CNTs to attain tailored properties. The evolution from single-material-based technologies to multifunctional materials necessitates adaptation and innovation. Modeling FFRCs presents a multifaceted challenge particularly in analyzing geometries across four different length scales. The intricate geometry models, material behaviors, and involvement of multiphysics phenomena amplify the complexity of modeling and analyzing CNT-grafted carbon fiber composites. Despite attempts to simplify interfaces, employ ad hoc assumptions, and estimate property ranges, the pursuit of accurate and consistent results remains ongoing. This underscores that the spectrum of mechanical, electrical, or thermal properties displays considerable variance and altering even a single processing parameter at any scale (nano, micro, meso, macro) can result in a distinctly divergent set of parameters and behaviors in CNT-grafted/modified materials. Consequently, specialized outcomes only relevant to specific material and process parameters emerge. Furthermore, stochastic modeling approaches for fuzzy fibers and FFRCs are still in the process of establishment. Pioneering work has focused primarily on the mechanical properties of these multifunctional materials. Achieving a comprehensive, all-encompassing multiphysics coupled model across all scales remains a distant goal. However, the field is rapidly evolving with an increasing focus on pertinent research.

6 | CONCLUSION

This comprehensive review article offers an extensive overview of fuzzy fibers and their applications in composites, encompassing both their experimental and modeling characterizations. The article delves into diverse techniques employed for modeling and characterizing these fibers, as well as the various production and experimental methods utilized. The integration of CNTs into conventional composites at the fiber matrix interface has demonstrated remarkable potential in enhancing their elastic, thermal, and electrical properties. Additionally, CNT incorporation imparts multifunctional attributes like electromagnetic shielding and piezoresistivity. While advancements are evident in the longitudinal aspects, the transverse properties exhibit even more substantial enhancements. CNTs also contribute to improvements in the IFSS, delamination behavior, debonding resistance, and fracture toughness of composites. This is primarily attributed to their radial growth pattern, which imparts superior performance in off-axis directions and improves fiber-matrix bond by improved wettability and contact area. Notably, the modification of interfaces emerges as a pivotal factor contributing to the overall advantages of CNT integration. The review concludes by emphasizing that despite the burgeoning interest in employing CNTs in fuzzy fibers, challenges still exist and warrant further exploration. The article briefly touches upon the prospective scope of future research and life cycle assessment of CNT-grafted carbon fibers.

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DATA AVAILABILITY STATEMENT

Data sharing not applicable—no new data generated, or the article describes entirely theoretical research.

ORCID

Renuka Sahu https://orcid.org/0000-0002-1090-9475
Sathiskumar A. Ponnusami https://orcid.org/0000-0002-2143-8971
Christian Weimer https://orcid.org/0000-0002-1815-6578
Dineshkumar Harursampath https://orcid.org/0000-0001-6855-303X

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