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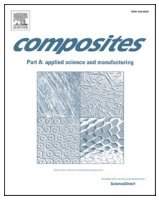

Static and dynamic mechanical properties of hybrid polymer composites: A comprehensive review of experimental, micromechanical and simulation approaches

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ABSTRACT

Over the last decade various investigations have been carried out on experimental and numerical approaches to predict the influence of dual nanofillers (DNFs) and multiscale fillers (MSFs) in polymers. DNFs and MSFs reinforced polymer composites exhibit exceptional mechanical and thermomechanical properties by designing and controlling the material and process parameters. This paper discusses the current state of art of the manufacturing methodologies, influential parameters, micromechanical models and simulation approaches for static and dynamic mechanical properties of hybrid polymer composites. An extensive literature review is conducted to discuss experimental and micromechanical modulus of composites. Multiscale modelling strategy to take into account the simultaneous inclusion of DNFs and MSFs of different length scales using stochastic FEM modelling techniques have been reviewed. This review serves as a stand-alone reference for investigations on experimental, micromechanical models and multiscale simulation strategies of static and dynamic mechanical properties of DNF and MSF reinforced hybrid polymer composites.

1. Introduction and state of the art

The development of advanced smart materials to replace the existing conventional materials and composites with superior materials of multifunctionality has been an area of wide research interest [1]. Ceramics and metals have been already replaced in many applications with polymers owing to its lightweight, low cost, ease of processing, high ductility and corrosion stability. However, conventional polymers face many challenges like inferior tensile strength, modulus, resistance to fracture, low thermal conductivity and stability, toughness etc. [2]. In order to overcome these shortfalls, polymer matrices can be reinforced with different types of fillers at any scale, macro, micro and nanoscale. The reinforcements embedded in a continuous matrix phase can be in the form of particles, fibers, rods, tubes, flakes or platelets, organic or inorganic, synthetic or natural [3]. Multiscale composites are a class of advanced materials in which more than one discontinuous phase (at both micro- and nano-scale) are dispersed in a continuous polymer phase and a distinct interface separates the constituents [4–6]. Over the past two decades, dual nano scale (two different fillers of nano size) filler reinforcements in a single matrix also gained considerable attention owing to its remarkable properties over the fiber reinforced composites. The performance of the material could be tailored with respect to the individual key features of the chosen fillers and the parent base polymer. The synergism of the fillers in the matrix and the characteristics of the interface developed in the matrix determine the performance of final composites. Once a conventional fiber incorporated in a matrix is replaced with nano filler, considerable reduction in the weight of the prepared component could be observed with reasonable mechanical properties. In addition, the ductility of the polymer could be improved with the inclusion of nano reinforcements rather than resulting in brittle fracture observed in micro filler reinforced composites. Instead of a single reinforcement in the matrix, multiscale (fillers of different length)

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or dual nano reinforcements in the polymer always exploit the advantages of individual fillers and the additive contribution of these fillers are seen to remarkably enhance the performance of the materials [7]. These dual nano scale and multiscale composites have potential applications in all sectors where conventional materials are used, owing to its lightweight, high aspect ratio of fillers/fibers, high mechanical properties, durability and reasonable cost [8–10]. Even though micro fillers are less expensive, weight of the products is very crucial in sectors like automobile, structural, aerospace and military applications [11–13]. Hence, the conventionally used micro and macro scale fillers could be replaced with nano fillers or one could reduce the composition of micro fillers in the matrix by incorporating nano filler along with micro filler. Several studies have reported the property enhancement achieved in dual nano scale and multiscale filler reinforced composites with respect to the conventional composites [14–17]. Pedrazolli et al. [18] reported the hybridized effect of fumed nanosilica and graphene nanoplatelets along with short glass fiber in polypropylene thermoplastic matrix. These authors varied the glass fiber content from 5 to 20 wt% and analyzed the synergism of nanofillers with glass fibers (micro fibers) and studied its effect on microstructure and thermo mechanical properties of the multiscale composites. The study demonstrated improved strength and modulus with multiscale fillers and exhibited higher values of creep stability and dynamic modulus in the presence of nanofiller along with short glass fibers. Further, they investigated the interfacial shear strength, analyzed the failure behavior of the composites, and the dominant failure mechanism prevailed in the composites. It was noticed that, even the minimal inclusion of nanofiller improved the adhesion between the filler and the matrix which enabled better stress transfer at the fiber matrix interface. The stronger interfacial strength achieved due to the synergism of fillers in hybrid composites resulted in positive enhancement in the viscoelastic behavior of the composites [18]. Thostenson et al. [17] outlined the synergism of carbon nanotubes along with carbon fibers and the dispersion of carbon nanotubes around the fibers. The matrix/fiber interfacial characteristics were evaluated using fragmentation tests. It was observed that the interfacial strength of the hybrid multiscale composites enhanced due to the presence of nanotubes at the interface of matrix/fiber. They inferred that at the persistent fiber strength and diameter, the smaller fragment lengths impose strong interphase characteristics between the filler and matrix [17]. Reza et al. [19] reported significant enhancement in the impact resistance with the addition of 3 and 5 (volume %) of CNT into polyethene reinforced carbon fiber composite. The authors followed a multiscale stochastic approach using FEM (finite element method) to evaluate the impact and mechanical properties of representative volume elements (RVE's). The effect of parameters like curliness of carbon nanotubes, the L/D ratio of carbon fiber and CNTs, volume percentage of fillers on the tensile properties of carbon fiber/carbon nanotube/polyethene multiscale composites was delineated [19]. The compounded effect of untreated and treated carbon nanotube/ glass fiber multiscale fillers in polypropylene (PP) matrix was investigated by Gamze et al. [20]. The multiscale composites were prepared after surface modification of nanotubes using silane coupling agents which was verified using FT-IR and XRD analysis. The authors reported better tensile strength and modulus for surface treated glass fiber (GF)/ carbon nanotube (CNT)/ PP multiscale composites in comparison to PP/GF or PP/CNT composites. The simultaneous hybrid reinforcement effect of multiscale fillers resulted in positive enhancement in dynamic and static mechanical analysis results, which proves the reinforcement effectiveness of nanotubes in the matrix [20]. In another study [21], carbon nanotubes were introduced into carbon fiber wet laid polyamide composites and improvement in both mechanical, electrical and thermal properties were observed. The synergism of fiber and CNTs significantly enhanced the interfacial adhesion with matrix which in turn enhanced the mechanical and electrical conductivity [21].

Josime et al. [22] reported the inclusion of nano Al_2O_3 and clay nano platelets in cross linked polyethylene (XLPE) resulting in the hybrid effect of dual nano fillers in binary and ternary hybrid systems. The interaction between alumina and clay have created unique complex filler architecture with regard to the idealized surface morphology of conventional composites. The morphology of the composites could be controlled by adjusting the ratio of two different nano fillers and the synergism between nano fillers (filler-filler networks) leads to superior microstructure development [22]. In another study [23], authors discussed the synergism of cellulose nanocrystals (CNC) and graphene oxide nanosheets (GON) in poly vinyl alcohol (PVA) base matrix. The synergism enhanced the dispersion homogeneity avoiding agglomerations and the hybrid filler content at 5 wt% (1: 2 ratio) enhanced modulus, tensile strength and toughness and maintained reasonable ductility for PVA [23]. In another work [24] involving PVA polymer it was mentioned that the incorporation of exfoliated graphene oxide (GO) sheets enhanced the distribution and dispersion of CNTs in PVA matrix owing to the strong GO-CNT interaction and the complex scroll like structure developed due to synergism between fillers. This hybridized effect exploits the advantages of individual nano filler and shows superior mechanical properties in comparison to PVA composite films embedded with GO or CNT alone [24]. The extremely small dimensions of nanofillers leads to its superior characteristics like low weight, high aspect ratio, and exhibits specific characteristics like insulative, conductive, flame retardancy etc. The size, geometry (0D,1D, 2D) of nanofillers and the processing techniques, optimization of filler content in the base matrix are some of the prime influential parameters that decides the overall performance of the dual nanocomposites. The optimal composite performance can be achieved, by improving the dispersion quality and the interfacial bonding amongst the fillers and the matrix. To this effect, researchers have introduced functional moieties that could elevate the anchoring between the filler and the matrix [20,25–27].

This review comprehensively discusses the choice and formulations of polymer nano and multiscale hybrid composite systems and the relevance of co-existence of dual nanofillers or multiscale fillers in thermoplastic/ thermoset polymer matrices. The various processing methodologies for the preparation of aforementioned composite systems are discussed and elaborated on the technologies which was most industrially implemented that could improve the operational efficiency and multifunctionality of the composites. The challenges related to the uniform and individual dispersion of nanoparticle aggregates to achieve ultra-large interfacial area per volume, constraints/difficulties in the preparation and commercialization of prepared composites are also elucidated. Numerous works in literature demonstrating the substantial enhancement in multifunctional properties of DNFs and MSFs reinforced polymer composites were studied and cited. This review documents the complex morphological structures developed due to synergism of multi scale and dual nano fillers and interprets its correlation with dynamic and static mechanical behavior of the composites. The experimental observations are verified with several micromechanical and simulation approaches for the dynamic and tensile properties of the composites. In addition, the issues related to the multiscale and dual scale nano filler dispersion, adhesion characteristics owing to the surface treatment of fillers, and the synergistic effect accountability of multiscale and dual nanoscale fillers in theoretical modeling and simulation modeling approaches are also explored. This review comprehensively documents the micromechanical models used for the evaluation of static, dynamic mechanical properties and discusses some of the relevant works involving multiscale finite element modeling strategies, representative volume element method (RVE) based micromechanical modeling of polymer composites.

The uniqueness of novel DNFs and MSFs polymer nanocomposite systems will enable the replacement of traditional filled polymer composites by utilizing the new properties imparted by the DNFs and MSFs and exploiting the synergism between the fillers which could occur, when the nanoscale morphology and the fundamental physics connected with a property coincide. The excitement surrounding DNF and MSFs

polymer nanocomposites science and technology provides unique opportunities to develop multifunctional revolutionary materials which represents a radical alternative to the traditional macro and micro filler composites.

2. Development of hybrid nano and multiscale filler reinforced polymer composites

2.1. Selection of hybrid nano and multiscale fillers

It was detailed in many studies that the size of fillers incorporated as reinforcements in polymer matrices has a profound effect on the final performance of the composites [28–30]. The extent of dispersion, bonding, adhesion amongst the matrix and the filler, surface interactions between the filler and the matrix are all dependent on the size of the fillers used [31]. The aforementioned effects improve as the filler size decreases and hence incorporation of dual nanoscale fillers or single nano filler along with a micro filler plays a prominent role in the development of advanced materials [32]. Fillers of nano dimensions possess a very large surface area to volume ratio and the properties such as electrical resistivity, chemical reactivity, catalytic reactivity, adhesion depend on the nature of interface. In addition, at nanoscale, the confinement of electrons, molecular motion, energy quantization, and electromagnetic forces become very active. This leads to improved intermolecular bonding, hydrogen bonding, van der Waals effect, hydrophobic effect, catalytic, magnetic effect, surface energy etc. The key characteristics of nano dimension reinforcements are shown in Fig. 1 [33]. These prevalent effects yield dramatic results during the development of dual scale nanocomposites and multiscale composites [34–35].

Examples of reinforcing nanomaterials are carbon nanotubes, nanosilica, clay nanoplatelets, halloysite nanotubes, graphene nanoplatelets TiO_2, ZnO, Al_2O_3 etc. [36–40]. Each nanofiller possess specific characteristics based on the structure and the dimensions, say for carbon nanotubes owing to the one-dimensional structure of CNTs electrons are confined across the diameter where electrons are free to move (electron delocalization) throughout the length of the CNT [41–42]. Hence, CNTs exhibit high thermal conductivity on account of the coordinate vibrations of carbon atoms which produces heat conduction. In addition, CNTs possess high aspect ratio with nano diameter and length in microns and based on zig zag, chiral and arm chair configurations and number of walls in MWCNTs, desired characteristics could be achieved [43–44]. Likewise, each nanofiller offers one major specific characteristic for example, nanosilica (insulative), halloysite nanotube (flame retardancy), graphene (electrical, optical) etc. [1,45–46].

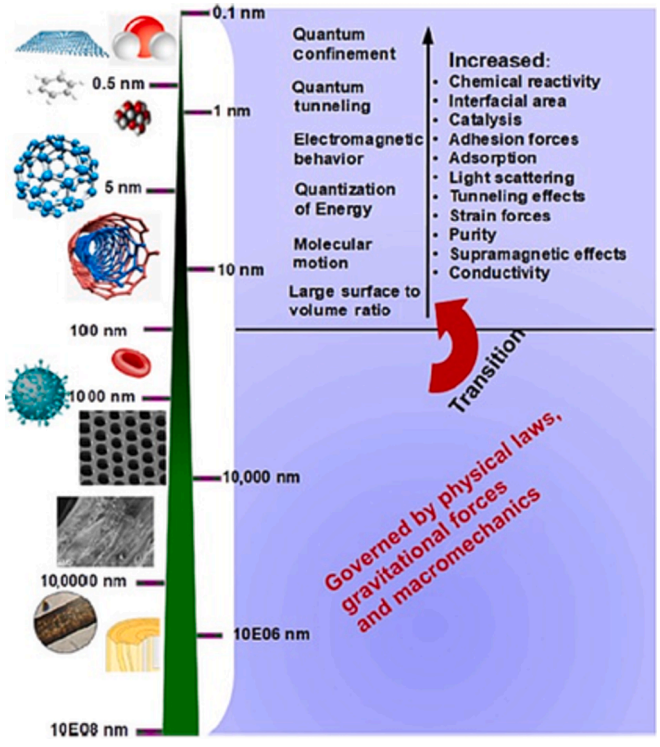

Fig. 1. Characteristics of nanoscale fillers. Reprinted with permission from Elsevier [33].

Some of the major micro scale reinforcements include glass fibers, aramid fibers, carbon fibers, basalt fibers etc. [47–50]. Glass fibers are lightweight, strong and robust which are preferred mostly as micro scale filler due to its low cost and process friendly nature. Glass fibers are inorganic materials that manifest good fire resistance and high strength that could be utilized to produce light weight advanced materials for automobile or aircraft components [51]. On the other hand, carbon fibers possess outstanding mechanical, thermal and electrical properties, and the resultant carbon fiber incorporated composites demonstrates low density, improved stiffness, excellent electrical conductivity, chemical stability and coefficient of thermal expansion which are applicable in the design of components for automobiles, defense equipment's, air vehicles and sport goods [52–53]. Another class of fiber which possesses high modulus, strength, resistance to corrosion and temperature are basalt fibers which are referred to as green industrial material. The reasonable cost and process friendly nature makes it a better choice than carbon fiber reinforced composites [54–55]. Organic fibers with impressive stiffness and strength are another group of micron size fibers which could even replace asbestos. These materials exhibit outstanding heat and flame resistance properties, mechanical properties, abrasion resistance and could be used in load absorbing applications at elevated temperature [56–57]. Aramid fibers are lighter than glass fibers, and shock resistance, energy absorption prior to failure are their attractive features.

Even though intensive research works were carried out in the field of nanocomposites, the mechanical properties achieved are not so exciting in comparison with conventional micro composites. Another major challenge is the dispersion of nano reinforcements as they have a natural tendency to bind together and form aggregates due to weak Van-der Waals forces of interaction [2,58–60]. In the case of conventional micro composites, abrupt brittle fracture occurs during loading, and the large volume fraction of fillers have to be included to acquire exciting mechanical properties. This always poses a serious challenge in the production of large structures using micro composites.

Several approaches were unfolded in the development of composite structures involving the co-existence of dual nanofillers or multiscale (micro-nano) fillers in the matrix. These approaches could utilize the advantages of individual fillers and could mitigate the less desirable properties. The collective contribution of dual nano or multiscale fillers in the polymer matrices is expounded as hybrid effects [61–62]. The number of investigations on hybrid multiscale filler reinforced composites and dual nanoscale filler reinforced composites have increased recently owing to the attractive properties applicable to various demanding applications [63–64]. Fig. 2 explains the dispersion of dual nanofillers (0D and 1D) in dual nano hybrid composite system and in multiscale composite system where different scale fillers (micron scale (glass fiber) and nanoscale (CNTs, nanosilica, nano clay graphene etc.)) are involved. In the morphological analysis of dual nanofiller and multiscale filler reinforced composites, one could observe both loose and dense packed aligned nanofiller over the other filler. The various reasons for the difference in bonding of one filler over the other are mentioned in Fig. 2.

Table 1 shows the list of research work reported in the field of dual nano and multiscale hybrid composites in the past ten years. Various processing methodologies and their merits are reported in Table S1 under the supplementary section S1. Hongkun et al. [65] reported a

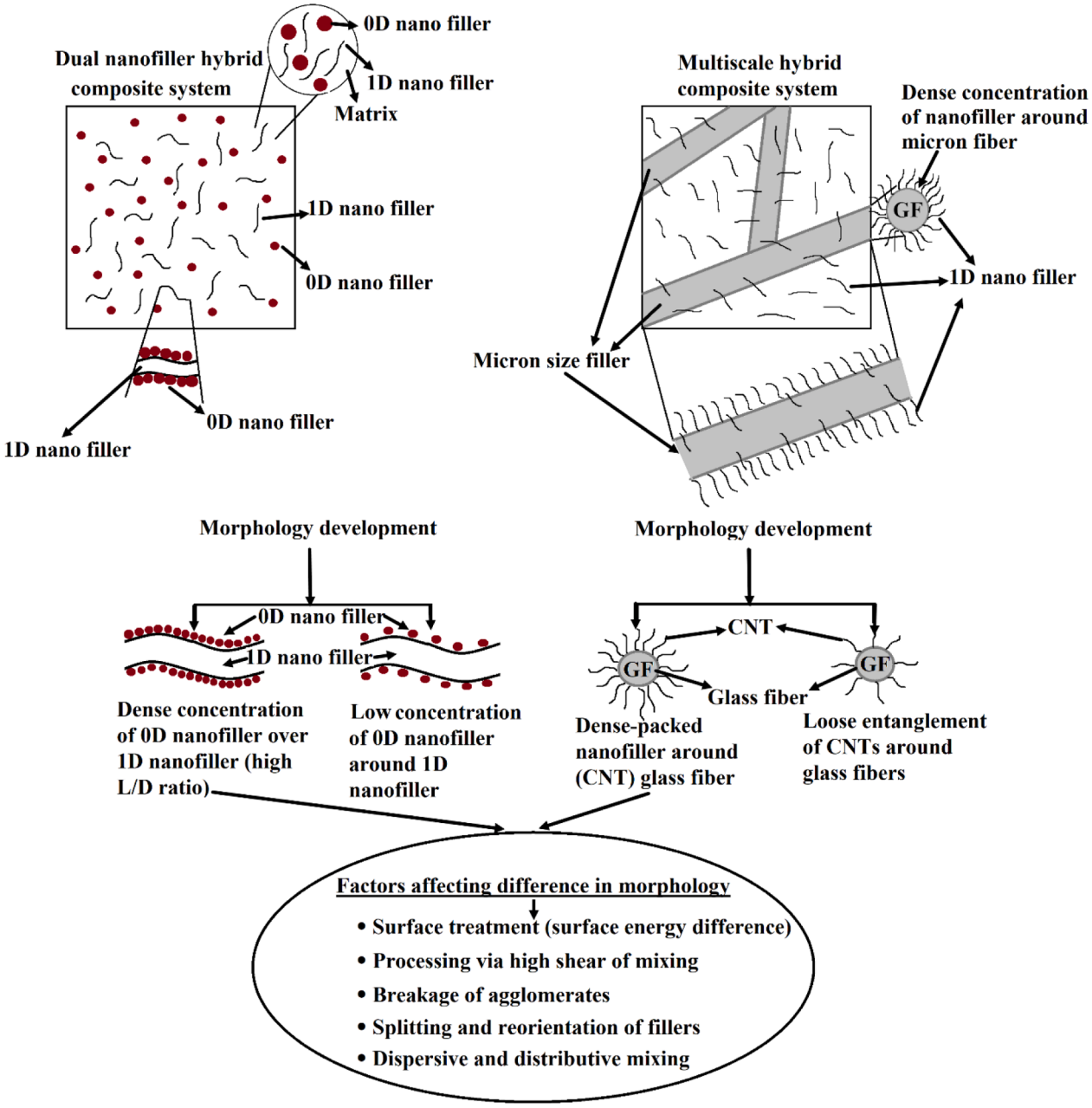

Fig. 2. Scheme representing the distribution and dispersity of dual nano and multiscale fillers in the matrix leading to different morphologies.

facile approach for the preparation of multifunctional iron oxide nanoparticle attached graphene nano sheets in polyurethane matrix. The integrated properties of FeO nanoparticles along with graphene nano sheets in polyurethane hold great promise in fields like magnetic resonance imaging, microwave absorbing and electromagnetic interference shielding. It was elucidated that the multihydroxyl groups on Fe_3O_4 nanoparticles leads to highly reactive nature when attached to graphene nanosheets creating 2D nanoplatform [65–66]. It was observed that tensile modulus enhanced from 1.2GPa to 2.2 GPa and tensile strength enhanced to 150 MPa with regard to 76 MPa of epoxy/glass fiber composite with the addition of 0.3 wt% of r-GO [72]. The enhanced flexural properties are assigned to the higher interfacial area which enables effective stress transfer between the filler and the matrix. The high aspect ratio of nanofillers can enhance the interfacial bonding among the filler and the polymer and restricts the movement of polymer chains, which improved the tensile modulus and flexural properties [72,83].

A recent report [73] addresses the multiscale modeling of epoxy hybrid composites containing carbon nanotubes and carbon fibers and molecular dynamics (MD) simulations are performed to interpret the interfacial strength at a molecular level. In another study [74] of the same composite system, effective electrical conductivity and electrical percolation threshold increased even at low content of short carbon fiber in the presence of CNTs owing to the high aspect ratio of multiscale fillers [74]. Thostenson et al. [84] reported that carbon nanotubes are grown on individual carbon fibers which resulted in stiffening of the interfaces of filler/matrix aiding effective load transfer. Wang et al. [64] reported that uniform dispersion of CNTs in epoxy matrix along with glass fibers was achieved using surfactant and ultrasonication. The surfactant molecules are physically adsorbed on the surface of

Table 1
List of research work carried in dual nano and hybrid multiscale polymer composites.

	Base polymer/ Reinforcements	Processing/ Surface functionalization	Effect on properties	References
Dual nano filler reinforced polymer composites				
1.	Polyurethane/ Graphene nanosheets/ Fe_3O_4	Solution processing technique	Strong supraparamagnetism, electrical conductivity, high chemical reactivity, good solubility	[65–66]
2.	Epoxy/ halloysite nanotubes/ reduced GO	Exfoliation/ reduction/ curing/sand milling	Enhanced tensile strength, storage modulus, fracture toughness	[67]
3.	Polyamide 6 (PA 6)/ Graphene/CNT	In-situ polymerization/ melt spun	Functionalized graphene (0.2 wt%) + CNT (0.3 wt%) enhanced tensile strength by 2.4 times and Young's modulus by 132% than PA6	[68]
4.	Polyimide (PI)/ Graphene Oxide (GO)/ Carbon nanotube (CNT)	Vacuum filtration solution hybrid dispersion/in situ polymerization	At GO:CNT ratio of 3:1 in PI excellent mechanical, tribological properties and thermal stability and achieved low coefficient of friction	[69]
5.	Polyurethane (PU)/ functionalized CNT (fcnt)/ graphene nanoplatelets (fGnP)	Solution mixing	At fGnP: fCNT ratio of 3:1 tensile modulus enhanced by 86.5% and tensile strength by 30% and reduced electrical resistivity	[70]
6.	PU/CNT/ Graphene nanoplatelets	Ultrasonication/ solution mixing	Tensile strength of PU increased by 43% at GnP: CNT content of 0.25 wt% (1:1 ratio)	[71]
Multiscale hybrid polymer composites				
1.	Epoxy/glass fiber/r-GO (reduced graphene oxide)	Compression molding and hand lay-up method	The inclusion of 0.3 wt% of r-GO nanofiller improved the tensile strength by 97% and flexural strength by 44.5%. Ductile fracture mechanism was observed owing to the presence of dual nano fillers.	[72]
2	Epoxy/carbon fiber/carbon nanotube	Molecular dynamics simulation	The addition of nanotubes in short carbon fiber reinforced non conducting polymer composites enhanced the modulus, strength, fracture toughness, electrical and thermal conductivities.	[73–74]
3	Polyimide/ carbon fiber/ carbon nanotube	Chemical method/ Hot pressing technique	Excellent friction and wear properties	[48]
4	Epoxy/carbon fiber/carbon nanotube	Chemical vapor deposition	Enhanced interfacial shear strength confirmed with the presence of nanotubes at matrix/fiber interface	[17]
5	Epoxy/glass fiber/carbon nanotube	Resin transfer molding	Surfactant modified CNTs promotes CNT percolation network, higher glass transition temperature, enhanced dynamic and static mechanical properties of hybrid composites	[64]
6.	Epoxy/Glass/ MWCNTs/GO nanoplatelets	Hand lay-up method -Composite laminate plates of glass epoxy laminate-Nano fillers are added to resin followed by mechanical stirring and ultrasonication	Enhanced initiation fracture energy of 179% (aligned MWCNTs), 349% (aligned GONPs) and 66% and 127% increment in maximum load, in reinforcement of composite adhesive joints	[75]
7	High impact Polystyrene (HIPS)/short silane treated glass fiber/ Aluminium trihydroxide micro filler/ nanoclay	Melt compounding using a micro compounder	In the presence of hybrid fillers at different length scales interfacial shear strength at filler/ matrix regions has varied.	[76]
8.	PP/Glass fiber/ Organo modified layered silicates/ micrometric calcium carbonate	Melt blending in internal mixer, followed by preparation of thick films of composites via hydraulic hot press compression molding.	In hybrid composite flexural modulus enhanced by 60% and flexural strength by 130%, appreciable enhancement in storage modulus	[77]
9.	Polypropylene/ Glass fiber/ inorganic fullerene and nanotubes Polyphenylene Sulphide/ Carbon fiber/ inorganic fullerene and nanotubes	Melt processing and compression molding with a hydraulic hot press	Enhanced wear, thermal stability, storage modulus	[78]
10.	PP/Glass fiber (40 wt%) /ZnO (2 wt%)	High speed mixer followed by twin screw extrusion	Enhanced crystallization temperature and crystallization rate of PP	[79]
11.	PP/Glass fiber/ SiO_2	Melt Mixing via twin screw extrusion	Enhanced tensile and crystallization properties	[80]
12	Epoxy/Glass fiber/MWCNT/ SiO_2 or Fe_3O_4 nanoparticle	Dip coating of nano SiO2 and Fe3O4 on MWCNT grafted glass fiber and embedment in epoxy matrix	85% increase in IFSS (CNT/Fe_3O_4 nanofiller system) and 75% increase in IFSS (CNT/SiO_2) in comparison with glass fiber micro composite, Generation of multifunctional composite interfaces	[81]
13.	PP/Glass fiber/ melamine cyanurate fumed silica	Melt blending in two roll mill /hot compression	Enhanced flame retardancy PP/GF/ IFR-SiO_2 at SiO_2 content of 20 wt% with LOI of 32.4%	[82]

nanotubes which cause a steric repulsion force and overcome van der Waals forces of attraction. They mentioned that glass fibers are muffled by curly long CNTs which offers CNT percolation network in hybrid multiscale composites and creates a conduction path [64]. Likewise, the surface treatment and synergism of multiscale and dual nano fillers have been envisaged which have paved the way to the novel investigations on behavior of multi and nanoscale composites as next generation materials using cutting-edge technology.

2.2. Manufacturing methods

Mia et al. [67] proposed the preparation methodology of epoxy/ halloysite nanotubes (HNTs)/ reduced graphene oxide (RGO) dual nanoscale composites as a two-step process. Initially HNT/GO hybrids are prepared by the addition of GO and HNT s suspension of weight ratio 1:1 in a flask followed by half an hour magnetic stirring (exfoliation and reduction). Then the suspension is treated with hydrazine hydrate and heated to 90 ^{o}C for 1 h. Further the product was washed, centrifuged and dried in vacuum (60 ^{o}C). In the next step, sand milling method is used to prepare suspensions of epoxy composite via zirconia balls (1000 rpm) as milling media and added curing agent and degassing [67]. Another group of researchers [69] prepared CNT/GO hybrids by solution dispersion method followed by ultrasonication and vacuum filtration. In the first step, CNTs were treated with mixed acid solution (HNO_3: H_2SO_4 volume ratio of 1:3) to attach carboxyl functional groups on CNT and graphene oxide by modified Hummer's method. The carboxyl attached CNTs and GO of varying weight ratios are then added to ethanol by ultrasonication followed by mechanical stirring and vacuum drying at 60 ^{o}C for 10 hrs. Then CNT/GO hybrids are ultrasonically dispersed in dimethyl acetamide (DMAC) followed by addition of oxidianiline (ODA) and pyromettalic dianhydride (PMDA) to form GO/CNT/PAA (poly amic acid) solution. In the next step GO/CNT/PAA nanocomposite are coated on the glass substrates and vaccum dried at 70^{o} C and thermally treated from 100 to 300 ^{o}C for 30 min to remove volatiles and complete imidization reaction to obtain GO/CNT/PI films [69].

Zhou et al. [68] discussed the preparation of dual nanocomposites incorporating functionalized CNTs/functionalized graphene powder in polyamide 6(PA6) by in situ-ring polymerization to form PA6/fCNTs/fG composites. Graphene oxide was functionalized with styrene maleic anhydride (SMA) using modified hummer's method and CNTs were functionalized with hydroxyl groups. In the next step, functionalized graphene and CNTs are reacted with aminopropionic acid and caprolactum (ring polymerization) under ultrasonication and then the mixture temperature was raised to 270 ^{o}C for 9 h and samples were cooled to room temperature [68]. The composites fibers are then melt spun at 270 ^{o}C using piston spinning machine. Amir et al. [71] delineated the methodology of preparation of PU foams reinforced with dual nanofillers (MWCNTs and GnPs) of varying content. The nanofillers at varying content are added to polyol and ultrasonically dispersed at 2000 rpm for 5 min. PUs fabricated by mixing polyols and isocynates are added to nanofiller/polyol mixture at specific ratios to form PU/GNP/CNT hybrid dual nanocomposite [71]. Similar preparation methodology for fabrication of polyurethane/f-graphene nanoplatelets (fGnP)/fCNT composites and functionalization of fillers were explained by Rostami et al. [70].

Wang et al. [85] reported the fabrication methodology of epoxy/CNT/carbon fiber hybrid multiscale composites by compression molding technique. Initially, predetermined quantities of CNTs kept in acetone were agitated via tip sonication (15 min) and mixed with known quantities of epoxy resin. In the next step, to evaporate acetone solvent fully, the mixture of epoxy, carboxyl functionalized CNTs and the solvent acetone was kept in a water bath (60 ^{o}C) followed by magnetic stirring. In the dispersion of CNTs/curing agent/epoxy resin carbon fiber fabrics were dipped to prepare a prepreg of 8 layers which gives 48.9% fiber volume content and pressed and cured from 4 to 8 MPa with temperature ranging from 80 to 200 ^{o}C using a hot compression press [85].

In another work [48] carbon fiber-nanotube (CF-CNT) hybrid was prepared and incorporated in PI resin using hot pressing technique. Firstly, amino groups are chemically and physically adsorbed on to the carbon fiber, and –COOH treated CNTs were dispersed in acetone and kept under reflux for 12 h with magnetic stirring and achieve the completion of reaction between –NH_2 on –NH_2-CF surface and –COOH on –COOH-CNTs surface respectively. PI resin and the prepared CF-CNT hybrids are mechanically blended and filled in the mold and then heated to around 350 ^{o}C at a pressure of 30 MPa. Subsequently, the mold was naturally cooled below 100^{o}C and PI/CF-CNT hybrid multiscale composite was released from the mold [48]. In another work [84] by chemical vapor deposition method, carbon nanotubes are decorated on carbon fibers by sputtering a layer of catalyst on the carbon fiber surface. A hot tube furnace loaded with catalyst coated carbon fibers was used to grow CNTs on the carbon fiber surface. Carbon fibers surrounded by sheath of nanotubes were further incorporated in the matrix to form hybrid multiscale composite [84]. Another group of researchers [64] followed the resin transfer molding (RTM) wherein CNT/epoxy dispersion was injected into the closed die where plies of glass fibers are preplaced. Then the RTM assembly was cured for 80, 120 and 200^{o} C each at 2 hrs to prepare hybrid multiscale composite with varying content of CNTs.

In the preparation of thermoplastic polypropylene/glass fiber/CNT multiscale composites Karsili et al. [20] followed by a melt mixing technique via twin screw extrusion. CNTs are first treated with HNO_3 –H_2SO_4 (1:1) acid mixture and attached –COOH groups on CNTs are then treated with silane (3-Amino propyl tri ethoxy silane). Silane groups are attached on carboxyl treated MWCNTs and are further melt mixed with glass fiber and polypropylene in a co-rotating twin screw extruder [1]. Compounding of PP with glass fibers and MWCNTs were performed at 230 ^{o}C at screw speed of 100 rpm followed by injection molding of standard test specimens [20]. Pedrazzoli et al. [18] also reported the preparation methodology of PP reinforced with glass fibers along with graphene nanoplatelets and nanosilica particles. The silica nanoparticles (untreated and treated) with dimethyl-dichlorosilane and the exfoliated graphene nanoplatelets are melt mixed with silane treated glass fibers and then compression molded with hot press maintaining a temperature around 190 ^{o}C [18]. Taraghi et al. [86] discussed the preparation of PP/glass fiber/CNT hybrid multiscale composites. Initially, PP was fully melted and stabilized with the compatibilizer PP-grafted maleic anhydride (PP-g-MaH). The PP/MWCNT mixture which was premixed in the tumbler initially, was then melt mixed in the extruder. To achieve a uniform composition of mixture, glass fibers were slowly added to the melt mix in the extruder. The extrudate was then an injection molded to standard test specimens [86]. The typical melt compounding process for the preparation of dual nano and multiscale hybrid composites is presented in Fig. 3.

2.3. Parameters influencing the properties of dual nano and multiscale filler reinforced composites

Polymer Matrix: The choice of polymer plays an important role in the preparation of dual nanoscale and multiscale composites. The continuous matrix phase protects the reinforcing fillers and fibers from external load, abrasion, thermal and other environmental conditions. The matrix holds the fillers and fibers firm within the matrix and absorbs the maximum amount of energy when it undergoes deformation upon loading and enables effective transfer of stress effectively from matrix to reinforcing fillers. Considering the requirements, it is preferable to choose a matrix that could deform to its maximum before failure. An array of thermoplastic materials are available that ranges from cheap commodity plastics to high end specialty ones. The basic drivers for the wide adoption of thermoplastic resins are due to its recycling options, enhanced impact properties, unlimited shelf life and wide processing window. One of the drawbacks associated with thermoplastics is its high

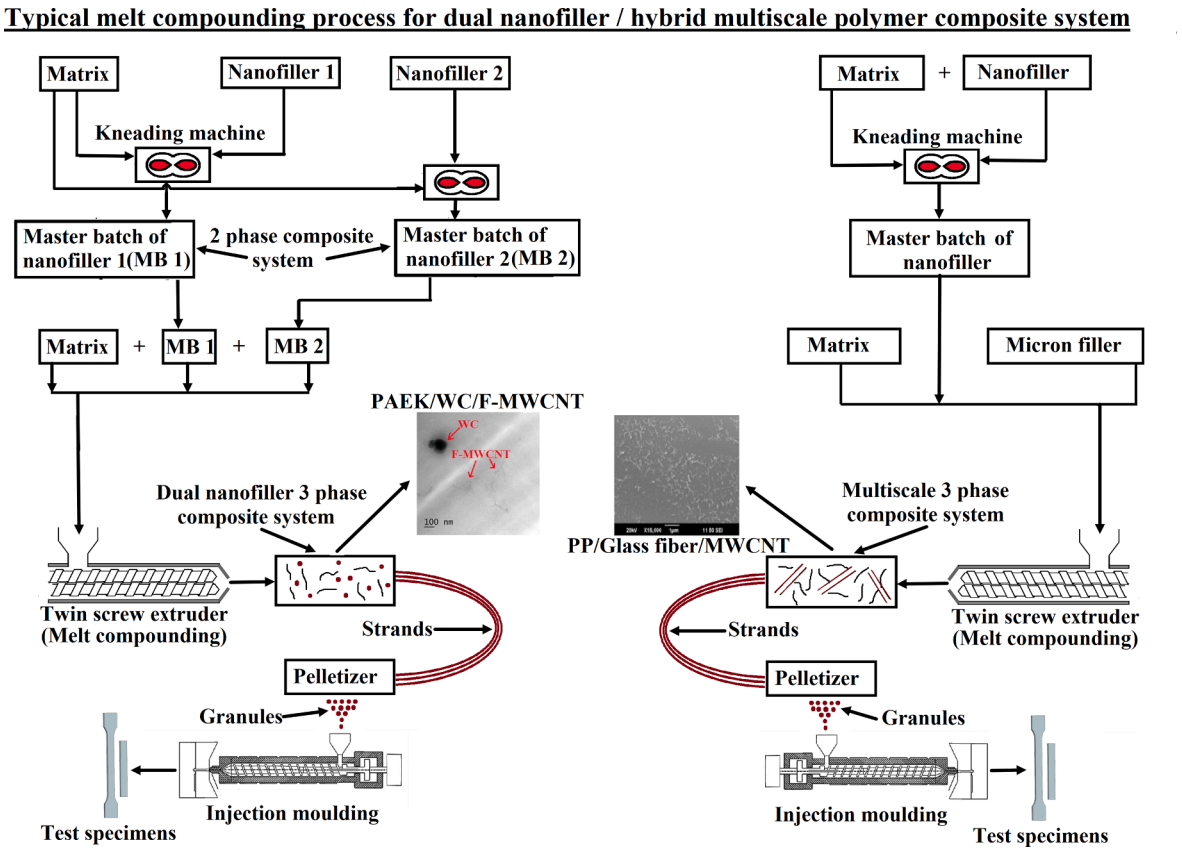

Fig. 3. Schematic representing melt compounding process including masterbatch preparation, compounding in extruder and injection molding to test specimens.

viscosity relative to thermosets which restricts their flow through reinforcements. Several studies have disclosed the incorporation of nanofillers/micro fillers over a range of thermoplastic matrices like isotactic polypropylene (iPP), polyphenylene sulphide (PPS), poly (ether ether ketone) (PEEK) and nylon-6. In the thermoset category, widely used polymers are epoxies, phenolics resins, polyesters, vinyl esters and urethanes.

Geometry of nanoscale fillers: The dimensions of dispersed nano fillers in polymer matrices is one of the prime factors that affects the final properties of composites [26,28,87–88]. First category is two dimensional (2D) layered nanoscale fillers such as graphene, MXene, layered silicates as thin sheets of thickness of few nanometers and length varying between hundreds to thousands of nanometers. The other class of nanofillers, has two dimensions in nano scale and third dimension is longer, creating one-dimensional (1D) structure like carbon nanotubes, halloy site nanotubes, carbon nanofibers etc. The third class of nano scale fillers have all dimensions in nanometers which are *iso*-dimensional (aspect ratio very low) nanofillers like quantam dots, spherical silica, tungsten carbide and semiconductor nanoclusters which are classified under three dimensional (3D) nanofillers.

Interfacial area: Uniformly dispersed isotropic and anisotropic fillers of nano dimensions can introduce large interfacial area per unit volume between nanofillers and the matrix. Morphology developed in dual nanoscale composites could be controlled based on nanoscale features. The nano dimensions of reinforcements and large interfacial area could produce exciting physical and mechanical properties with regard to conventional composites. [89–90].

Surface modification of nanofillers: Surface modification of nanofillers is performed with functional moieties particularly to prevent nanofiller aggregation and promotes uniform dispersion of fillers in the matrix [91–92]. Enhanced anchoring of the fillers as well as between the fillers and the matrix chains could be achieved owing to the surface modification and functionalization with coupling agents/compatibilizers [93–95].

Nanofiller loading: This parameter takes another important role in deciding the overall properties of the dual nanoscale and multiscale composites. It was reported in several studies [96] that higher content of nanofiller could lead to particle aggregation since very small nanosize particles could bind themselves owing to van der Waals forces of attraction. Enhanced properties were reported for the composites at low level loading of nanofillers with better dispersion. Surface free energy of fillers and compatibility between dual nano fillers are to be considered while selecting the of nanofillers in dual and multiscale composites. Optimized filler content has to be maintained to achieve effective interfacial strength, adhesion characteristics and nanofiller bridging amongst fillers and with matrix.

Fiber geometry, orientation and content: In the case of multiscale composites, fiber length contributes to the overall performance of the composites. Short fiber reinforced micro composites are cost efficient, exhibit versatility and display flexibility in processing. One of the drawbacks associated with short fiber reinforced composites is that during melt processing in twin screw extrusion and injection molding, further attrition of fibers occurs owing to the high shear rates involved. Whereas in the case of fiber (long) reinforced composites processing has to be performed through special extrusion/ pultrusion techniques. The fibers are introduced only at the die end of the extruder and the products exhibit magnificent tensile, impact properties and durability. Generally, with increase in micro filler content the properties of composites are enhanced but the amount of micro scale fillers to be incorporated in the matrix should be high to achieve reasonable mechanical properties. This may lead to production of heavy weight components which can be avoided by reducing the amount of microscale filler and incorporating nanofiller. Fiber orientations such as unidirectional, bidirectional, random and multidirectional orientations also play a significant role on

overall performance of composites. Shazed et al. [97] outlined the distribution of orientation of fibers on the fractured specimen of PP/CNT/CF samples and represented in Fig. 4. The distribution pattern observed in SEM images by using image processing software reveals the random orientation of carbon fiber in PP/CNT/CF multiscale composite. The fiber orientation efficiency factor could be determined considering the transverse direction to the tensile load which is perpendicular to the fracture surface.

The evaluation of nanofiller dispersion and distribution is very crucial, since mechanical and thermal properties are strongly influenced by the morphologies obtained. The key paramete which one would look into, to ensure the uniform dispersion and distribution of DNFs and MSFs in polymer matrices is that the dispersed phases should exhibit at least one dimension less than 100 nm. The nanofiller content should also be at optimum or low contents which could appreciably enhance the mechanical, thermal, flammability and barrier properties of the prepared composites without affecting their processability. The nanoparticles usually exist in the form of agglomerates and a large number of individual nanoparticles are bonded together in the form of microscopic agglomerates by attractive forces like electrostatic, ionic and van der Waals forces. Owing to the bonding effects between nanoparticles when they are added to the polymer melt, the viscosity of the polymer melt/solution increases. At higher content of nanofillers the viscosity of the melt will be increased to an extent affecting the processability. This is specifically important in the case of thermoset resin where fluidity is required to penetrate through the fibers, which are used as reinforcements. These agglomerates when mixed with the polymer matrix in solution processing or melt processing will be subjected to forces like shear, ultrasonic and centrifugal forces to disperse individual particles to the nanoscale in the polymer matrix. In melt processing techniques like twin screw extrusion which has wide industrial applications, specialized screw design configurations are available which provides remarkable mixing capability. Screw designs with varying pitch, length to diameter ratio, helix angle, rpm of the screw, different processing stages across the length of the screw, intermeshing twin screws could contribute to the effective mixing. These parameters influence the differential pressure across the length of the screw, shear and feed flow rates which could precisely control the degree of mixing.

3. Static mechanical properties of hybrid nano and multiscale filler reinforced polymer composites

3.1. Tensile properties evaluation

The salient mechanical properties of multiscale and dual nanofiller reinforced polymer composites from several studies are reported in Table 2. Shazed et al. [97] reported the enhancement in mechanical properties of carbon nanotube coated randomly oriented short carbon fiber incorporated PP hybrid composites. Carbon fibers treated at a reaction temperature of 700 ^{o}C for 30 min were incorporated along with CNTs in PP matrix and it could be observed that tensile modulus reached 3.52 GPa (enhancement by 104.6 %) in comparison with 1.72GPa of neat PP/carbon fiber composite. The tensile strength of PP/carbon fiber (CF)/Carbon nanotube (CNT) composite was reported as 33.63 MPa (enhancement by 64%) with regard to 20.5 MPa of neat CF/PP composite [97]. The significant enhancement in tensile properties with regard to micro composite is due to the synergism of carbon nanotubes coated on carbon fiber and CNTs acts as a binder at the interfaces of carbon fiber and the matrix. Similar work was performed by Sharma et al. [98] and outlined the mechanical strength of CNT grown carbon fiber reinforced epoxy composites. The methodology of growth of CNTs on carbon fiber in epoxy matrix is illustrated in Fig. 5. Epoxy/carbon fiber/CNT reinforced composites showed an enhancement of 69% in tensile strength with respect to neat epoxy/carbon fiber composite [98]. They explained that CNTs enhanced the interfacial bonding between the fillers and the matrix and the partial alignment of carbon fibers and nanotubes along fiber axis during the fabrication are the two prominent reasons for the enhancement of tensile properties. The direct growth of CNTs on fibers encouraged the mechanical anchoring and increased the fiber surface roughness and improved the overall interfacial area. Eventually, in epoxy/carbon fiber/CNT composites CNTs have a major contribution in strengthening the matrix-fiber interface. The authors reported that specimens without CNTs exhibited brittle fracture without plastic deformation allowing quick propagation of cracks. The methodology of growing CNTs on carbon fibers before incorporating into matrix has modified the surface morphology of fibers and strengthened the fiber–matrix interface.

Karsli et al. [20], evaluated the mechanical properties of PP/glass fiber/carbon nanotube multiscale hybrid composites and reported the augmentation in tensile properties for hybrid composite with regard to PP/CNT nanocomposite or PP/Glass fiber micro composite. In order to enhance the mechanical properties of PP composites, glass fibers were sized with silane coupling agents and nanotubes were treated with acid mixture to attach carboxyl functional moieties. A possible interaction was expected between silane treated glass fibers and carboxyl treated CNTs and this synergism can cause positive hybrid reinforcement that could enhance the gross mechanical properties of the composites. The higher polarity of glass fibers would have intensified the hybridization effect and improved the overall polarity of the matrix. The authors reported that apart from surface treatment of glass fibers and carbon nanotubes, adequate stress transfer at the interfaces of glass fibers and CNT embedded in PP matrix is required to enhance the comprehensive tensile and dynamic mechanical properties of composites. The hybrid PP/MWCNT/ glass fiber (GF) system, exhibited higher notched izod impact strength at 4 wt% CNTs and at 30 wt% glass fibers with regard to PP/GF micro composite or PP/CNT nanocomposite since the simultaneous inclusion of fillers reduced the agglomeration effects owing to the high polarity of glass fibers [20]. Iman et al. [86] also reported the elevation in tensile properties of PP/MWCNT/Glass fiber composites and found that Young's modulus of hybrid multiscale composite reinforced with carbon nanotubes (1 wt%) and short glass fiber (20 wt%) enhanced by 300% in comparison with neat PP [86–119]. The flexural modulus and strength reported for PP/MWCNT (1 wt%)/GF (20 wt%) hybrid multiscale composite was 15,876 MPa and 186.7 MPa with respect to 1549 MPa and 48 MPa for PP/GF composite. The incorporation of MWCNT in PP/GF has dramatically improved the strength and flexural modulus [86,120].

Fawad et al. [121] detailed the preparation of amino-modified double walled carbon nanotubes (DWCNTs) /carbon fiber-based epoxy multiscale hybrid composite laminates by resin infusion technique. They explained the interlaminar toughness-mode I (three-point bending) and impact properties of the nano and hybrid composites. The incorporation of minimal amounts of CNTs of about 0.025, 0.25 and 0.1 wt% in epoxy resin has enhanced the flexural strength by 5 % and flexural modulus by 35%. Further, a 6% enhancement in impact energy absorbed, and 23% decrement in interlaminar toughness was reported. The morphological studies on DWCNTs/CF/epoxy multiscale composites revealed the excellent dispersion of carbon fibers and CNTs and at certain locations non uniform distribution of CNTs amongst the layers of carbon fibers could be seen due to the bundling of CNTs. Some of the prominent failure mechanisms explored in morphological analysis portrayed pull out of fibers, fracturing, development of debonding zone between the fiber and the matrix, cracking matrix, bridging of cracks which was in agreement with earlier studies on similar composites [17,106,122–123]. Some previous attempts reported that inclusion of multiscale fillers decremented the flexural strength owing to the poor interfaces between the fillers and the matrix [124]. Impact test analysis reported that amongst the hybrid multiscale composites of epoxy/carbon fiber/amine modified DWCNT, the increment of CNTs from 0.05 to 0.1 wt% could enhance the impact strength from 3 % to 6 % which is not very significant. In hybrid epoxy/carbon fiber/0.05 wt% DWCNT-NH_2 multiscale composites owing to their larger volume, carbon fibers were supposed to

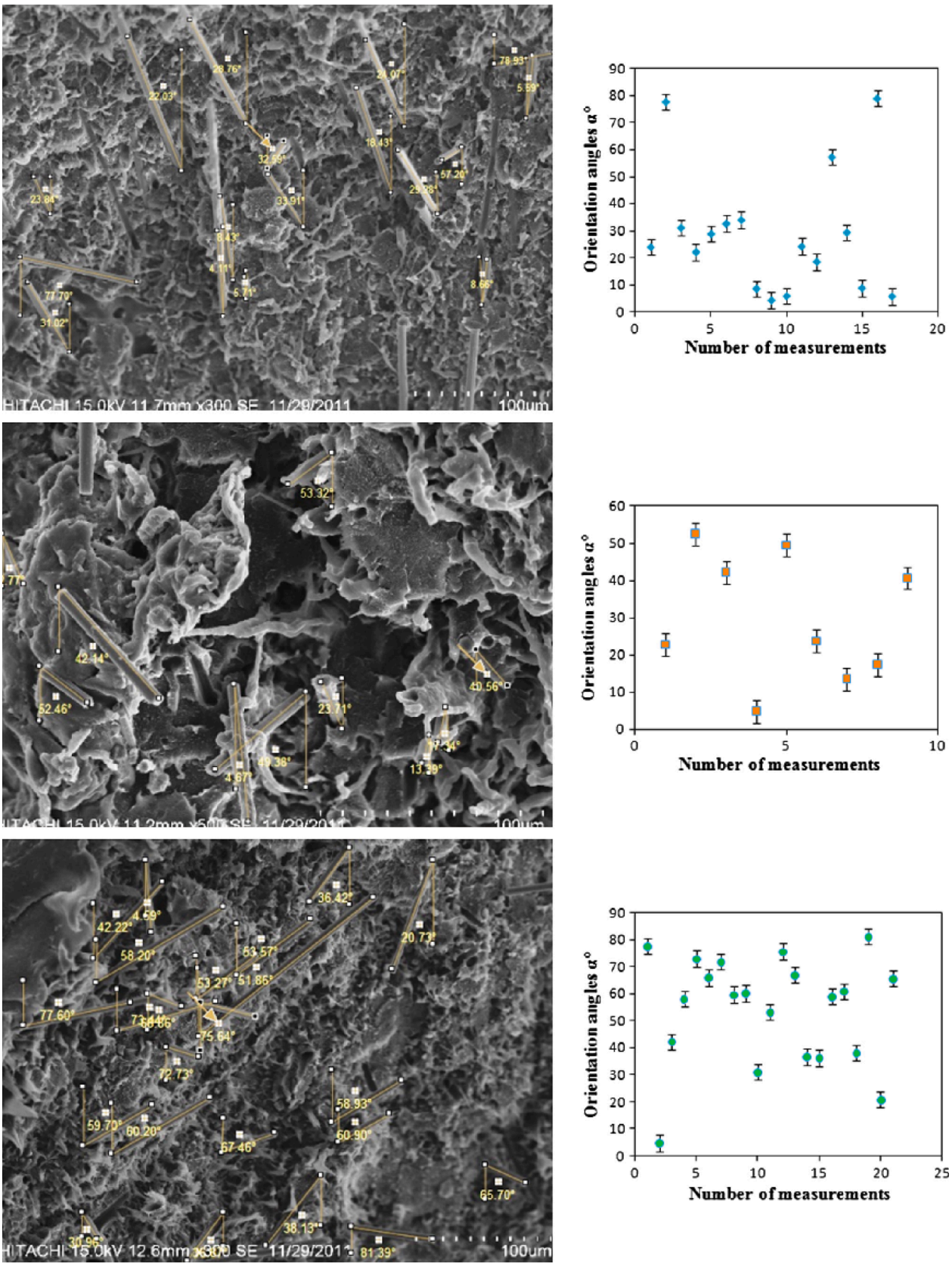

Fig. 4. Scanning electron microscopic images of fractured specimens of PP/CNT/CF composites showing fiber orientation geometry upon tensile test loading with distribution of orientation angles [97]. (Reprinted with permission from Elsevier).

Table 2
Mechanical strength of multiscale and dual nanoscale filler reinforced polymer composites.

Sl no	Matrix	Nanofiller	Micro filler	Tensile strength (Exp)	Young's Modulus (Exp)	Impact strength	Flexural strength	Ref
Multiscale filler reinforced thermoplastic polymer composites								
1	PP	CNT	Carbon fiber	33.63 MPa	3.52GPa			[99]
2	PP	CNT(4 wt%)	Glass fiber (30 wt%)	38 MPa (7.8 kJ/m^2 (Notched Izod))	6.2GPa	————	————	[20]
3	PEEK	CNT 0.5 wt%	Carbon fiber 70 wt%	———	———	————	Strength = 1580 MPa Modulus= 80GPa	[100]
4	HDPE 70 wt%	SiC	wheat straw fiber 30 wt%	23 MPa	———	————	33 MPa 2300 MPa	[101]
5	Polyester 60 wt%	Nano clay	Vakka fiber 40 wt%	95 MPa	2.6GPa	————	Strength = 145 MPa Modulus = 4GPa	[102]
6	LPET 40 wt%	CNT 0.9 wt%	Glass fiber 60 wt%	343 MPa	15.1GPa	————	Strength = 402 MPa Modulus = 14.1GPa	[103]
7	Polyamide 6	CNT 1 wt%	Carbon fiber	1100 MPa	58GPa	———	———	[104]
8	PEEK	Graphene 1 wt%	Carbon fiber 30 wt%	150 MPa	7000 MPa	———	———	[105]
9	Polypropylene	CNT 5 wt%	Glass fiber 20 wt%	51.4 MPa	2500 MPa	———	———	[106]
10	PPEK	Carbon fiber 60.3 wt%	CNT 1 wt%	———— (Impact strength: 1.41 J)	———	1.41 J	———	[107]
11	Polypropylene 86 wt%	short glass fiber 10 wt%	Nanosilica 4 wt%	44 MPa	2.6GPa	————	———	[45]
Multiscale filler reinforced thermoset polymer composites								
1	Epoxy	CNT	Carbon fiber	450 ± 5 MPa	———	————	————	[98]
2	epoxy	Nano polyvinyl alcohol 0.1 wt%	Carbon fiber 50 wt%	700 MPa	45GPa	————	————	[108]
3	epoxy	Aramid nanofibers 0.5 wt%	Kevlar 80 wt%	570 MPa	25GPa	————	————	[109]
4	Epoxy	Alumina 1 wt%	Carbon fiber	13.95 MPa	1.10GPa	————	Strength = 686 MPa Modulus = 1.74GPa	[110]
5	Epoxy	Nano silica 3 wt%	Carbon fiber kevlar	525 MPa	———	————	Strength 650 MPa	[111]
6	Epoxy	MWCNT 0.3 wt%	Carbon fiber 52 wt%	692 MPa	54.1GPa	————	————	[112]
7	Epoxy	$CaCo_3$ 2 wt%	Carbon fiber	750 MPa	40.38GPa	————	strength381MPa modulus= 40GPa	[113]
8	Epoxy	MWCNT 1 wt%	Carbon fiber 60 wt%	447 MPa	15.9GPa	————	————	[114]
9	Epoxy	$AgNO_3$ 5.21 wt%	Carbon fiber	5810 MPa		————	————	[115]
Dual nanoscale filler reinforced polymer								
10	Nylon6,6	Activated graphene oxide 1 wt%	Mwcnt 1 wt%	40 MPa	0.7GPa	———	———	[116]
11	epoxy	Boron carbide 2.5 wt%	Tungsten disulphide 4 wt%	33 MPa	———	————	Strength 55 MPa	[117]
12	PAEK	Boron carbide .375 wt%	MWCNT .375 wt%	104 MPa	3.2GPa	————	————	[83]
13	PAEK	Tungsten carbide 0.75 wt%	MWCNT 0.75 wt%	108 MPa	3.6GPa	————	————	[118]

absorb more energy during an external impact. According to Mode 1 fracture toughness analysis, carbon fiber reinforced matrix in the absence of CNTs exhibited a stick and slip fracture followed by propagation of crack, and crack cease behavior explains instability in the development of cracks. Average strain energy release rates (GI) of hybrid multiscale epoxy composite reported was 0.31 kJ/m^2 and there was a decrease of approximately 23% with the inclusion of CNTs owing to the hackle formation as the predominant failure mode [121]. Zhang et al. [125] reported the KI and G_c values of epoxy/short carbon fiber/nanofiber reinforced composites at 10 vol% carbon fiber and 0.5 vol% carbon nanofiber. The composite possesses higher fracture toughness and strain energy release rates with respect to individual filler composites. In the presence of multiscale fillers KI value raised by 427% and GI value by 232% at 10 vol% of CF and 0.75 vol% of carbon nano fiber. The crack pinning effect in the presence of nanofiller, debonding, pullout and breakage of nanofibers were elucidated as major failure mechanisms [125].

Praveen et al. [126] reported the synergism of silicate nano clay (layered) and carbon black in styrene butadiene rubber (SBR). It was delineated that the inclusion of 10 phr nano clay and 20 phr carbon black resulted in 153% enhancement in tensile strength, 144% stress increment at 100% strain and 157% increase in strain at break owing to the synergism of dual nanostructures contributing to the stiffening effect and effective stress transfer [126]. Rahmanian et al. [127] reported the

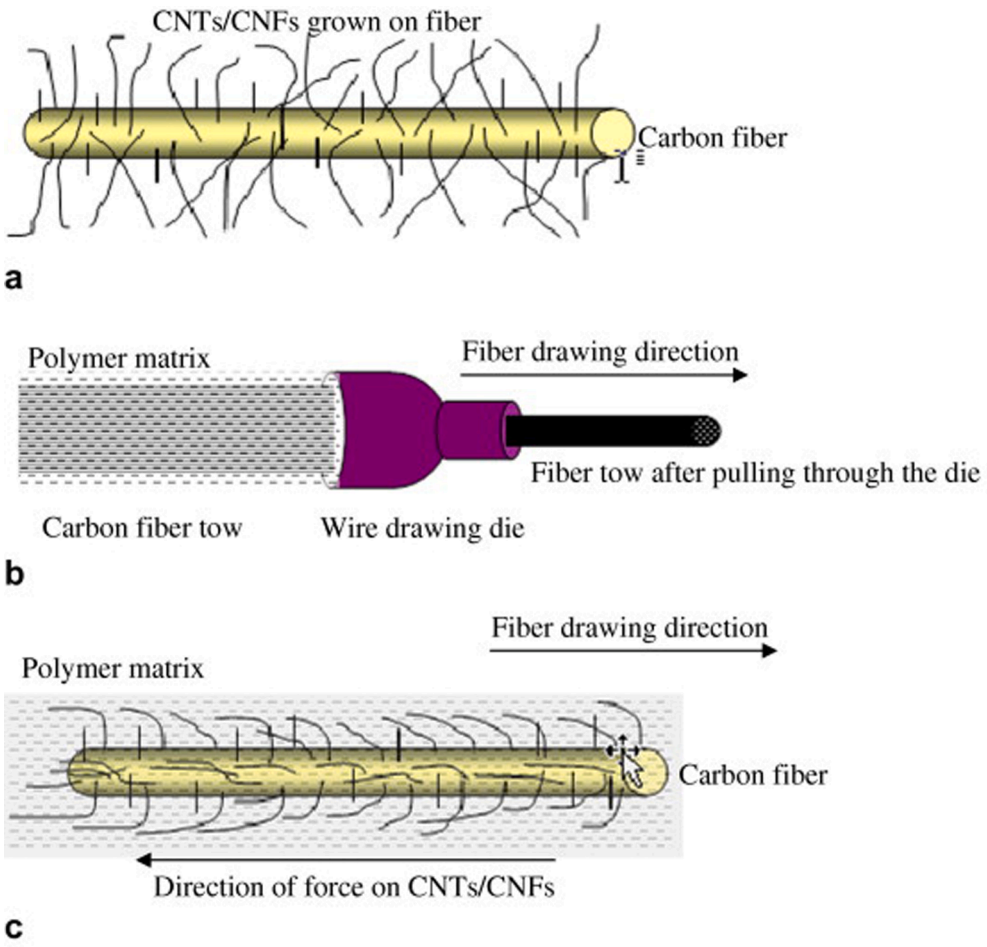

Fig. 5. (a) Representation of growth of carbon nanofibers/ carbon nanotubes on the surface of carbon fibers (b) Drawing of carbon fiber immersed in polymer pulled out through die to form a single fiber tow sample (c) Representation of partial alignment of carbon nanofibers /carbon nanotubes along the carbon fiber axis owing to drawing through die [98]. (Reprinted with permission from Elsevier).

effective stress transfer in multiscale composite, achieved by growing CNTs on carbon fibers. The synergic reinforcement of fillers caused an appreciable increment of 39% in elastic modulus and 37% in strength at optimum content of 0.3% CNT/1% carbon fiber in epoxy matrix [127]. Enhanced thermo- mechanical stability of composites was also reported by the incorporation of CNT-graphene nano hybrids in polymer matrices [128]. Zhang et al. [125] reported that epoxy reinforced with nanoscale carbon nanofiber and microscale short carbon fiber improved modulus, strength and fracture toughness with regard to micro or nano fillers reinforced composites. It was reported that inclusion of carbon nanofiber (0.75 wt%) along with short carbon fiber (10 wt%) in the epoxy matrix enhanced the ultimate tensile strength (80 to 91.4 MPa) and Young's modulus from 3.7 GPa to 5 GPa. This explains that at nanofiller content beyond 0.25 wt% in multiscale composite the increment in modulus and strength was appreciably higher than the superposition of individual contribution of separate fillers. It also proves the synergistic play of dual fillers on strengthening and stiffening of the epoxy matrix. The stress fields developed in the epoxy matrix between short carbon fibers were homogenized due to the presence of well distributed carbon nanofibers located in the space between carbon fibers, fiber enabling efficient transfer and redistribution of stresses. The strain softening effect was also reported for hybrid composites, assigned to the release of overstress on short carbon fibers responsible for the generation of subcritical cracks causing carbon nanofiber related failure events [125]. Another group of researchers investigated the development of multiscale reinforcements in composites via growing CNTs on microscale alumina nanoparticles for enhanced mechanical and electrical properties [65,129]. Mittal et al. [130] investigated the tensile properties of vinyl ester polymer composites reinforced with a multiscale filler system of MMT/Glass fiber. Surface treated MMT reinforced glass fiber/vinyl ester composites exhibited higher tensile strength (~290 MPa) elastic modulus (47.5GPa), flexural strength (282 MPa) and flexural modulus (9.2GPa) with regard to untreated MMT/Glass fiber multiscale composite. This is due to the enhanced interfacial interaction and dispersion achieved vide surface treatment of nanoclay with 3-APTES (3-Aminopropyl triethoxy silane) incorporated in vinyl ester/glass fiber matrix. The interplanar length between the nano platelets could be improved and each platelet encouraged compatibility with neighboring fillers and matrix when MMT was treated with nano silica [130]. In another work [131] involving epoxy /alumina/CNT hybrid nanofiller composite system, appreciable load transfer and interfacial strength between the filler and the matrix was observed. They reported that epoxy resins with varying nanofiller hybrid content (1, 3 and 5 wt%) improved the flexural modulus by 9.9%, 22.5% and 35.27% respectively with regard to neat epoxy. High modulus, strength and aspect ratio of CNT, and its content may block the epoxy chain motions and raise the cross-link ratio which could contribute to higher flexural modulus [131]. Investigations were carried out by other researchers in epoxy system reinforced with dual filler hybrids [132].

3.2. *Micromechanical and simulation modeling of tensile properties*

To enhance the reliability of experimental observations, the validation and theoretical prediction of tensile properties of composites are of

paramount importance [37,133–136]. Various analytical models have been used by researchers to predict the overall performance of composite materials by taking into consideration the individual properties of nano filler, microfiber or dual nano fillers and the matrix. The mathematical model named Halpin-Tsai model is used to predict the transverse and longitudinal modulus of the composite based on geometry, orientation of the filler, volume fraction of fillers, elastic properties of the fillers and the matrix. To evaluate the modulus of randomly oriented short fiber reinforced polymer composite, Halpin and Tsai proposed a series of equations.

$$E_L = E_M\left(\frac{1+c\eta_L v_f}{1-\eta_L v_f}\right) \qquad where \quad \eta_L = \frac{\left(\frac{E_f}{E_m}\right)-1}{\left(\frac{E_f}{E_m}\right)+c} \tag{1}$$

(1)

$$E_T = E_M\left(\frac{1+\alpha\eta_T v_f}{1-\eta_T v_f}\right) \qquad where \quad \eta_T = \frac{\left(\frac{E_f}{E_m}\right)-1}{\left(\frac{E_f}{E_m}\right)+\alpha} \tag{2}$$

(2)

In the Eqs. (1) and (2) E_L and E_T represents longitudinal and transverse modulus,E_m denotes Young's modulus of the matrix, E_f indicates Young's modulus of fiber, η_L and η_T denotes longitudinal and transverse efficiency factor, c is shape factor depending on fiber aspect ratio (l/d), v_f is volume fraction and ∝ is a geometric factor.

In the case of multiscale composites, primarily the Halpin-Tsai model was used to evaluate the tensile modulus of PP/CNT composite [134–135]. The obtained modulus is then utilized as the matrix modulus to evaluate the modulus of hybrid composite (PP/CNT/CF composite) by taking into consideration the fiber geometry. A modified Halpin-Tsai equation can be given as in Eq. (3) incorporating l/d ratio of CNTs and effective modulus (Eeq) of carbon nanotubes.

$$E_C = \frac{3}{8}\left[\frac{1+2\left(\frac{l_{CNT}}{d_{CNT}}\right)\left(\frac{\left(\frac{E_{eq}}{E_M}\right)-1}{\left(\frac{E_{eq}}{E_M}\right)+2\left(\frac{l_{CNT}}{d_{CNT}}\right)}\right)*V_f}{1-\left(\frac{\left(\frac{E_{eq}}{E_M}\right)-1}{\left(\frac{E_{eq}}{E_M}\right)+2\left(\frac{l_{CNT}}{d_{CNT}}\right)}\right)*V_f}*E_M\right] + \frac{5}{8}\left[\frac{1+2\left[\frac{\left(\frac{E_{eq}}{E_M}\right)-1}{\left(\frac{E_{eq}}{E_M}\right)+2}\right]*V_f}{1-\left(\frac{\left(\frac{E_{eq}}{E_M}\right)-1}{\left(\frac{E_{eq}}{E_M}\right)+2}\right)*V_f}*E_M\right] \tag{3}$$

(3)

Banarjee et al., [137] proposed another modification on the H-T model (Eqs. (4)–(9)) for the multiple reinforcements in the matrix which takes into account volume fraction of all fillers. Eqs. (1) and (2) can be modified as:

$$\frac{E}{E_m} = \frac{1+\varepsilon\left(\eta_1 V_{f1}+\eta_2 V_{f2}\right)}{1-\left(\eta_1 V_{f1}+\eta_2 V_{f2}\right)} \tag{4}$$

$$\eta_1 = \frac{\left(\frac{E_{f1}}{E_m}\right)-1}{\left(\frac{E_{f2}}{E_m}\right)+\varepsilon} \tag{5}$$

$$\text{and } \eta_2 = \frac{\left(\frac{E_{f1}}{E_m}\right)-1}{\left(\frac{E_{f2}}{E_m}\right)+\varepsilon} \tag{6}$$

E is the elastic transverse and longitudinal modulus,V_{f1} and V_{f2} (volume fractions) denotes the content of filler 1 and filler 2 respectively, η_1 and η_2 are the transverse/longitudinal efficiency factor for filler 1 and 2, $'\varepsilon'$ is the shape factor based on the dimensions of filler [137].

$$\frac{G}{G_m} = \frac{1+\varepsilon\left(\eta_1 V_{f1}+\eta_2 V_{f2}\right)}{1-\left(\eta_1 V_{f1}+\eta_2 V_{f2}\right)} \tag{7}$$

$$\eta_1 = \frac{\left(\frac{G_{f1}}{G_m}-1\right)}{\left(\frac{G_{f2}}{G_m}\right)+\varepsilon} \tag{8}$$

$$\text{and} \quad \eta_2 = \frac{\left(\frac{G_{f2}}{G_m}-1\right)}{\left(\frac{G_{f2}}{G_m}+\varepsilon\right)} \tag{9}$$

Where G_{f1}, G_{f2}, G_m represent the shear modulus of filler 1, filler 2 and the matrix, respectively. The overall properties of short fiber (random orientation) reinforced composites depend upon the shape of heterogeneity of phases incorporated in the matrix, interface developed among components and load transfer at fiber/matrix interface. Voigt-Reuss model [97,138] was used to evaluate the elastic modulus of the composite (E_c) with randomly oriented fiber distribution with two types of fillers with load applied in a specified direction as in Eq. (10).

$$E_C = \frac{3}{8}E_L + \frac{5}{8}E_T \tag{10}$$

The modified rule of mixtures (RoMs) could be employed to evaluate and analyze the tensile modulus of fiber reinforced polymer composites in which filler was oriented in random directions (Eq. (11)) [133].

$$E_c = E_m\left(1-v_f\right) + \eta_L\eta_0 E_f v_f \tag{11}$$

The orientation of fillers was also taken into account by the Krenchel equation,

$$\eta_0 = \frac{\sum_n a_{fn} Cos^4\alpha_n}{\sum_n a_{fn}} where \sum_n a_{fn} = 1 \tag{12}$$

The parameters η_L is correction factor for fiber length (~1), η_0 denotes Krenchel orientation efficiency factor, a_{fn} is the ratio between the cross sectional area presented by a group of fibers oriented at an angle to the applied load direction and the total area of all the fibers at a given cross section of the composite and n = 1, 2, 3,4……..n. In those cases when internal geometry of fillers becomes more complicated with the inclusion of heterogeneous filler phase, RoM model is used to evaluate fiber orientation distribution factor η_0. To determine the through thickness fiber orientation efficiency (η_0) the Krenchel equation was modified as in Eq. (13).

$$\eta_0 = \frac{N_{f1}Cos^3\alpha_1 + N_{f2}Cos^3\alpha_2 + N_{f3}Cos^3\alpha_3 + \ldots\ldots\ldots\ldots + N_{fn}Cos^3\alpha_n}{N_{f1}Sec\alpha_1 + N_{f2}Sec\alpha_2 + N_{f3}Sec\alpha_3 + \ldots\ldots\ldots\ldots + N_{fn}Sec\alpha_n} \tag{13}$$

The parameter N_{f1} represents the total fiber fraction,N_f oriented at angle α_1 in any field of view. Using a series of fields of view, the through thickness fiber orientation angles can be measured to evaluate the corresponding efficiency factor. Tensile properties evaluated for PP/CNT/CF multiscale composites [99] are shown in Fig. 6. Karsli et al., [20] also evaluated the elastic modulus of glass fiber reinforced CNT/ PP composites using the RoM model as mentioned in Eq. (11). The highest modulus values were reported for the composite with 30 wt% glass fiber and 4 wt% CNT in PP matrix.

Nagar et al. [139] reported the applicability of continuum based micromechanical models such as Mori-Tanaka (M−T) and Halpin- Tsai (H-T) on functionalized (amine) layered graphene/ functionalized (amine) multiwalled carbon nanotube dual nanofiller reinforced hybrid epoxy composites. Mechanical properties were estimated with regard to weight fraction and aspect ratio of fillers. In the M−T model three principle directions (orthogonal) are used to determine the longitudinal (E11) and the transverse modulus (E33) and are expressed, as per Eqs. (14) and (15).

$$\frac{E_{11}}{E_m} = \frac{1}{1+\varphi_f(A_1+2\gamma_m A_2)/A} \tag{14}$$

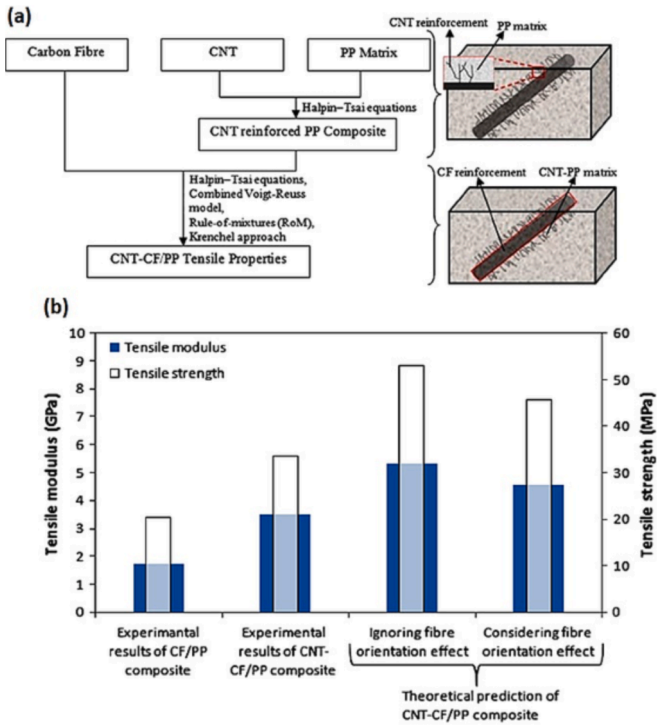

Fig. 6. (a) Heirarchy to evaluate the tensile modulus of PP/CNT/CF hybrid multiscale composite (b) Tensile properties evaluated for PP/CNT/CF hybrid multiscale composite with and without fiber orientation effects [99].(Reprinted with permission from Elsevier).

$$\frac{E_{33}}{E_m} = \frac{1}{1+\varphi_f(-2\gamma_m A_3 + (1-\gamma_m)A_4 + (1+\gamma_m)A_5 A)/2A} \tag{15}$$

Where φ_f is weight fraction of filler, γ_m denotes the Poisson's ratio of the matrix, and A, A1, A2, A3, A4 and A5 are evaluated from the matrix and filler properties and components of Eshelby tensor which is dependent on the elastic constant of the matrix and aspect ratio of fillers. In aspect ratio (l/d) 'l' and 'd' are the major and the minor diameters of an ellipsoidal disk-shaped inclusions. The authors [37,139] reported the comparison of M−T and H-T analytical models for graphene/epoxy nanocomposite and epoxy/graphene/CNT dual nano filler reinforced composites. A synergistic combination of dual nanofiller reinforced hybrid composites showed enhanced properties via M−T and H-T mathematical models owing to the higher filler content, and in the regimes where higher aspect ratio is maintained the tendency of fillers to embed in the matrix simultaneously increases and in turn the strengthening efficiency [37,139]. Pedrazolli et al. [18] proposed the applicability of Halpin-Tsai model, Mori-Tanaka model and Christensen Waals model in glassfiber/nanosilica and glass fiber/graphene nanoplatelet composites. Halpin-Tsai model considers the modulus of filler and the matrix, geometry of fillers, its aspect ratio, assumes uniform dispersion of fillers, orientation in particular directions and thorough bonding at the filler/matrix interface. One of the drawbacks associated with the model is that it does not account for the aggregation effects, properties at the interphase and considers unidirectional alignment of the filler along the direction of the applied tensile load. To overcome the drawbacks of the H-T model, Tsai and Pagano proposed models (Eq. (10)) that could be used to calculate the modulus of composites reinforced with random oriented short fibers in a plane. In continuation with these proposed models, Christensen and Waals suggested the evaluation of elastic modulus of 3-dimensional random distribution of short fibers throughout the composite volume as follows (Eq. (16)). This model can be used to evaluate the Young's modulus of randomly oriented filler reinforced composite systems in 3D plane in terms of Poisson's ratio, fibre volume fraction and its aspect ratio. The Young's modulus of both uni-directional and randomly oriented filler composites are strongly dependent on the reinforcements length and hence on Poisson's ratio.

$$E_c = \frac{\varphi_f}{6}E_f + \left[1+(1+\gamma_m)\varphi_f\right]E_m \tag{16}$$

E_f is elastic modulus of filler,γ_m is the poisson's ratio of filler,φ_f is filler volume fraction. Chow [140] suggested a simplified equation for the determination of effective modulus of unidirectionally aligned ellipsoidal fillers reinforced in polymers at finite concentrations (φ_f). The major to minor axis (aspect ratio of particle) characterizes the shape of the particle. In the case of an isotropic (transverse) composite with fillers oriented in the direction of injection molding, the effective elastic (compressive) properties of the composites can be determined according to Eq. (17).

$$\frac{E_c}{E_m} = 1 + \left(\frac{k_f}{k_m} - 1\right)G_1 + \frac{2\left(\frac{\mu_f}{\mu_m} - 1\right)K_1}{2K_1G_3 + G_1K_3} \tag{17}$$

where μ_f, μ_m, k_f, k_m represents the shear and bulk moduli and the subscript 'f' and 'm' denotes filler and matrix, K_i, G_i denotes Eshelby tensor components [140].

Pedrazzoli et al., [18] also suggested a two-population model method to combine the effect of microscale glass fiber and nanoscale graphene and nanosilica in the PP composites. Yoo et al. [141] proposed a similar method to model the merged effect of exfoliated clay and intercalated tactoids. When two fillers are added to the same matrix, they should be considered as separate populations to accurately predict their experimental modulus. Two population models can be of additive and multiplicative approaches [142]. The effect of individual filler on the tensile modulus can be evaluated without double counting the contribution of the matrix in the additive approach. The additive approach can be evaluated according to the following Eq. (18).

$$\frac{E_c^{add}}{E_{nm}} = \frac{E_{nm/f1}}{E_{nm}} + \frac{E_{nm/f2}}{E_{nm}} - 1 \tag{18}$$

Where 'E_{nm}' represents modulus of neat matrix, $E_{nm/f1}$ represents modulus of matrix reinforced with filler 1 and $E_{nm/f2}$ represents modulus of matrix reinforced with filler 2 and 'E_c^{add}' is the overall composite (polymer reinforced with filler 1 and filler 2) modulus respectively by additive approach. There are two approaches in the multiplicative two population modeling. The foremost step is to consider the contribution of filler 1. Then 'neat matrix/filler1' composite is taken as the matrix for the filler 2 addition. Further the effect of each filler can be multiplied.

$$\frac{E_c^{mult}}{E_{nm}} = \frac{E_{nm/f1}}{E_{nm}} * \frac{E_{\frac{nm}{f2}}}{E_{\frac{nm}{f1}}} \tag{19}$$

Where E_c^{mult} is the overall composite modulus containing dual fillers by multiplicative approach. Another approach is to consider the effect of filler 2 inclusions at first. The modulus of neat matrix/filler 2 composite is taken as the base matrix for filler 1 incorporation. Then the contribution of filler 1 is calculated using the effective modulus of neat matrix/filler 2 composite as the matrix. The overall composite modulus of dual filler reinforced composite using the second approach in the multiplicative dual population model can be given as in Eq. (20) [142].

$$\frac{E_c^{mult}}{E_{nm}} = \frac{E_{nm/f2}}{E_{nm}} * \frac{E_{nm/f1}}{E_{nm/f2}} \tag{20}$$

Pedrazzoli et al. [18] reported that additive and multiplicative approaches predict similar stiffening effects, in PP/glass fiber/graphene nanoplatelet composites. They inferred that slightly higher modulus values were obtained by multiplicative model since this model considers nanocomposite as the matrix for the micro fiber reinforcement.

Christensen and Lo [143] presented models to determine the effective bulk (K) and shear modulus (G) of polymer composites reinforced with hard spherical particles as in Eqs. (21)–(29).

$$K = K_m + \frac{\varnothing(K_f - K_m)}{1 + (1-\varnothing)R_m(K_f - K_m) + X} \tag{21}$$

$$G = G_m + \frac{\varnothing(G_f - G_m)}{1 + (1-\varnothing)Q_m(G_f - G_m) + Y} \tag{22}$$

$$X = \frac{a}{1 + R_f(K_m - K_f)} \tag{13}$$

$$Y = \frac{a}{1 + Q_f(G_m - G_f)} \tag{24}$$

$$R_z = \frac{3}{3K_z + 4G_z} \tag{25}$$

$$Q_Z = \frac{6(K_z + 2G_z)}{5G_z(3K_z + 4G_z)} \tag{26}$$

Where Φ is volume fraction of filler and 'm' and 'f' corresponds to matrix and filler phases.

$$\varnothing = \varnothing_f + \varnothing_i = \varnothing_f(1 + a) \tag{27}$$

$\varnothing_i$ is volume fraction of interphases and 'a' is interphase parameter which denotes the extend of interfacial interaction or interphase properties. When a = 0, there is no adhesion at the interface or absence of interphase.

$$a = \frac{A_c \rho_f r_i (E_i - E_f)}{E_f} \tag{28}$$

The term $A_c \rho_f r_i$ explains the role of filler and interphase size on 'a' parameter whereas the term $\frac{E_i - E_f}{E_f}$ represents the effect of filler and interphase modulus. For nano composites with spherical inclusions, $A_c = \frac{3}{\rho_f r}$ and hence interface parameter 'a' can be evaluated as

$$a = 3\frac{\left(\frac{r_i}{r}\right)(E_i - E_f)}{E_f} \tag{29}$$

Ji model [26,143] (Eqs. (30)–(33)) proposed a three-phase model taking into account the matrix, filler and the interphases and considers spherical inclusions as reinforcements in the matrix and are as follows.

$$E = E_m \left[(1-\lambda) + \left(\frac{\lambda - \beta}{(1-\lambda) + \frac{(\lambda(k-1))}{lnk}} \right) + \frac{\beta}{(1-\lambda) + \frac{(\lambda-\beta)(k+1)}{2} + \beta\frac{E_f}{E_m}} \right]^{-1} \tag{30}$$

$$\lambda = \sqrt{\left(\left(\frac{r_i + r}{r}\right)^3 \varnothing_f\right)} = \sqrt{\left(\frac{r_i}{r} + 1\right)^3 \Phi_f} \tag{31}$$

$$\beta = \sqrt{\Phi_f}\,,\ k = \frac{E_i}{E_m} \tag{32}$$

In the absence of interphase Ji model reduces to two phase model.

$$E = E_m \left[(1-\beta) + \frac{\beta}{(1-\beta) + \beta\frac{E_f}{E_m}} \right]^{-1} \tag{33}$$

The parameters r_i and r_f indicates the radius of interphase region and radius of filler particle respectively. The values E_f, E_m, E_i denote the filler modulus, matrix modulus and interface modulus and k is a factor that defines the ratio of interphase modulus to matrix modulus. The 'k' value will be minimum when the interphase modulus is same as matrix modulus ie; when $E_i = E_m, k = 1$, while 'k' can take a maximum value when interphase modulus is equal to the filler modulus [26,143].

Christensen Lo and Ji models which takes into account the interface characteristics can also be used to predict the elastic modulus of dual nanocomposites and multiscale composites in which one filler inclusions are of spherical geometry. Another category of models are percolation models which accounts the interphase zone and is dependent on the reinforcement filler radius, particle curvature and non-bond interaction between polymer chain and fillers. The insufficient penetration of polymer chains into the inter particulate domain and the reduced volume fraction of interphase zone leads to the proposal of continuum percolation models that incorporates three phase microstructures consisting of nano reinforcements, interphase and the matrix [144].

As already discussed, the major assumptions in analytical model includes perfect adhesion between the phases, exclusion of the variations in the shape and size of the inclusions and the absence of microcracks and impurities. The overall mechanical properties of the composites are strongly dependent on the content and the size of the reinforcing particles. However, classical analytical models do not take into account the size of the inclusions, but the Eshelby's solution, applied in analytical models considers the inclusions in the form of spheres, ellipsoids, disks and cylinders of infinite length. Hence, the applicability of analytical models are limited to composites with such inclusions. Owing to the differences in the real and simulated microstructure, results obtained from micromechanical models mostly deviate from the experimental data. Based on the geometry, content, ratio of the properties of matrix and fillers, various analytical models turn out to be accurate and it is impossible to select the best model of homogenisation. Hence, in spite of their physical correctness and mathematical rigour, micromechanical models could not account for all the microstructural features of real composite systems and the model results can often deviate from experimental modulus values. Those models that take into account the microstructural characteristics of real composites explicitly or implicitly would be adequate for each composite system. Explanations on the choice of best suitable models for the prediction of tensile modulus are reported in supplementary section S2.

Finite element modeling has been integrated in commercial software packages like ABAQUS and ANSYS and is used to predict the mechanical behavior of filler reinforced polymer composites [145–147]. In finite element analysis (FEA) spatial discontinuities of non-homogeneous materials are taken into account by constructing preprocessed mesh. The continuum domain is discretized into sub-domains (finite elements) without overlaps and gaps. The sub-domains are interconnected at nodes [145]. The flowchart to develop the global stiffness matrix are shown in Fig. 7.

Raffie et al. [148] predicted the tensile characteristics of polymer reinforced with carbon nanotubes (radially grown) on carbon fiber on the basis of a bottom–up modeling approach. This stochastic modeling approach includes nano, micro, meso and macro scale levels of polymer composites (Fig. 8). For each scale of analysis, a representative volume element was defined and effective parameters of each scale were identified. The isolated CNT is at nanoscale and its mechanical properties are evaluated. In order to account for the non-bonded van der Waals interactions (ie; upper micro scale) the interaction between CNT and surrounding polymer is also explored. At meso scale, the tensile properties of polymer/CNT nanocomposite are derived considering radial arrangement of CNTs. Eventually, tensile characteristics of single fuzzy fiber which encompasses core carbon fiber surrounded with CNT embedded in base polymer are evaluated. As a final step, the tensile parameters of the unidirectional and short fuzzy fiber reinforced composites are computed at macro scale. Non straight shapes of CNTs and CNT volume fraction are taken as random parameters [148]. In each scale of simulation, the parameters considered for modeling are reported in Table 3. The multiscale inclusion of fillers in the simulation procedure of hybrid composite necessitates the importance to define each scale

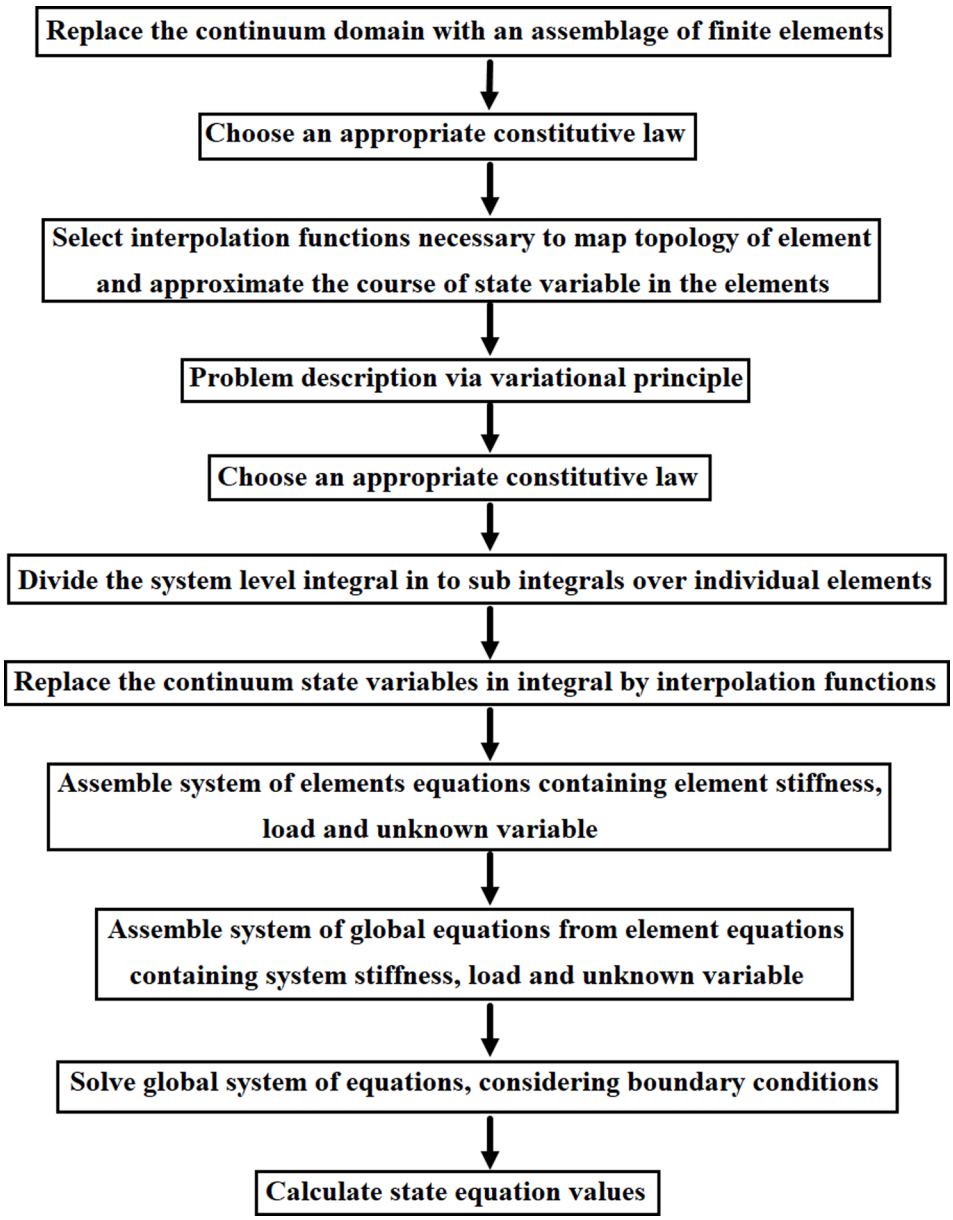

Fig. 7. Flowchart to develop global stiffness matrix and solve for system of equations [145].

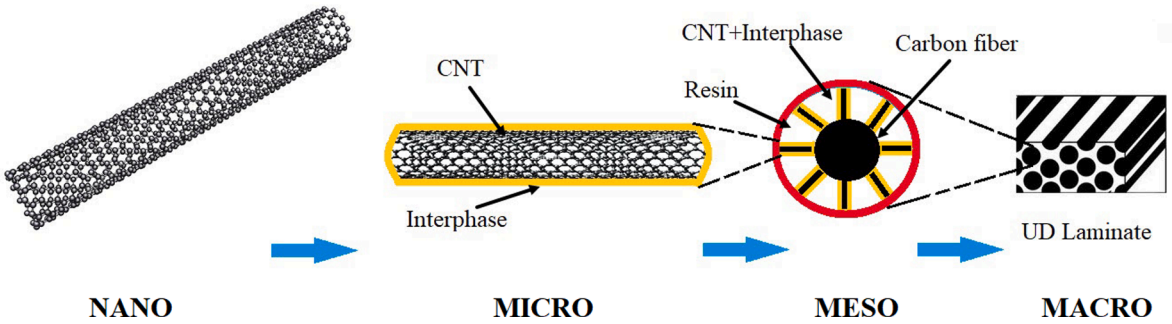

Fig. 8. Representation of each scale of simulations in stochastic modelling approach [148].

representative volume element (RVE) modeling separately.

Computational modeling details at each scale of analysis of carbon fiber/CNT reinforced polymer composite are reported in Table 4. Nanoscale continuum mechanics approach has been used in nano scale modeling where each C–C bond is replaced with a continuum element and the lattice structure of CNT can be simulated. A finite element model of carbon nanotube could be created following the structural mechanic's approach. The C–C bond of the nanostructure is replaced with an

Table 3
Simulation parameters involved in each scale of analysis of carbon nanotube grown on carbon fiber reinforced polymer composites via stochastic modelling [146].

Effective Scale	Parameters
Nano	C–C interactions, bond length, diameter of CNTs and CNT chirality
Micro	Interaction between CNT and polymer, CNT length
Meso	volume fraction of CNTs, radial orientation of CNT, and waviness of CNT
Macro	Volume fraction of carbon fibers

Table 4
Computational modeling details at each scale of analysis of CNT/Carbon fiber multiscale polymer composite [148].

Scale of analysis	Input data	Computational tool	Output results
Nano	CNT nano structure	FEM	Mechanical properties of isolated CNT
Micro	vdW interactions between CNT and polymer	FEM	Mechanical properties of developed equivalent fiber
Meso	CNT volume fraction, CNT curvature, mechanical properties of the core fiber and resin and equivalent fiber	Mori-Tanaka, coordinate transformation, random selection of CNT volume fraction, bounding technique for CNT curvature	Mechanical properties of isolated fuzzy fiber
Macro	Mechanical properties of resin and fuzzy fiber and fuzzy fiber volume fraction	Halpin-Tsai	Mechanical properties of U-D FFRP

equivalent beam element. The interatomic potential energies of molecular space to the strain energies of structural mechanics, are used to identify the mechanical and geometrical properties of the beam element [148]. Many researchers [149–152] have simulated the beam-based FE lattice structure of CNT as a discrete continuum structure. Young's modulus of CNTs isolated by FE models are reported in literature which is in the range of 1032 to 1046 TPa.

In microscale modeling, the CNT/polymer interface is modeled accounting for load transfer between CNT and matrix [153]. In the absence of surface treatment, CNT interacts with polymers through weak secondary non-bonded interactions. RVE defined at microscale consists of CNT, surrounding polymer and the interphase region between CNT and polymer containing non-bonded vanderwaal's interactions. In the simulation modeling approach, CNT is modeled at nanoscale and the interphase and polymer is modeled at microscale with isotropic behavior using continuum elements. The vdW interactions are represented by spring elements using semi continuum modeling [153]. Several researches have been reported on CNT embedded polymer modeling using semi-continum modeling [153–157]. In the next step, non-linear static analysis was carried out on the micro scale RVE FE model and the CNT and the interphase combination is converted into an equivalent fiber as an individual inclusion which behaves as a transversely isotropic material and the effective properties could be tabulated. In addition, CNT lengths are captured in micro scale. The RVE at mesoscale includes core micron fiber and the surrounding nanofillers with polymer. The nanofiller and the surrounding polymer around the core micro fiber can be considered as the secondary interphase which is micro interphase between core fiber and the polymer. At the meso scale, CNT and the interphase regime could be converted into equivalent fiber at very low scale of micro and this equivalent fiber can be considered as embedded reinforcements in polymer. Furthermore, suitable micromechanical models (like Halpin-Tsai, Mori-Tanaka etc.) formulations can be used to compute the stiffness tensor of nanofiller/polymer composite. After evaluating the effective properties of micro interphase and the core fiber, the effective tensile properties of fuzzy fiber can be modeled at the meso scale. Subsequently, the mechanical properties of hybrid composites can be evaluated using the appropriate micromechanics rule. Different scales of modeling can be stochastically employed considering the curliness of CNT structure and its content as the random parameters. Matlab software can be used to model the process and the output from each scale of modeling could be the input to the very next scale of modeling [148]. Flowchart of computational stochastic modeling steps are shown in Fig. 9.

In another work, simulation modeling of dual nanofiller reinforced polymer was discussed. Leon et al. [158] discussed the combined effect of secondary nano reinforcement ie; graphene nanoplatelets around the MWCNTs. The macro–micro model strategy was implemented and CNTs are modeled as cylinders of high aspect ratio. The interphase between the CNTs and the graphene platelets surrounding the CNTs embedded in the polymer was also modeled at micro scale [158].

Banerjee et al. [137] performed micro mechanical investigation of RVE using the finite element method of glass fiber/carbon fiber reinforced unidirectional hybrid composite. The fillers are considered as circular fibers and packed in hexagonal fashion, and the effect of volume fractions of two micro scale fillers within the unit cell is elucidated. A smooth linear variation was observed with volume fraction in stiffness properties predicted by FEM, and found to be in close proximity with analytical results (Table 5). The RVE of the hybrid composite is as shown in Fig. 10 and the meshed model is represented in Fig. 11. The RVE of the composite is studied using FEA ABAQUS software. The composite is considered to be under uniform strain distribution called micro strain and the corresponding macro stresses could be evaluated using FEA. The averaged stress to produce macro deformations are called macro stresses [137].

Macro stresses and macrostrain are related using the Eq. (34)

$$\sigma = C\varepsilon \tag{34}$$

Where C represents homogenized composite (stiffened matrix) elastic constant. Further, RVE is subjected to macrostrains and on the opposite faces of the element and the corresponding periodic boundary conditions are applied. RVE based constraints depend on symmetry, loading conditions, and periodicity of unit cells. Banerjee et al. [137] selected 10 random locations inside the RVE varying volume fraction of fillers in hybrid composite. One of the observations was, with fiber locations none of the elastic constants showed significant variability. It could be inferred that spatial variation of micro stresses does not affect elastic constants since they were evaluated using volume averaging of micro stresses of all elements [137,159]. The comparison of longitudinal tensile strength obtained for composites using FEA and analytical methods is presented in Table 5. The parameters v_c and v_g represent the volume fractions of carbon and glass fibers incorporated in the hybrid composite.

The steps involved in the random spatial distribution of fillers in RVE modelling was discussed under supplementary information in section S3.

4. Dynamic mechanical analysis of hybrid multiscale and dual nano filler reinforced composites

Dynamic mechanical analysis (DMA) is a significant tool for explicating the behavior at the filler/matrix interface of multiscale and dual nanofiller reinforced hybrid composites [76,160]. Static mechanical analysis results can be corroborated with temperature dependent dynamic mechanical tests and could predict the energy absorption capability of dual and multiscale composites. The morphological analysis and tensile properties can be correlated with dynamic mechanical properties and could indicate the adhesion factor and substantial entanglement

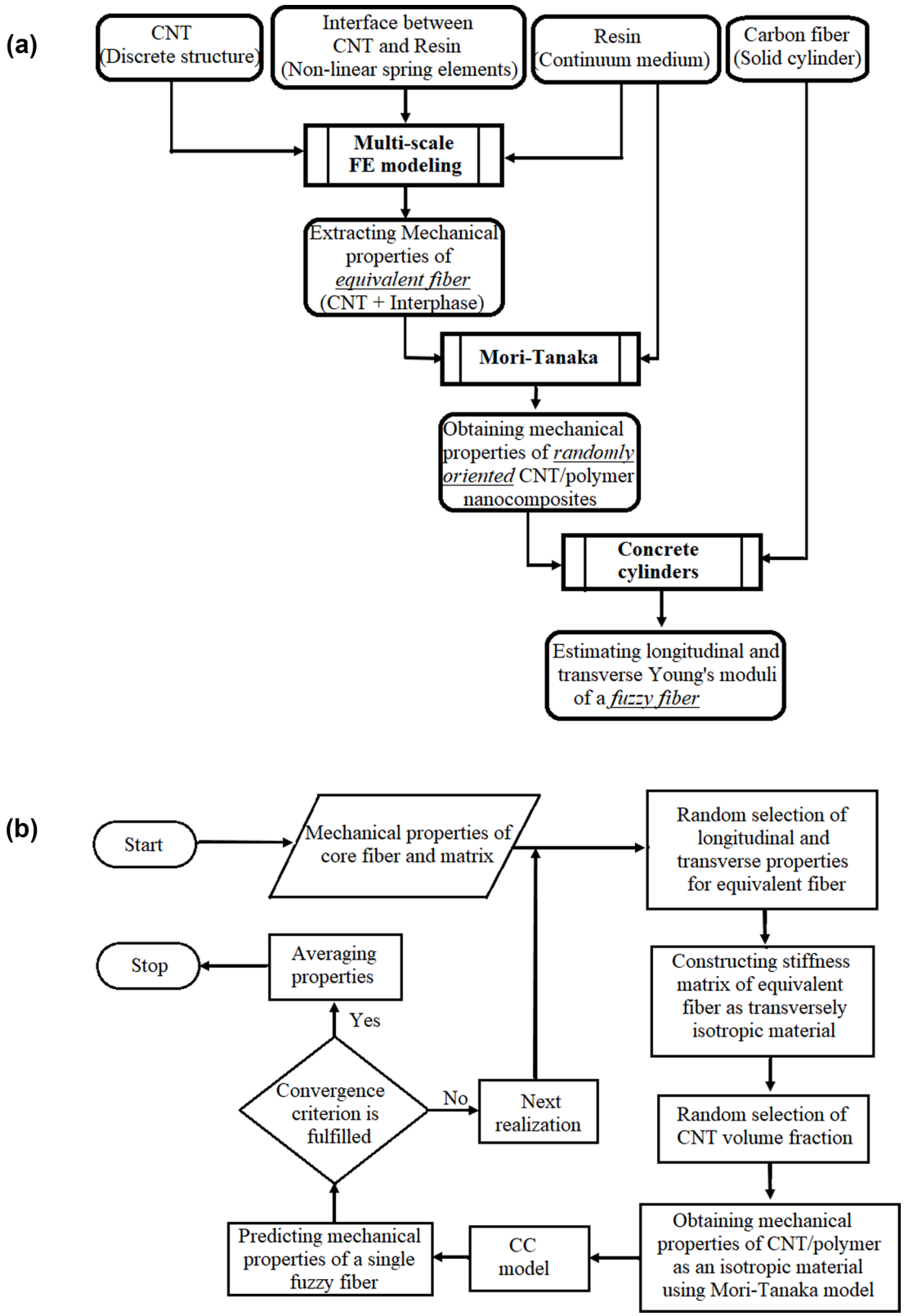

Fig. 9. A) flowchart representing the evaluation of mechanical properties of a fuzzy fiber b) flowchart for computational stochastic modeling of fuzzy carbon fiber reinforced mwcnt hybrid polymer composite[148].

offered by the fillers with polymer chains towards the overall performance of the composite. The mechanical tolerance of multiscale and dual nanoscale composites and inter- laminar, inter- fiber- nanofiller network versus polymer dynamics could be elucidated from dynamic mechanical studies.

The viscoelastic properties like storage, loss modulus and tanδ of polymer composites could be studied using DMA. Peng et al., [161] reported the storage modulus PP/MWCNT/glass fiber multiscale hybrid

Table 5
Longitudinal tensile strength for composites [137].

Composite	Specimen	FEA	Analytical	% Difference
Carbon/epoxy		2130	2229	4.43
Hybrid	H1(v_c: v_g: 0.54: 0.06)	1972	2069	4.67
	H2 (v_c: v_g: 0.42: 0.18)	1665	1748	4.77
	H3 (v_c: v_g: 0.3, 0.3)	1360	1428	4.78
	H4 (v_c: v $_g$: 0.18, 0.42)	1055	1108	4.79
	H5 (v_c: v_g: 0.06, 0.54)	750	788	4.81
Glass/epoxy		598	628	4.74

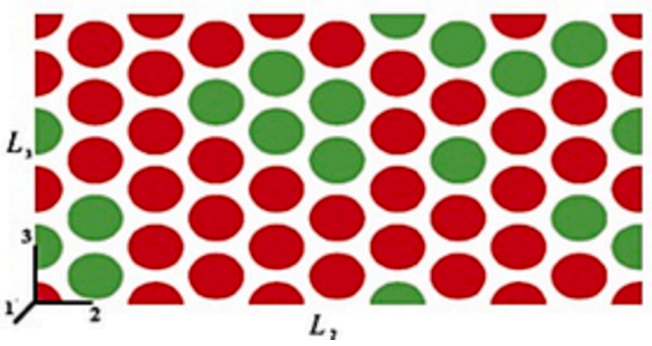

Fig. 10. RVE for hybrid composite containing two micro scale (carbon fiber, glass fiber) fillers [137]. (Reprinted with permission from Elsevier).

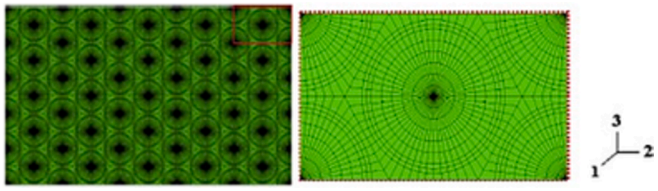

Fig. 11. RVE finite element model and the meshed model of the repetitive unit [137].(Reprinted with permission from Elsevier).

composites. In comparison with pure PP, the incorporation of glass fibers appreciably enhanced the storage modulus of PP in the whole temperature range owing to the reinforcing effect of glass fibers and effective stress transfer between filler and the matrix contributing to improvement in stiffness. A reasonable enhancement in Tg (beta transition) of the polymer phase (PP) in hybrid composite was observed in comparison with pure PP or that of PP/glass fiber composite which shows the synergism of dual scale fillers in PP matrix. In addition, a second transition peak is identified in the range of 60–90 °C which explains the alpha transition. The alpha transition in semi-crystalline PP explains the relaxation and sliding of tied polymer chains from crystalline blocks of PP. The broadness of peak intensity of PP hybrid composites explains the strong immobility caused on the polymer chains in the presence of multiscale fillers.

In our earlier studies [162] it was reported that the highest storage modulus of PP/MWCNT/GF hybrid composites was at the optimum MWCNT content (3 wt%) and the glass fiber (20 wt%) content. At higher loadings of MWCNT (5 wt%), storage modulus significantly dropped. The higher loss modulus of hybrid multiscale composite in the vicinity of the Tg of the polymer indicated superlative interfacial anchoring between the filler and the matrix demonstrating appreciable dissipation of energy and improvement in mechanical properties. The shift of alpha α peaks to higher temperature side showed hindered mobility of crystalline regions in the polymer and the drop in peak heights represents improved bonding at the interfaces of fillers and matrix. The lowered and broadened tan δ peak was significant for hybrid composite at its optimum content in comparison with PP/MWCNT nanocomposite at CNT content of 3 wt%. This proves that the presence of nano filler alone in the matrix could not restrict the mobility to a higher degree, whereas the synergistic inclusion along with microscale fillers drastically improved the reinforcing efficiency. Various parameters like efficacy of fillers, volume of constrained region, effectiveness of dispersion, degree of entanglement, adhesion factor etc. can be evaluated from DMA analysis that describes the synergism of multiscale fillers which improves the mechanical properties [1]. The variation in storage, loss modulus and tan delta peak with temperature of PP/MWCNT/Glass fiber hybrid multiscale composites are depicted in Fig. 12.

The effectiveness of filler dispersion can be computed from composite storage modulus according to the Eq. (35) [163–165],

$$E_f = \frac{\left(\frac{E_g^{'}}{E_r^{'}}\right)_{composite}}{\left(\frac{E_g^{'}}{E_r^{'}}\right)_{matrix}} \tag{35}$$

Where E_f denotes dispersion effectiveness, $E_g^{'}$ and $E_c^{'}$ denotes the storage modulus at the glassy and rubbery region of the composite and base polymer matrix. This factor is evaluated from the relative decrement in storage modulus at glassy and rubbery regions. The fragments of anchored chains of polymer between the filler and the matrix could not contribute to the dissipated/ lost energy which could be explained in terms of loss ratio of energy at T_g. The volume fraction of constrained regions (C) could be quantified as Eq. (36) [163–165].

$$C = 1 - (1 - C_0)\frac{F_C}{F_0} \tag{36}$$

Where F_C and F_0 represents energy loss ratio of composite and neat polymer respectively at T_g and C_0 is the percent crystallinity of pure polymer. The energy loss ratio 'F' can be calculated from the peak of tanδ as given in Eq. (37) [165–166]

$$F = \frac{\pi \tan\delta}{\pi \tan\delta + 1} \tag{37}$$

The degree of entanglement can be evaluated from the storage modulus at rubbery region (Eq. (38)), [163–166]

$$\varphi = \frac{E_r^{'}}{6*R*T} \tag{38}$$

Where 'R' and 'T' represent universal gas constant and temperature respectively.

The reinforcing efficiency (r) of fillers in the matrices could be related with storage modulus (E) and volume fraction (v_f) of fillers as per Eq. (39) [83,164–166]

$$E_c = E_m\left(1 + rv_f\right) \tag{39}$$

In our work [162] it was identified that the extent of entangled polymer chains between fillers was higher for hybrid composite at MWCNT content of 3 wt% and glass fiber at 20 wt% and was found to be $13.6*10^5$ mol/m^3 with regard to nano or micro composites. The volume of constrained regions (C) can be evaluated from the drop in peak of tanδ which was maximum for hybrid composite (C = 0.723). The reasonable reinforcing efficiency factor of hybrid composite is assigned to the synergism of intertwining of nanotubes with PP chains and forming a complex network of polymer chains around the surface of glass fibers which generates strong interfaces [160].

Sharma et al. [167] reported the synergistic reinforcing effect of dispersed CNTs and bucky paper (interleaved) on dynamic mechanical properties of Kevlar fiber reinforced multiscale epoxy composites. It was found that bucky paper interleaves along with dispersed MWCNT synergism caused substantial enhancement in interfacial and interlaminar properties. Bucky paper promotes efficient load transfer between the Kevlar fibers embedded in the matrix. Dispersed MWCNTs performs dual action, firstly it restricts the localized movement of segmental fragments

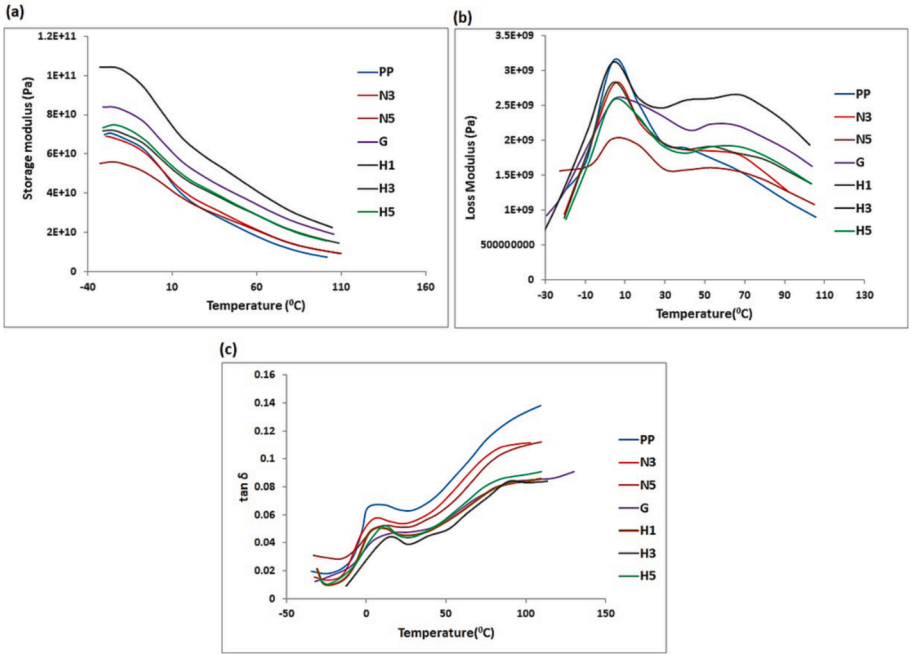

Fig. 12. (a), (b), (c) The variation of storage, loss modulus and damping parameter with temperature for PP/MWCNT/Glass fiber multiscale hybrid composites [162].

of polymer chains and secondly acts as an effective bridge between Kevlar yarns. It was noticed that with the synergistic inclusion of MWCNTs in the form of bucky paper interleaves and its uniform dispersion in the resin led to enhanced tensile strength, elastic modulus, toughness and strain at break. Even after the transition region (200-250 ^{o}C) of the polymer, storage modulus of Kevlar fiber reinforced/ bucky paper interleaved epoxy multiscale composites exhibited better thermo-mechanical stability with regard to Kevlar reinforced epoxy composite or MWCNT reinforced epoxy composite as revealed by DMA studies. Sharma et al. [167] confirmed the enhanced storage modulus further in comparison with degree of entanglement (ϕ) and effectiveness of filler dispersion (E_f). Degree of entanglement based on storage modulus at glassy region was reported for Kevlar/MWCNT/epoxy composites and effectiveness of filler dispersion which relates storage modulus from glassy to rubbery region was also evaluated. Maximum effectiveness could be observed when there is efficient load transfer between the fibers and dispersed CNTs network and lower E_f denotes better filler effectiveness [167].

Pravin et al. [126] reported the synergism of hybrid fillers (carbon black and organo modified nanoclay) in styrene butadiene rubber (SBR). Appreciable increment in storage modulus and steady decrease in tanδ maxima was noticed with increase in content of nano clay and carbon black/nanoclay incorporated SBR multiscale composites. Enhanced dynamic mechanical properties of dual filler systems was dedicated to the development of complex intercalated layered silicate structure of nanoclay and carbon black dual structures which could immobilize the fragments of polymer chains [126]. Rahmanian et al. [99] reported the inclusion of both carbon fiber and CNT reinforcements in epoxy matrix and the influence of fillers on storage modulus of composites. It was identified that storage modulus increased by 41% at CNT content of 0.3 wt% and carbon fiber content of 1 wt%. From the loss factor curves T_g of neat epoxy was found to be 70 ^{o}C and only slight differences in peak height of tanδ occurred which justifies that CNT and carbon fiber or its combination could not influence the movement of polymer chains at T_g. Beom et al. [168] explored the effect of plasma treatment of fillers on the mechanical properties of Polycarbonate (PC)/CNT/CF (carbon fiber) multiscale hybrid polymer composites. For plasma treated samples the composite storage modulus enhanced by 39 GPa (increment of 387 %) as compared to PC/CF composite. The authors inferred that although plasma treatment was effective for micro composite (PC/CF) rather than hybrid composites, hybridization effect of CNTs along with carbon fiber enhanced the surface roughness, functionality and bridged the gap between modulus of carbon fiber and polycarbonate [168].

Viscoelastic properties of CNT /carbon fiber strengthened epoxy hybrid composites under hygrothermal conditions recorded higher storage and tanδ values in comparison with conventional carbon fiber reinforced composites [169]. The dynamic mechanical behavior and viscoelastic characteristics of graphene oxide–carbon nanotube reinforced acrylonitrile butadiene styrene (ABS) composites was investigated recently [170]. Graphene oxide reinforced/carbon nanotube hybrid composites exhibited higher dispersion effectiveness as compared to individual filler based reduced graphene oxide and MWCNT based ABS composites. The authors commented on the dual filler network achieved within the composite that improves the interfacial interaction that could enhance the viscoelastic properties [170]. The viscoelastic properties of epoxy glass fabric composites reinforced with silicon carbide and alumina nano hybrid multiscale composites was discussed by Bommegowda et al. [171]. Storage modulus of composites was appreciably increased (lie in the range of 8,000 to 12,500 MPa) and silicon carbide filled epoxy presented highest storage modulus and reasonable shift in glass transition temperature [171]. Nayak et al. [172] reported the influence of dual nanofillers (Al_2O_3 and TiO_2) along with glass fiber in polymer composites and its synergism that leads to enhancement in interlaminar shear strength. However, they noted a decrease in storage modulus and loss modulus with the addition of nano fillers at the optimum content of 0.3 wt% Al_2O_3, 0.15 wt% TiO_2 [172].

Dynamic mechanical torsion studies of polyetheretherketone (PEEK) reinforced with 20 wt% carbon fiber and 2.5 wt% carbon nanofiber was reported by Tewatia et al. [173]. Storage modulus enhanced with multiscale hybrids before and after the glass transition temperature. In the case of PEEK composite tanδ peak was observed at 154 ^{o}C whereas in hybrid PEEK/2.5CNF/20CF maxima of tanδ peak was detected at 146 ^{o}C. The slight decrement in T_g indicates the movement of molecular chains at lower temperatures in hybrid composite owing to the inhibition of bulk crystallization in the presence of higher content of carbon fiber and carbon nanofiber [173]. Asimpour et al. [174] explored the

DMA parameters of MWCNT/graphene nanoplatelets/carbon fiber reinforced epoxy composites. They observed that the addition of 0.5 wt % of MWCNTs in carbon fiber reinforced epoxy composites exhibited higher storage modulus in the range of 20 and 60 ℃ in glassy state with regard to other multiscale composites. The MWCNT incorporated carbon fiber reinforced epoxy composites which were thus capable of storing more energy than similar GNP reinforced carbon fiber reinforced epoxy composites. DMA parameters are strongly influenced by the properties of interface and density of crosslinks. The bonding, crosslinks between filler and the matrix, structure of nanofillers and nature of polymer have prime roles to influence viscoelastic behavior. The T_g of epoxy/carbon fiber/0.5 wt% CNT increased to 84 ^oC with regard to 80 ^oC of neat PEEK [174].

Manu et al.,[83] reported the variation in storage modulus with temperature of PAEK polymer reinforced with dual nanofillers like boron carbide and carbon nanotubes. They reported that boron carbide/CNT system exhibited enhanced storage modulus in comparison with neat PAEK. The composite system at 0.75 wt% of treated boron carbide and functionalized MWCNTs presented the highest storage modulus value of 1350 MPa. They commented that silane treatment of boron carbide was less influential in enhancement of storage modulus of composites particularly at high temperatures of around 150 ℃ and the effect of acid treatment on CNTs is more influential. The higher loss modulus values at 0.75 wt% boron carbide/functionalized carbon nanotubes in PAEK indicates improved viscous response by dissipating maximum energy as heat which is an indication of restriction on the segmental mobility of polymer chains [83]. Fig. 13 reports the variation in storage, loss modulus and damping factor of PAEK/Boron carbide/MWCNT nanocomposites.

From our own experimental studies [26,83,114,162] and the studies reported in the literature [22–23] it could be interpreted that the storage and the loss modulus values enhanced substantially with the incorporation of dual nanoscale fillers and with multiscale fillers. The synergism of either dual nanofiller system or hybrid multiscale filler system constraints the mobility of polymer chains and shifts the Tg peak to higher temperature side which clearly depicts the superior interfacial interaction between the filler and the matrix. With the inclusion of rigid fillers with different geometry along with fibres, the entanglement density of polymer chains increase. The entanglement density and the constrained regions generated in the polymer composites could be evaluated from the DMA plots using various theoretical models discussed under section 4. Higher the volume fractions of constrained regions, better is the effectiveness of reinforcement and entanglement density of polymer chains which in turn will be reflected in the microstructure of the prepared composites. The stiffness and the damping characteristics of the composites could be analysed using DMA and could be corelated with morphology. The TEM images could vividly depict the uniform distribution and dispersion of fillers in polymer matrices and the strong embedment of fillers in matrices without reducing the aspect ratio of fillers. When the dispersion of nanofillers in uniform, the resistance offered to the movement of polymer chains enhances the storage modulus. The strong interface between the matrix and filler aids the dissipation of energy when the material turns viscous. This is due to the friction between polymer chains which raises the loss modulus values. The numerical values of DMA entanglement density, constrained regions, effectiveness of reinforcement etc. are reported in different studies of dual nanofiller and multiscale filler reinforced composites [22–23,26,162–163].

Various theoretical models can be used to predict the storage modulus of dual and multiscale filler reinforced composites. Einstein [160,175–177] proposed a simple model which takes into account the volume fraction of fillers in the matrix (Eq. (40)).

$$G_C = G_m(1 + v_f) \tag{40}$$

G_c and G_m represent composite storage modulus and the matrix modulus and v_f represents volume fraction of filler embedded in the

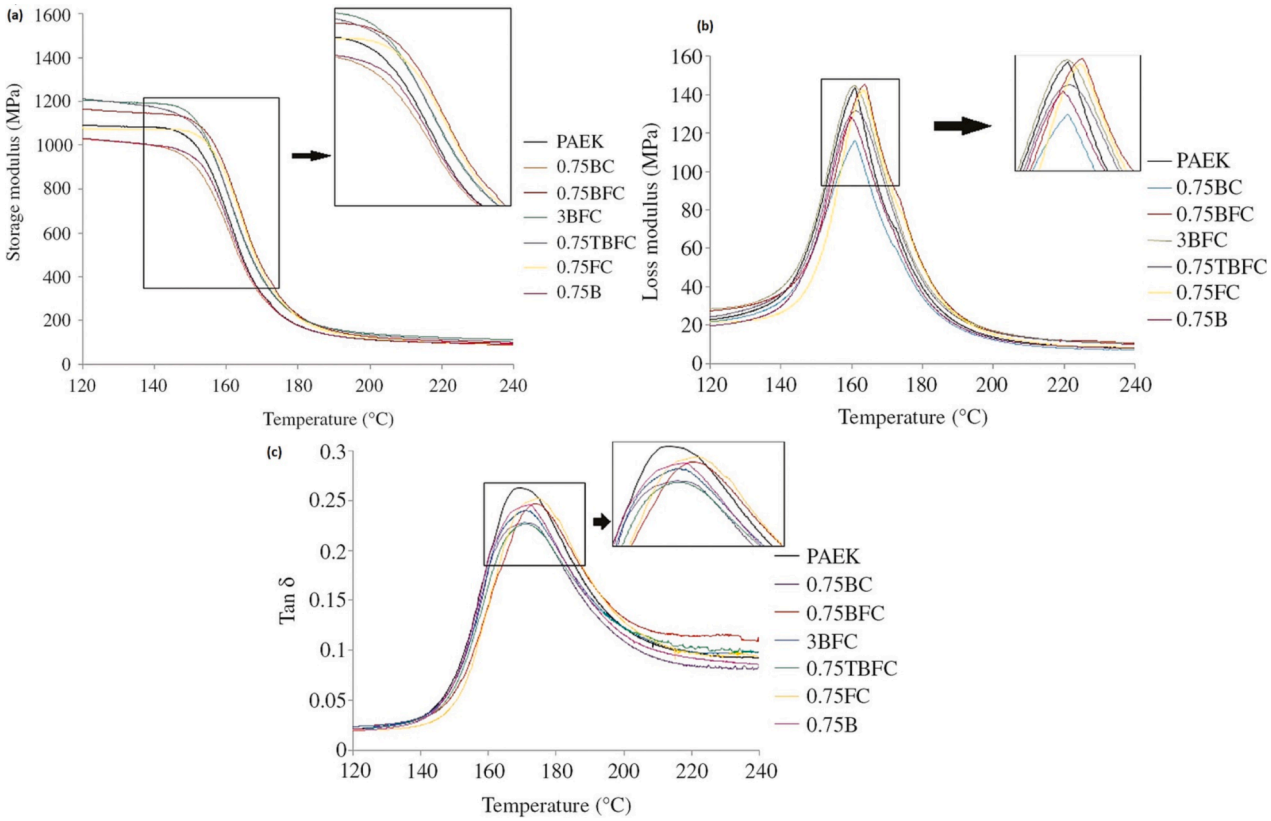

Fig. 13. The variation in storage, loss modulus and tanδ peak of PAEK/Boron carbide/ MWCNT nano hybrid composite systems [83].

matrix. Guth [160,176–177] proposed a polynomial model (Eq. (41)) to predict storage modulus which also considers filler content in the matrix.

$$G_C = G_m\left(1 + 2.5v_f + 14.1{v_f}^2\right) \tag{41}$$

Cohan proposed a model (Eq. (42)) which takes into account the aspect ratio of fillers for evaluating storage modulus of composites.

$$G_C = G_m\left(1 + 0.675\left(\frac{l}{d}\right) + 1.62\left(\frac{l}{d}\right)^2 v_f + 14.1{v_f}^2\right) \tag{42}$$

Another model was proposed by Mooney [160,176–177] which includes crowding factor 'S' (Apparent volume to the true volume occupied by the filler) (Eq. (43)).

$$G_C = G_m exp\left(\frac{2.5v_f}{1 - Sv_f}\right) \tag{43}$$

The rule of mixtures suggested by Nielsen [160,176–177] could effectively predict the damping factor of composites with reinforcements (Eq. (44)).

$$tan\delta_C = v_f tan\delta_f + v_m tan\delta_m \tag{44}$$

In certain cases where fillers offer very low damping, the first term in Nielsen model could be eliminated and equation can be modified as in Eqs. (45) and (46) [160,175,177]

$$tan\delta_C = v_m tan\delta_m \tag{45}$$

$$tan\delta_C = (1 - v_f) tan\delta_m \tag{46}$$

Where $\tan\delta_c$, $\tan\delta_m$, $\tan\delta_f$ are the maxima of tanδ of composite, matrix and filler respectively. Since the interaction of fillers with matrices impose constraints which reduce tanδ values a stiffness term is incorporated, which is the ratio of matrix and the composite modulus [160,175–177]. The modified equation is given as in Eq. (47)

$$tan\delta_C = v_m\left(\frac{E_m}{E_c}\right) tan\delta_m \tag{47}$$

Halpin-Tsai equation [176] is also used to compute storage modulus of composites and also considers the geometry of filler in the model (Eqs. (48) and (49)).

$$G_C = G_m\left(\frac{1 + \xi\eta\varphi_f}{1 - \eta\varphi_f}\right) \tag{48}$$

$$\text{Where} \quad \eta = \frac{\frac{E_f}{E_m} - 1}{\frac{E_f}{E_m} + \xi} \tag{49}$$

For spherical nanoparticles, the factor ξ can be taken as 2. Lewis and Nielson [178] presented the generalized form of Kerner equation as in Eqs. (50)–(52) [176]

$$\frac{G_C}{G_m} = \left(\frac{1 + AB\varphi_f}{1 - B\psi\varphi_f}\right) \tag{50}$$

$$A = \frac{7 - 5\gamma}{8 - 10\gamma} \tag{51}$$

$$\text{and } B = \frac{\left(\frac{E_f}{E_m} - 1\right)}{\frac{E_f}{E_m} + A} \tag{52}$$

A is related to filler geometry and γ is poison's ratio of the matrix. For very large ratios of E_f/E_m B can take a value of 1. If filler modulus is much higher than the matrix modulus then factor ψ can be evaluated using the Eq. (53)

$$\psi = 1 + \frac{\varphi_m}{\varphi_{\max}}\left[\varphi_{\max}\varphi_f + (1 - \varphi_{\max})\varphi_m\right] \tag{53}$$

The parameter ψ depends on maximum packing fraction (ϕ_{max}). In the case of nano sized fillers, since the specific surface area is very large, the volume fraction of the interfacial region is significant and at those regions polymer chain mobility is restricted by the filler. Hence regular micromechanical models may underpredict the modulus of multiscale or dual scale nanocomposites whereas the Eq. (53) predicts a realistic modulus [9,176,178]. The storage modulus of dual and hybrid multiscale nanocomposites could also be predicted by another model using Havriliak- Negami (HN) equation (54) [179].

$$E^*(\omega) = E_\infty + \frac{E_0 - E_\infty}{\left[1 + (i\omega\tau)^\alpha\right]^\beta} \tag{54}$$

This includes calculation of four temperature-independent HN parameters (α, β,$E_0 and E_\infty$) and one temperature dependent parameter 'τ'.$E_\infty and E_0$ are modulus at glassy regime at high frequency and modulus at rubbery regime at low frequencies. The parameters 'α' and 'β' are two adjustable fitting parameters in which α is related to the width of loss modulus peak and b controls the asymmetry of the loss modulus peak and τ is relaxation time of polymer chains that can be evaluated using Eq. (55) [179–180].

$$\tau = \frac{1}{i\omega} exp\left[\frac{1}{\alpha} log\left\{exp\left|\frac{1}{\beta} log\left(\frac{E_0 - E_\infty}{E^* - E_\infty}\right)\right| - 1\right\}\right] \tag{55}$$

In this review an effort has also been made to explicate several models to demonstrate their effectiveness to predict the storage modulus of nanoscale and multiscale hybrid polymer composites. Understanding and modelling of viscoelastic behaviour of polymers and composites with multiple relaxations and at quasi-static and high strain rates is of paramount importance in applications where the composites are subjected to dynamic loading conditions. The evaluation of storage modulus by using these models can be performed using experimental storage modulus obtained using a DMA analyser via shear mode, double cantilever mode, temperature sweep, frequency sweep, dynamic stress–strain studies etc in a temperature range extending up to the melting point of the base matrix. Most of the models discussed under section 4 could be used to predict the storage modulus and the model equations are of a non-linear fitting polynomial form. The accuracy and the suitability of each model in prediction of viscoelastic response of composite system depends on many factors like geometry, content of fillers, frequency, strain rate, type of base matrix, mode of analysis etc.

4.1. Simulation modeling of dynamic mechanical properties

Many researchers have discussed the significance of numerical simulation of interfacial damping between fillers and the matrices. Hwang et al. [181] explained the strain energy method in finite element analysis to investigate the fiber/ matrix interface of carbon fiber reinforced composites. The characteristics of interfacial damping was further explored by Dai et al. [182] and broadened the strain energy method to optimize the damping behavior of CNT/polymer composites by adjusting the filler parameters. Gong et al. [183] investigated the enhancement in damping properties of carbon fiber reinforced composites by interfacial sliding of 2D multilayer graphene oxide in epoxy/ graphene oxide/carbon fabric multiscale composites [183]. They noticed that the deposition of graphene oxide on the carbon fiber can elevate the damping properties of carbon fiber reinforced polymer composites. It was determined experimentally that the damping properties of carbon fiber reinforced plastics were enhanced by 113% at 1 Hz and at a strain of 0.23%. Based on the strain energy method of ANSYS, a numerical parametric analysis was introduced to model interfacial properties of multilayer graphene oxide carbon fiber reinforced composites. In the first step of analysis, the elastic constants of the carbon fabric layer were extracted using Digimat 5.1.1 code, a multiscale design software for analyzing the mechanical behavior of complex multiscale composites. The damping loss factor of the GO/matrix layer was

evaluated using the properties obtained from the first step and the damping parameters of GO/GO interphase by utilizing the properties of GO/matrix layer obtained in the second step. Flow sheet for analysis is as shown in Fig. 14. Harmonic response analysis simulation module in ANSYS was used under the condition of small elastic strain of 2%.

The damping loss factor of fabric layer can be evaluated using Eq. (56)

$$\eta_{CFRP} = \frac{\eta_{fiber} * U_{fiber} + \eta_{matrix} * U_{matrix}}{U_{fabric} + U_{matrix}} \tag{56}$$

And the damping properties of GO/carbon fiber reinforced polymer composites could be obtained from Eq. (57)

$$\eta_{GO-CFRP} = \frac{\eta_{fiber} * U_{fiber} + \eta_{GO-matrix} * U_{GO-matrix}}{U_{fabric} + U_{GO-matrix}} \tag{57}$$

and finally based on strain energy method the damping loss factor of the composite can be calculated by Eq. (58)

$$\eta_{composite} = \frac{\eta_{fiber} * U_{fiber} + \eta_{matrix} * U_{matrix} + \eta_{interphase} * U_{interphase}}{U_{fiber} + U_{matrix} + U_{interphase}} \tag{58}$$

The authors inferred that sliding at interfaces dissipates large amounts of energy and appropriate modulus and loss factor of 0.20 was chosen for the GO-GO interphase on the basis of assumptions used in simulation modeling which conforms to the experimental data. From the literature survey it could be observed that only limited work has been carried out in the simulation modeling of DMA properties. There is a lot of scope to explore the modeling techniques that can be applied in multiscale composites for predicting damping behavior using RVE.

5. Conclusions

From the above discussions it could be inferred that the performance of dual nanofiller polymeric systems and multiscale filler reinforced polymeric composites is influenced by various factors like filler type, base matrix, geometry of fillers and interfacial/adhesion characteristics. Since one has to incorporate dual nanofillers or a single nanofiller along with a micron size filler to prepare the composites, a proper choice of the processing technique is of crucial significance to reach uniform dispersion and distribution of fillers in the matrix. Various processing techniques have been critically analyzed in the current article that could finally reflect on the physical, thermal, electrical and mechanical properties of the composites. Furthermore, the synergism of dual nanofillers or multiscale fillers appreciably enhance and impart superior thermal, mechanical and electrical properties along with the inherent properties of nano fillers like conductivity, flame retardancy, sensing capability, self-healing and energy storage applications. The developed hybrid filler polymeric composites could be named as multi-functional materials and the properties can be tailored to meet the requirements for different applications.

A detailed survey of the scientific reports was carried out to understand the enhancement of modulus, fracture toughness, thermal stability, electrical conductivity network, tribological characteristics, flexural strength, glass transition temperature, interfacial shear strength, crystallization characteristics etc. owing to the compounded effect of hybrid fillers. The influence of various surface treatment techniques on fillers that could enhance their surface energy of which in turn augments the adhesion characteristics between the filler and the matrix is explored. Exceptional enhancement in mechanical properties was reported via functionalization of nanofillers using silane, amine, carboxyl treatments.

Though there are few review articles available on multiscale composites, the effect of and dual-scale nano fillers on static and dynamic mechanical properties are not reported in a comprehensive manner. The suitability of a number of micromechanical models for hybrid composites was detailed and the effective elastic modulus of composite materials was predicted via several micromechanics models and compared with experimental results. Simulation modelling approaches of modulus via finite element method were also discussed to predict the elastic modulus and comparison with analytical and experimental results are performed. Multiscale stress transfer modelling procedure was investigated and stochastic multiscale approach was found to be most appropriate in prediction of elastic properties in the case of multiscale composites. In this approach a separate RVE was defined capturing effective parameters at each scale of nano, micro, meso and macro. In hierarchical multiscale modelling, the output of each scale are fed as input data to the next upper scale. Nano and micro scales were modelled using semi-continuum modelling approaches and upper scales of meso and macro are modelled using micromechanical models. Stochastic implementation of developed model was discussed which also considers non-deterministic parameters like the orientation, volume fraction, waviness/wrinkle, agglomerations etc. The detailed investigations included in this review will help material research groups to gain a broad understanding on the development methodologies, experimental, analytical and simulation strategies of dual nano and hybrid multiscale composites. These classes of advanced materials are potential candidates for next generation multifunctional materials and plays a primary role in materials innovation research.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

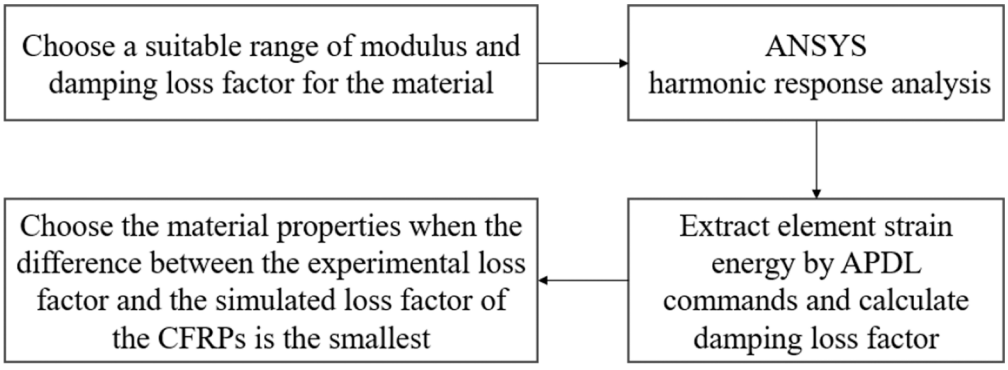

Fig. 14. Flow chart for determining the unknowns using known parameters for evaluation of damping loss factor of multiphase carbon fiber reinforced graphene/epoxy composites.

Data availability

Data will be made available on request.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.compositesa.2023.107741.

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