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# A review on Nanocomposites
# Part 1: Mechanical Properties

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## ABSTRACT

*Micromachining of nanocomposites is deemed to be a complicated process due to the anisotropic, heterogeneous structure and advanced mechanical properties of these materials associated with the size effects in micromachining. It leads to poorer machinability in terms of high cutting force, low surface quality and high rate of tool wear. In part 1 of this two-part review paper, a comprehensive review on mechanical properties of various nanocomposites will be presented while the second part of the paper will focus on the micro-machinability of these nanocomposite materials.*

***Keywords**: nanocomposites; micromachining; manufacturing; materials; mechanical properties*

## 1. INTRODUCTION

The word ”nanocomposite” was first introduced by Blumstein in 1961 [1]. The primitive nanocomposite was investigated in an attempt to improve the thermal stability of nano-silicate reinforced polymethyl methacrylate (PMMA) [2] in 1965. Nanocomposites share similar terminologies as conventional composites in terms of their constituents except for the reinforcement size that is typically in the range of hundreds of nanometers. The reduction from micro-range to nano-range of fillers provides remarkable reinforcements in nanocomposites while requiring much lower content of fillers than the composites with conventional sizes(hereafter, call composites or conventional composites for short), hence leading to inconsiderable increases in weight [3]. Nowadays, many nanocomposites have been discovered and commercially applied in various industrial areas including (and not limited to) aerospace [4], automobile [5] and medicine [6] due to their superior properties, namely mechanical [7], thermal [8], electrical [9], electrochemical, electromagnetic [10], and gas barrier properties [11]. Due to these superior properties, further applications of nanocomposites have been found in terms of manufacturing micro-structured components following the miniaturization trend of modern production. The discovery of nanocomposites has found many applications in microelectronics (Figure 1a, b) [12]. Tang et al. [13] have concluded that carbon nanotubes (CNTs) reinforced polyimide nanocomposite is applicable for microelectronics devices due to its ideal electrical conductivity, storage modulus and environmental stability. Other applications such as high performance transistors from poly-4-vinylphenol (PVP)/ TiO2 nanocomposite [14] or high energy density capacitor from poly-vinylidene-fluoride/TiO2 nanocomposite [15] have exhibited better operations than their neat matrix counterparts. Moreover, this miniaturization trend could cover not only microelectronics but also micromechanical devices. Nanocomposites might be considered as alternatives to composites and alloys in manufacturing micro-products [16]. For example, manufacturing airframe [17] or wings [18] of micro-air vehicles (MAVs) using conventional composites [19] such as carbon fiber, glass fiber or Kevlar reinforced plastics could be replaced by CNTs or carbon nanofiber (CNF) nanocomposites that have higher strength-to-weight ratio and flexibility. The artificial wings for MAVs have been developed using CNT/Epoxy and CNT/PP nanocomposites by Kumar et al. [20]. Additionally, the additions of ceramic nanoparticles improved the tribological property, wear-resistance and overall mechanical properties of metal nanocomposites [21]. Therefore, these nanocomposites could be used in manufacturing pistons or linkage rods [22] (Figure 1c), micro-gears [23] (Figure 1d-f). Due to their huge potentials to produce micro-products, it would be necessary to investigate on the mechanical properties as well as feasible processing methods to fabricate nanocomposites. Most of the recent techniques (Table 1) to manufacture nanocomposites are incapable of producing a final product (in terms of dimensional and geometrical accuracies as well as surface quality), instead, a near-net-shape is produced. Therefore, post-processing or finishing technologies will be always required. Some manufacturing processes such as LIGA (Lithography, Electroplating, and Molding) [24], laser micromachining [25], microextrusion additive manufacturing [26], micro-EDM (Electrical Discharge Machining) [27] , and mechanical micromachining [28] have been used to generate small precision component and micro-structured parts [29].

## 2. NANOCOMPOSITES

In general, nanocomposites still keep the distinct characteristics of both matrix and fillers that make them different from alloys while their final properties depend on their matrix-filler interface bonding, the arrangement of fillers inside the matrix as well as the geometry and content of the fillers. Nanocomposites are also distinguished from composites in which one of the filler’s dimensions is in the range of 1-100 nm**.** Table 2 demonstrates a few fundamental geometries of typical nano-fillers. The specific properties of each category, as well as the effect of filler phases on them, will be discussed in the next sections. Based on the unique properties of different nanocomposites, their potential or commercial applications will be given. In parallel with the discussion about nanocomposites, a comparison between nanocomposites and composites will be addressed to identify the basic differences in terms of the influences of size, content and properties of the filler on their properties.

### 2.1 Differences between nanocomposites and composites

As mentioned earlier, the basic difference between nanocomposites and traditional composites is the size of fillers. The revolution of composites takes place with the size reduction of the fillers from few millimetres in traditional composites to micro-scale (1-100 µm) in modern composites and recently, nanocomposites with the fillers having dimensions in nano-scale. The original for size reduction of filler is to attain a homogenous distribution of filler within the matrix hence reduce the stress concentrations within the composite structure [30] that subsequently improve its mechanical properties. Furthermore, smaller fillers can obtain higher surface energy that makes stronger bonding with the matrix [31] that is, in turn, improves stiffness and strength of the system [32]. In essence, the reinforcing mechanism of composite could be divided into two main types based on the filler scale. For micro-filler, the continuum mechanism is employed to indicate that the micro-fillers bear a fraction of transferred load from the matrix hence the efficiency of reinforcement depends on the adhesion of matrix-filler interface [33]. For nano-filler (10-100 nm), the strengthening mechanism is applied when the matrix-filler interaction is in the molecular level. This mechanism proclaims that the nano-fillers restrict the plastic deformation of the matrix by impeding its dislocations, subsequently leads to the improvement of strength and hardness.

Some researchers have investigated the influence of filler size on the mechanical properties of polymer composites. It was observed that in micro-range of fillers, their size effect on the tensile modulus of composites was unobvious. Some experimental results indicated that the moduli of epoxy-based composites were not considerably improved, or even decreased [34] while using various particle size of $Al_2O_3$ (1-12 µm) [35], glass (4.5-62 µm) [36], or silica (2-47 µm) [37]. The same trend could be seen with PP/ $CaCO_3$ [38], poly-benzoxazine/$CaCO_3$ [39] or polyester/ Al [40] (Figure 2). In addition, the relation between the filler size and tensile strength of composite also not clear. While some studies reported that tensile strengths of composites remarkably increased with the size reduction of fillers, using micro silica particles to reinforce epoxy [37, 41], another result showed no trend of tensile strength variation of epoxy/$Al_2O_3$ when decreasing the filler size [35]. In general, it could be seen that the influence of filler size in micro-scale on mechanical properties of composite is unremarkable.

From the discussion above, it leads to the requirement to investigate the effect of filler size in nano-scales to identify whether their influence on mechanical properties of composites are more sensitive than the micro-counterparts or not. Onuegbu and Igwe [42] adapted various filler sizes (150, 300 and 420 nm) of snail shell powder to reinforce PP. Tensile modulus, flexural strength and impact strength were observed to be improved with the decrease of filler size. However, those improvements were not considerable. For example, the tensile strengths increased by around 5% when decreasing the filler size from 300 to 150 nm for every weight fraction. The improvement of the mechanical properties of composites was only considerable when keep reducing the size of filler below 100 nm. This phenomenon was verified when a comparative study between micro and nano-fillers in terms of mechanical performance of polymer composites was given by Devaprakasam et al. [43]. In this research, micro-silica (100 nm – 4 µm) and nano-silica (40 – 60 nm) were employed and the results exhibited a less variation in the modulus and hardness of the nanocomposite than the composite while applying different loadings. It was explained by the homogenous distribution of nano-fillers, strong interfacial matrix-filler bonding in comparison with micro-fillers. Some other researches also indicated the dominant effect of filler sizes on mechanical properties when they were reduced below 20 nm, as seen in Figure 3. This dominance was also verified by Edwards [44] that a high degree of reinforcement could be achieved using the fillers with sizes in below 100 nm.

This threshold of filler size in which the mechanical properties of composites increase remarkably is called ‘critical size’, according to [45]. Kumar et al. [46] investigated the effect of CNTs ratio on strengthening behavior of polyethene-based nanocomposites. They claimed that employing high-aspect-ratio CNTs provided great hardness and elastic modulus improvement due to high contact surface area and strong interfacial bonding of CNT-polymer. In addition, the presence of mechanical locking was more frequent in terms of high-aspect-ratio CNT due to its smaller diameter and longer length (Figure 4). However, most of the relevant studies only focused on experimental works while a few theoretical models or simulations were found (Figure 5). Constitutive models would be essential to provide quantitative analysis and explanation in terms of strengthening mechanism. Chowdhury and Okabe [47] have applied molecular dynamic (MD) method to investigate the effect of matrix density, chemical cross-links on the interface and geometrical defect of CNTs on interfacial shear strength (ISS) and consequently, CNT pull-out (Figure 5a). From the simulation, it was concluded that high matrix density, presence of cross-link and small cross-link switching contributed to high ISS. A 3D representative volume element (RVE) method has also been applied to assess the effects of CNT waviness, diameter, volume fraction, Poisson's ratio and matrix modulus on interfacial strength of polymer/SWCNT [48] (Figure 5b). Based on the aforementioned studies, it could be observed that investigating the interfacial bonding between nano-fiber and matrix plays a critical role in load transfer assessment and consequently, the mechanical strengthening efficiency of the reinforcement. The stress transfer behavior of SWCNT in epoxy matrix has been studied by Xiao and Zhang [49] using the Cox model for solid fibers [50] (Figure 5c). The main used indicator was stress transfer efficiency δ which is the ratio of maximum tensile stress ($\sigma_{max}$) to maximum shear stress ($\tau_{max}$) in the interfacial section. The effect of SWCNT length, diameter and thickness are shown in Figure 6a. Moreover, a comparison of stress transfer efficiency δ between SWCNT and carbon fiber (CF) has also been expressed with the assumption of the same hollow structure and dimension of CF as SWCNT. The analytical results showed a remarkable improvement of δ (128%) in case of epoxy/SWCNT compared to that in epoxy/CF composite. The increase Young's modulus and structural change (from solid to hollow structure) contributed 69% and 31%, respectively to this enhancement (Figure 6b). Based on that, it could be seen that nano-fibers with higher aspect ratio and tensile properties can provide higher reinforcing efficiency compared to their micro-counterpart. Some researchers have claimed an equivalent or even higher strengthening effect could be achieved from using low loading of nano-fillers compared to that of micro-fillers. Fornes [51] indicated that the modulus of nylon 6 would be doubled by mixing only approximately 6.5 wt.% nano-clay montmorillonite (MMT) platelets whereas the same effect could be achieved when using three times volume of glass fiber or Campbell [52] claimed that a desired strength and stiffness of composite could be achieved when using 50 wt.% microparticles or 70 vt% of micro-fibers. Therefore, it could be seen that nanocomposites can provide excellent mechanical properties with low filler contents that can only be obtained using high filler content in conventional composites [53] since they proved as an efficient material to be used in demanding applications which require excellent strength to weight ratio. The improvement of stress transfer efficiency δ due to nanotube's diameter increase has also been confirmed by Li and Saigal [54] using shear-lag analysis and the representative volume element (RVE) (Figure 5d). In addition, they also considered the effect of fiber volume fraction as it has not been considered in previous studies.

The strengthening efficiency of nano-fillers such as CNT in MMNCs was also investigated and its mechanism is following load transfer [50], Orowan strengthening [55] and thermal expansion mismatch [56]. The strength improvement that is contributed by metal matrix grain size refinement [57], resulted from the addition of CNT has been found by [58]. Similar to polymer-based nanocomposites, the effect of reinforcement size also significantly contributed to strengthening behavior of MMNCs [59-61]. However, there were only a few models published that provided a comprehensive explanation of the strengthening mechanism of CNT reinforced MMNCs. Barai and Weng [62] have developed a two-scale model to analyze the elastoplastic behavior of CNT reinforced MMNCs that considered CNT agglomeration and interface properties as two main factors affecting the load transfer. Dong et al. [63] have built a dislocation model that combined the effect of both matrix grain size and filler size on the strengthening mechanism of metal/CNT nanocomposites (Figure 7). They claimed that the load transfer effect was improved at small grain size and high volume of CNT.

### 2.2 Mechanical properties of nanocomposites

From the discussions about the differences between nanocomposites and composites in the previous section, it could be intrinsically seen that reinforcing a matrix material by nano-fillers could lead to higher effectiveness of reinforcement than micro-fillers due to their advanced mechanical properties and novel nanostructures. Among many nano-fillers have been applied recently, CNTs, graphene and nano-clays are the most common reinforcing materials due to their compatibility and high efficiency of reinforcement with three main matrix materials including polymers, metals and ceramics. The major objective of the most relevant studies is to investigate the changes between the neat matrix and the matrix reinforced nano-fillers in terms of mechanical, thermal and electrical properties that are most common. Within the scope of this review, only the mechanical properties will be considered due to the requirements for the other sections related to the micromachining of nanocomposites. The effectiveness when reinforcing nano-fillers depends on many factors which can be classified into three main groups: (i) Nano-filler nature, (ii) Filler-matrix interaction and (iii) Fabricating methods.

#### *2.2.1 CNT based nanocomposites*

CNTs are allotropes of carbon that made of a cylindrical rolled-up single layer of the carbon atom. The diameters and lengths of CNTs range from 1-100 nm and 0.1 – 100 µm, respectively [64] with the tubular structure make them very high aspect ratio materials with the surface areas are in the range of 200–900 $m^2/g$ [65]. CNT was first discovered by Iijima in the transmission electron microscopy (TEM) image in 1991 [66] and the first single-walled carbon nanotube was synthesized in 1993 by the same author [67]. Many applications have been found for CNT in different domains such as drug delivery [68], health care [69], electronics [70], electrics and thermal applications [71]. Due to their high strength-to-weight ratio, aspect ratio, thermal and electrical properties [72, 73], CNTs have found huge potential applications on composite reinforcement.

##### *2.2.1.1 CNT reinforced polymer matrix nanocomposites*

CNTs have been employed to reinforce polymers due to their better interfacial interaction in comparison to ceramic [74] or metal matrix [75] and the similar characteristic of organic structure. Therefore, it has been witnessed that there were some polymer/CNTs nanocomposites with tensile strengths ranging from 0.1 to 5 GPa and Young's modulus from 5 to 200 GPa [76]. Generally, the mechanical properties were improved when using CNTs as reinforcement in some polymers such as epoxy [77-80], polystyrene (PS) [81], polyethylene [82, 83], PMMA [84, 85], poly (p-phenylene benzobisoxazole) (PBO) [86], polyvinyl alcohol (PVA) [87], polyester elastomers (PEE) [88], polycarbonate (PC) [89], polyamide-6 [90] and nylon-6 [91]. The optimum loading of CNTs has to be considered carefully to avoid agglomerations or bundles that negatively affect the mechanical properties of PMNCs, usually when CNTs content exceeds 2-3% [92]. Figure 8 shows some improvements in terms of Young's moduli and tensile strengths when reinforcing polymer by CNTs with the corresponding dispersion techniques. Generally, it could be seen that the level of CNTs distribution in polymer matrixes, their interfacial interaction and processing methods significantly affect the load transfer from the matrix to CNTs, hence decide their effectiveness of reinforcement in terms of mechanical properties of polymer-based nanocomposites. From Figure 8, it was observed that both Young's modulus and tensile strength of nanocomposites improved with the addition of CNTs. However, there are different thresholds of CNTs loading at which the tensile strengths decrease or even lower than pristine polymers. It is possibly due to the re-agglomeration [93-95] of CNTs due to insufficient dispersion techniques, high loading of fillers hence creating the more stress concentration and reducing the effectiveness of CNT as reinforcement. Specifically, the poor interfacial interaction between CNT and polyester even leads to the negative influence on tensile strength of nanocomposite [96, 97]. In terms of dispersion methods, solution mixing, in situ polymerization dry mixing and melt mixing are the most common methods for fabricating polymer based nanocomposites. Esawi et al. [98] have investigated the effects of dispersion methods (dry mixing and solution mixing) on the mechanical properties of PP/CNT nanocomposites (Figure 9). The results have indicated a higher level of CNT distribution when using dry-mixing in comparison with solution-mixing. The degradation of the polymer, high viscosity due to the addition of solutions that limited the CNT distribution contributed to the lower improvements of mechanical properties when applying solution-mixing. In addition, employing ultra-sonication in solution-mixing might damage the CNT structure, hence also contributed to low mechanical properties of nanocomposites [99]. Associated with the effect of CNT content, the influence of CNT structure on the mechanical properties of CNT-based nanocomposites has been also indicated considerable. Since CNTs have various types: SWCNTs, double-walled CNTs (DWCNTs) and MWCNTs, their different structures and properties lead to various efficiencies in terms of mechanical reinforcement. Figure 10 shows the improvements in tensile properties when reinforcing epoxy with different types of CNTs. Theoretically, the addition of SWCNTs and DWCNTs exhibited higher reinforcement of mechanical properties than MWCNTs due to their higher mechanical properties, aspect ratio and specific surface area. In addition, the multi-layer structure of MWCNTs basically leads to the low effective surface area in comparison with less-layer structure CNTs. However, this reinforcing effectiveness also depends on how homogenous the CNTs distribute within the polymer matrix. In this case, DWCNTs showed no agglomeration as SWCNTs that explained for a higher improvement of tensile properties. This phenomenon was also verified by Fornes et al. [100] and Sennett et al. [101] that the dispersion of MWCNTs within PC matrix was much more effective than SWCNT regardless the fabricating routes. They explained that SWCNT had a high propensity to re-agglomerate during the synthesis making the exfoliation more difficult in comparison with MWCNT.

Besides the structure, CNTs alignment also has a dominant effect on the mechanical properties of polymer-based nanocomposites. This feature could be attained using several methods such as shear flows [102], ex-situ alignment [103], force field-induced alignment [104], magnetic field-induced alignment [105], electrospinning-induced alignment [106] and liquid crystalline phase-induced alignment [107].

Figure 11 shows some experimental results that exhibited the improvements of tensile properties of CNT based nanocomposites with aligned CNTs in comparison with non-aligned CNTs. It could be basically explained by the isotropic nature of nanocomposites when CNT alignment was employed and also leading to better distribution and reducing agglomeration of CNT when filling into the polymer matrix. In addition, functionalization has been considered as an effective treatment of CNTs to improve their interactive adhesions with polymer matrix, hence enhance the reinforcing effectiveness of mechanical properties through load transferring. Khare et al. [108] have indicated that reinforcing epoxy by amido-anime functionalized CNTs (f-CNTs) led to a higher improvement of mechanical properties in comparison with pristine-CNTs (p-CNTs) (~ 51% of Young’s modulus). Some main reasons were given including lower interphase compression, matrix structure integrity, suppression of matrix mobility, stable-covalent bonds of epoxy-FCNTs and subsequent facilitation of load transfer (Figure 12).

Unlike the improvements of tensile strength and Young’s modulus, the addition of CNTs exhibited different variations of fracture strain and toughness. Figure 13 shows the different trends of fracture strain improvement between thermoplastic and thermosetting reinforced by CNTs at various weight content. It could be seen that the flexibility of thermoplastic nanocomposites was significantly decreased at every filler loadings. Although relevant studies have shown this phenomenon, a comprehensive mechanism to explain it has been not proposed. Wang et al. [109] claimed that the degradation of flexibility when adding MWCNT-$NH_2$ into Polyimide (PI) was possibly due to strong interface interaction between matrix and filler. Consequently, the movement of polymer chains under loading could be restricted and hence, decreasing the flexibility of this material. However, the characterization of interfacial strength has not been made to support this claim. On the other hand, the fracture strains of thermosets could be enhanced by the addition of CNTs at certain levels of contents which is different from thermoplastic nanocomposites. When reinforcing epoxy by MWCNT, Chen et al. [110] indicated that the brittle epoxy phase was toughened by dispersed CNT. This enhancement of failure strain at certain low filler loadings (below 1 wt.%) was confirmed by Gojny et al. [111]. At higher CNT loadings, CNT agglomeration led to stress concentration and weaken the interfacial interaction of polymer-CNT, hence reducing the fracture strain. Scanning electron microscope (SEM) imaging was employed to demonstrate these explanations in terms of CNT distribution at different filler contents.

Regarding the fracture toughness, CNTs have been qualified as a potential reinforcing candidate to replace glass fiber (GF) or CF to attain higher toughening efficiency due to their high aspect ratio and stiffness [112]. The micro-mechanical toughening mechanism of polymer reinforced CNT nanocomposites can be expressed as follows: (1) crack bridging by CNTs and (2) CNTs de-bonding and pull-out or breaking depends on the interface strength and applied load [113, 114] (Figure 14). Based on this schematic, it could be seen that the highest fracture toughness could be achieved if CNTs are oriented transversely with the propagated cracks in which the bridging mechanism takes effect. Otherwise, it will not have considerable influence on fracture toughness in the case of longitudinal or random distribution of CNTs [115]. In addition, the effect of transverse alignment could be only effective at a low loading of CNTs. Some studies have shown a maximum enhancement of fracture toughness (around 51%) of polymer/CNT nanocomposites at 3 wt.% of fillers was used and decreased when exceeded this threshold due to filler agglomeration [115, 116]. On the other hand, the random distribution of CNTs did not show considerable enhancement in toughness at low filler content (< 1wt.%) [117]. In addition, Chen et al. [114] have analyzed the effect of CNT length and interface strength on fracture toughness. They claimed that the fracture toughness could not be improved with the consideration of interface chemical bond density or fiber length only but combined them together. The optimal values of these indicators were around 5-10% and 100 nm, respectively. Optimal CNT-bridging from their experiment has confirmed this theoretical analysis. Generally, reinforcing polymers by long CNT can achieve high fracture toughness due to its high load transfer and hence, improving the interface shear strength [118, 119]. The structure of CNTs has also influenced the fracture toughness of nanocomposites. Low contents of DWCNT (<0.5 wt.%) have shown a remarkable improvement of fracture toughness of epoxy-based nanocomposites due to its high compatibility with epoxy matrix (37). However, using suitable dispersion methods and functionalization could make MWCNT better reinforcing candidate to enhance fracture toughness than other types of CNTs (18). In general, the fracture toughness improvement when reinforcing polymer by CNTs has shown a complicated correlation between various factors including CNT content, structure, alignment, treatment as well processing technique. Table 3 shows some fracture toughness improvements of polymer reinforced CNT nanocomposites with the consideration of the aforementioned factors.

#### *2.2.1.2 Carbon nanotube reinforced metal matrix nanocomposites*

Although CNTs are theoretically considered an effective reinforcement for high strength-to-weight materials, few studies have concerned about using them to reinforce metals. The incorporation of CNTs in metals has some difficulties due to the inherent characteristics of both CNTs and metals, the fabricating conditions that negatively affect the interfacial adhesion as well as CNTs defects and subsequently, the improvement of mechanical properties of metal reinforced CNT nanocomposites. In general, there are two main challenges that hinder CNTs from achieving high effectiveness of reinforcement in the metal matrix. The first challenge is the poor interfacial adhesion between CNTs and metals. It is due to the nature of CNTs such as low compatibility with high surface energy (72.8 $mJ.m^{-2}$) [120] high surface tension (721 mN/m) [121], low wettability or hydrophobicity [122] and high possibility of agglomeration because of Van der Waals forces [123]. Because chemical bonding between CNT-metal is neglected, these physical factors are dominant in the interfacial interaction [121]. Secondly, the undesirable chemical reactions between CNTs and metals at high temperature, pressure from fabricating conditions such as sintering, hot milling. It leads to the formation of intermetallic such as $Al_4C_3$ [124], TiC [125], $Al_2MgC_2$ [126]. Generally, a minor formation of carbide can positively improve the interfacial adhesion while an uncontrolled process could lead to serious damages of CNTs and in situ carbide formations [127, 128] or enhance CNTs amorphization as well as their thermal decomposition [129]. Some solutions have been adapted to improve the wettability and distribution of CNTs as well as their interfacial strength with metal matrix such as chemical functionalization [130], surfactant [131], coating CNTs with Ni [132, 133], Si [134] or in situ synthesis CNTs within metal matrix [135]. In general, some main improvements in terms of tensile properties and hardness when reinforcing metals by CNTs are shown in Table 4. The effectiveness of the improvement of mechanical properties of metals reinforced CNTs depends on the homogenous distribution of CNTs [132, 136-139], interfacial strength of metal/CNT [132, 133, 139, 140], thermal expansion mismatch between metal and CNTs [141], grain refinement effect [133, 142], CNTs structure retention of plastic deformations of metal matrixes [132, 139, 140, 143], that all contributed to strengthening mechanism. The two last ones also contribute to the hardness improvement of metal/CNT nanocomposites.

### 2.2.2 Graphene-based nanocomposites

Graphene is a planar sheet of a single layer of $sp^2$-bonded carbon atoms that is considered as an original structure element of other carbon allotropes such as CNTs, graphite and diamond. This two-dimensional (2D) structure provides graphene with a much larger specific surface area of ~ 2600 $m^2/g$ than other carbon allotropes such as carbon black (CB) or CNTs [144]. Graphene exhibits exceptional mechanical properties such as Young's modulus (~1 TPa) or strength (130 GPa) [145] hence considered as the strongest material [145]. Moreover, this material is also an excellent conductor with high thermal and electrical conductivity of ~4000 $WmK^{-1}$ [146] and ~ 6000 $S.cm^{-1}$ [147], respectively and other properties such as gas impermeability, optical transmittance. Therefore, graphene has high potential in a wide range of applications such as flexible electronic devices, transparent coating material, energy storage, and especially, nanocomposites [148-150].

### *2.2.2.1 Graphene reinforced polymer matrix nanocomposites*

Many researchers have attempted to investigate the reinforcing efficiency of graphene in nanocomposites by considering various factors such as the effective modulus, filler dispersion [151], alignment [152], agglomeration [153] or fabricating methods [11]. All of these factors have certain influences on the filler distribution and graphene-matrix interfacial adhesion or stress transfer that subsequently affect the reinforcing effectiveness of graphene-based fillers.

The effective modulus of thermal expanded graphite oxide (TEGO) has been measured when it was mixed with PC and polyethylene. Only a slight improvement of modulus was experimentally observed in comparison with graphite-based composites while its effective modulus was around 70 GPa-7% of the value for defect-free graphene (~1 TPa) [154, 155]. This low effective modulus of graphene is explained by its wrinkled geometry once dispersed in the matrix [156], which consequently unfold under tensile load instead of stretching. The incomplete exfoliation could also lead to the aspect ratio reduction of graphene [51] that contributes to this phenomenon.

Graphene-matrix interfacial adhesion is considered as another crucial factor that affects the effective reinforcement [157, 158] since it makes ineffective dispersion as well as load transfer, hence resulting in a low modulus of nanocomposites [159]. Some significant increases of composite tensile moduli were observed when using graphene as reinforcement due to the roughness of the platelets [160] that attribute to mechanical interlocking within the matrix and hence, strong interfacial bonding.

In addition, the reinforcing effectiveness of graphene also depends on the processing method. For instance, melt mixing has been found less suitable in graphene-based nanocomposites since this method causes particle attrition [161] that possibly decrease the filler ratio. In situ polymerization, on the other hand, can generate good dispersion but it also causes polymer chain extension by graphene, resulting in a less modulus improvement of composites than solution-mixing method [11]. This comparison between different processing methods can be seen in Figure 15.

Table 5 summarizes some improvements in mechanical properties when reinforcing polymers with graphene. In general, the elastic moduli increased with the addition of graphene while the tensile strengths decreased. It is explained by the improvement of interfacial interaction of matrix-filler due to the high aspect ratio of graphene [162] that effectively bridging with the matrix molecules and leading to a high stiffening effect [163]. As a consequence, the tensile strength of nanocomposites would be reduced. Moreover, this reduction is also due to the fabricating methods, re-agglomeration of graphene that constitutes some defects in the polymer matrix. The presence of graphene also opposes the flow of amorphous phase and increase the crystallization of hard segment of a polymer matrix that subsequently contributes to the elastic modulus improvement and the tensile strength reduction of polymer-based nanocomposites [164].

Similar to CNTs, the addition of graphene into the polymer matrix leads to different variations in terms of fracture strain and toughness. Fracture strain showed a significant reduction in the case of thermoplastic/graphene nanocomposite while this property was improved for thermoset plastic reinforced with low loading of graphene (Figure 16). However, the fracture toughness improvement when using graphene seems to be higher than CNT. Domun et al. [165] have collected experimental values of fracture toughness when using graphene to reinforce epoxy from relevant studies (Figure 17). They claimed that among common nano-fillers (CNTs, graphene, and nano-clay), graphene exhibited higher fracture toughness enhancement in epoxy-based nanocomposites, mostly at low filler loadings (<1 wt.%). It is due to the higher surface area, aspect ratio of graphene, as well as its exceptional stiffness and strength, compared to CNT as aforementioned. Rafiee et al [166]., in a comparative study, have claimed that graphene exhibited better mechanical reinforcement including Young’s modulus, tensile strength and toughness than MWCNT. It was due to poor interfacial contact area, wetting and adhesion of MWCNT that showed ineffective reinforcing epoxy-based nanocomposites. On the other hand, higher aspect ratio allowed more interfacial contact between graphene and epoxy matrix in contrast with the only outer tube of MWCNT. In addition, wrinkled sheets of graphene also contributed to better interfacial binding, hence improving the toughening mechanism in epoxy/graphene nanocomposites. The micro-mechanical toughening mechanism of polymer reinforced graphene is also different from that in CNTs. While filler pull-out and de-bonding still exhibit along the cracks, there is no presence of graphene cracking. When the crack reaches to the graphene surface, it will be deflected and bifurcated then propagating around the filler. The ease of shearing between graphene sheets also allows the crack propagation goes through or penetrates within layers (Figure 18).

#### *2.2.2.2 Graphene reinforced metal matrix nanocomposites*

Graphene has been also applied to reinforce metal with the aim to improve their stiffness but subsequently leads to the reduction of ductility that was called ‘strengthening mechanism’. However, tensile strengths have been found an increase in some cases [167-169] possibly due to the straightening of wrinkled graphene during the plastic deformation or ball milling process [170]. In general, the strengthening mechanism is related to the improvements of yield strength [171], elongation [168], flexural strength [172], but Young's modulus and hardness are most fundamental objectives. The improvements of Young's modulus when reinforcing metal with graphene is ascribed to the homogenous distribution of graphene [173], compact interfacial bonding of metal-graphene [174] that lead to effective load transfer [175]. On the other hand, the improvements of hardness in nanocomposites are possibly due to the mismatch of thermal expansion coefficient [176] between graphene and metal matrix as well as the restriction or obstruction of graphene in plastic deformation [177-180]. In addition, the formation of metal carbide due to the chemical reaction between graphene and metal during synthesis contributes to the strengthening effectiveness. Al carbide ($Al_4C_3$) has been observed in Al/graphene nanocomposite synthesis using hot extrusion [171]. However, its role in terms of improvement of hardness has been still controversial whether it leads to the reduction of strength, hardness [171] or enhancement of bonding strength, load transfer [181]. The interfacial reaction can be improved by the formation of carbide because it decreases the contact angle of liquid alloy hence increase the wetting [180]. Furthermore, using metal powder through ball milling and sintering processes in high temperature can cause oxidation or the formation of metal oxide. Song et al. [182] reported that the hardness of Cu/ graphene nanocomposite was improved due to the presence of $Cu_2O$ during the synthesis but it was reduced when increasing graphene loading. Similarly, Lin et al. [180] indicated that the formation of $Fe_3C$ after sintering decreased the contact angle between liquid-alloy and GO, hence increasing GO wettability (Figure 19). In case no carbide is formed, the interfacial bonding of metal-graphene strongly depends on matrix nature (contact angle and cohesive energy of liquid-metal and filler) that has been addressed in case of Mg matrix [183]. Hwang et al. [184] have confirmed the compatibility between Mg and graphene with their high adhesion energy. In addition, pre-coating metals such as Ni on graphene surfaces have shown similar effects on improving the wettability of fillers in the Cu matrix [185] as compared to CNT cases [186]. Xu and Buehler [187] have pointed out that Ni-graphene has higher cohesive energy and interfacial strength than Cu-graphene.

In general, the improvements of mechanical properties of metal/graphene nanocomposites have shown their dependences on integrity of graphene [172], graphene exfoliation [188] and distribution [173], interfacial bonding [172], prevention of metal dislocation [179] beside the aforementioned roles of intermetallic formation due to sintering process (Table 6). These formations of metal carbide, metal oxide through the synthesis might contribute to these improvements but cannot be evaluated whether they have positive or negative effects on mechanical properties of graphene reinforced metal nanocomposites.

### *2.2.3 Ceramic based nanocomposites*

In this section, the other nano-filler based nanocomposites will be reviewed in terms of their mechanical properties. There were many types of nano-fillers instead of CNTs and graphene that make the discussion in details for all of them not feasible within the scope of this review section. Therefore, some common nano-filler based nanocomposites will be taken into consideration including polymer/nano-clay, polymer/ceramic and metal/ceramic nanocomposites. These selections are also based on the effective reinforcements of nano-fillers with their corresponding matrices as well as their important applications as engineering materials.

Beside of CNTs and graphene, nano-clay and ceramic nanoparticles have also been applied as reinforcement in polymer materials due to their advanced tensile properties. Nanoclays are potential candidates for nanocomposite reinforcements, especially in terms of mechanical properties. The most common type of nanoclays has been applied in reinforcing nanocomposites is montmorillonite (MMT). It is derived from absorbent aluminum phyllosilicate clay called” bentonite” and thus named “organoclays”. MMT has been applied for reinforcing polymers due to their high aspect ratio and unique intercalation/exfoliation natures [189]. The incorporation of organoclays into polymer matrices has exhibited superior strength and modulus [190]. The most applied polymers by nanoclay reinforcements are polystyrene [191], epoxy resin [192], poly(methyl methacrylate) [193], poly(ε-caprolactone) [194], PP [195], polyurethanes [196], polyimides [197]. However, nanoclays mostly require pre-exfoliation to attain homogenous distribution due to higher specific area and hence higher effective of load transfer could be achieved [198]. In general, the effectiveness of nanoclay reinforcements have been indicated strongly depend on intercalation methods [199], exfoliation nanoclays [200-202], the integrity of nanoclays [203] or matrices [204], and most importantly, the level of distribution of nanoclays [205] or interaction of polymer-nanoclays [201, 205]. However, the addition of nanoclays in polymer exhibited some negative effects on tensile strength of nanocomposites in some cases but no explanation was given [205].

On the other hands, silica nano-particles are also common nano-fillers that have been applied to reinforce polymers due to their high mechanical properties. The processing methods to fabricate these materials are similar to other polymer-based nanocomposites such as melt mixing for PP (63), PE (71n), PEN (62), PET (102), solution mixing for epoxy [206], poly(4-methyl-2-pentyne) (PMP) (123, 124), or sol-gel processing for epoxy [207]. In general, the additions of nano-silica in polymer matrices lead to the improvements of stiffness mostly due to the homogenous distribution, high aspect ratio and stiffness of silica nano-particles. However, it could be seen that their effectiveness of stiffness improvement are not high as CNTs, graphene or nano-clays (Table 7).

In the case of ceramic reinforced MMNCs, the main challenge is the incorporation of ceramic nano-particles into molten metal matrices due to their poor wettability. Besides high wetting could be seen in some metal/ceramic systems with strong (chemical) reactions and low contact angles (θ) such as Cu/WC ($\theta = 20^0$) or Au/$ZrB_2$ ($\theta = 25^0$), most of the other ceramic nano-fillers generate non-covalent (physical) bonds with liquid metals with low wettability such as Ag/$Al_2O_3$, Cu/$SiO_2$ ( $\theta = 120°–140°$); Au/BN($\theta = 135°–150°$) at high temperature [208]. The inhomogeneous distribution of ceramic nano-particles and their agglomerations within metal matrix have been indicated resulting from their low wettability associated with high specific surface areas. Some dispersion routes have been employed to overcome these obstacles including ex-situ and in-situ techniques [209]. The basic difference between these two methods is whether the reinforcements (ceramic nano-particles) are fabricated within the matrices (in-situ) [210] or separately synthesized outside by CVD [211], spray conversion process [212] or laser-induced gas-phase reaction [213] and then subsequently incorporated into metal matrices via powder metallurgy (PM) [214] or mechanical alloying (MA) [215]. Although the traditional PM method has been successfully applied to synthesis metal/ceramic nanocomposites, especially in aluminium-based matrices, it still exhibited obvious agglomerations of ceramic nano-particles and hence their inhomogeneous distribution [216, 217] (Figure 20). The low wettability of as-produced ceramic nano-particles, as well as their high specific surface area, have indicated the main reasons for this drawback. It could be improved by employing MA methods with better dispersion of ceramic nano-fillers with significant grain size reduction [218, 219]. In addition, the densification of MA is mainly conducted by hot pressing, HIP or extrusion. On the other hand, In-situ route has been claimed more suitable than the aforementioned methods in terms of generating the homogenous distribution of ceramic nano-particles such as $Al_3Ti$ [220] or TiC [221] at high loadings (50 vol.% and 18 vol.%, respectively) and subsequent ultrafine microstructures. In general, the additions of ceramic nano-particles lead to the improvements of tensile strength, yield strength and hardness associated with the sacrifice of ductility. All these changes in tensile properties follow strengthening mechanisms including Orowan strengthening, mismatch of thermal expansion coefficients between matrix and filler (CTE) and grain size refinement. In addition, the reductions of ductility have been claimed due to the presence of porosities within the system or brittle nano-fillers [216, 222] although some improvements of ductility could be seen when using Mg-based matrices [223, 224] without sufficient explanation. The role of ceramic nano-fillers on hindering the matrix dislocations and the effect of grain size reduction have discussed in most relevant researches that were the main reasons for hardness increments [216, 222, 223, 225-228]. Table 8 summarizes the mechanical properties of some common ceramic nano-particles reinforced MMNCs with their corresponding fabricating methods and discussion.

## 3. CONCLUSIONS

The first part of this paper has addressed relevant studies including the fabrication and characterization of nanocomposites. Based on that, it has been observed that the addition of nano-fillers as reinforcement improved mechanical properties of nanocomposites. The level of enhancement depends on many factors such as the nature of both filler and matrix, their interaction, filler size and fabricating methods. Strenthening and toughening mechanisms were also employed to explain these variations via modelling and experimental approaches. This review paper has focused on nanocomposite materials that require mechanical micromachining processes in order to produce the final product. These include polymer/CNT, polymer/graphene and metal/ceramic nanocomposites. This part of the review will be the basis to analyse the machinability of nanocomposites when employing mechanical micromachining techniques where the mechanical properies will be correlated with different machining measures.

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**Figure Captions List**

Figure 1 Micro-features as applications of micromachining of nanocomposites. (a - b) SEM images of MTA/MWCNT micro-capacitors and micro-resistors made by precise 3D printing (Reprinted from [12] Copyright 2016, with permission from Society of Photo-Optical Instrumentation Engineers (SPIE)); (c) SEM image of a stainless steel/Al2O3 piston and a linkage rod made by soft molding (Reprinted from [22] Copyright 2009, with permission from Acta Materialia); (d) SEM image of a stainless steel/titania micro-gear made by soft molding (Reprinted from [23] Copyright 2009, with permission from Elsevier); (e) Optical image of a Epoxy/SiO2 wheel and (f) SEM image of a epoxy/SiO2 micro-gearwheel made by UV-LIGA methods (Reprinted from [229] Copyright 2009 , with permission Elsevier)

Figure 2 Normalized tensile modulus of epoxy-based composites as a function of micro-filler size (Adapted from [35-37], [230])

Figure 3 Normalized tensile modulus of polymer-based nanocomposites as a function of nano-filler size (Adapted from [231, 232])

Figure 4 Graphical representation of the effect of CNT aspect ratio while interacting with polymer chains (Reprinted from [46] Copyright 2015, with permission from Elsevier)

Figure 5 Theoretical models representing the micromechanical strengthening mechanism of polymer/CNT nanocomposites via the analysis of interfacial stress transfer with the consideration of various factors: (a)

Effects of polymer matrix density, chemical cross-links and CNT defect (Reprinted from [47] Copyright 2006, with permission from Elsevier); (b) Effects of SWCNT waviness (Adapted with permission from [48] Copyright © 2011 Elsevier); (c) Effects of tube length and diameter (Reprinted from [49] Copyright 2004, with permission from Woodhead Publishing); (d) Effects of nanotube aspect ratio and fiber volume fraction (Reprinted from [54] Copyright 2006, with permission from Elsevier)

Figure 6 (a) Effect of SWCNT's diameter, thickness and length on the stress transfer efficiency (δ); (b) Effect of Young's modulus on the stress transfer efficiency (δ) (Assumed that CF had the same hollow structure with d= 3nm and t= 0.142 nm as SWCNT) (Fiber volume fraction Vf ~ 0.17 % )(Reproduced from [49]); (c) Effect of nanotube diameter on stress transfer efficiency (Vf = 0.1%); (d) Effect of nanotube volume fraction on stress transfer efficiency (d = 0.7086 nm) (Reproduced from [54])

Figure 7 (a) Schematic of the microstructure of CNT-reinforced MMCs with DPZs. (b) The entire composite is decomposed into CNT and effective matrix phase, and the effective matrix is comprised of DPZs and pure metal matrix (Reprinted from [63] Copyright 2014, with permission from Elsevier)

Figure 8 Mechanical properties of polymers reinforced CNTs with corresponding dispersion techniques (Adapted from [94, 96, 100, 233-236])

| | |
|---|---|
| Figure 9 | Effects of dispersion method on mechanical properties of CNT reinforced PMNCs (Reproduced from [98]) |
| Figure 10 | Effect of CNT structure on mechanical properties of polymer nanocomposites at a filler content of 0.3 wt.% (Adapted from [117]) |
| Figure 11 | Effect of CNT alignment on mechanical properties of epoxy-based nanocomposites (Adapted from [237, 238]) |
| Figure 12 | Effect of CNT functionalization on tensile modulus of CNT reinforced epoxy matrix nanocomposites (Adapted from [80, 108, 117]) |
| Figure 13 | Fracture strain improvement of CNT based polymer nanocomposites as a function of filler content (Adapted from [94, 96, 109, 110, 239-241]) |
| Figure 14 | Schematic representing fracture toughening mechanism of CNT reinforced polymer nanocomposites (Reprinted from [113] Copyright 2017, with permission from Elsevier) |
| Figure 15 | Normalized Young's moduli of TPU/graphene nanocomposites in different processing methods) (Adapted from [11]) |
| Figure 16 | Fracture strain improvement of graphene-based PMNCs as a function of filler content (Adapted from [164, 242-244]) |
| Figure 17 | Comparative fracture toughness improvement of epoxy-based nanocomposites using different nano-fillers [165] (Open access from The Royal Society of Chemistry) |
| Figure 18 | Schematic representing fracture toughening mechanism of CNT reinforced PMNCs (Adapted from [152] Copyright 2015 The Authors – Open access from Elsevier) |

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| Figure 19 | The formation of carbide when fabricating metal/GO (2 wt.%) nanocomposites (Reprinted from [180] Copyright 2014, with permission from Acta Materialia) |
| Figure 20 | The agglomerations of ceramic nano-particles in Al matrices: (a) Al/Al2O3 4 vol.% [217] and (b) Al/SiC 3 wt.% [216] (Open Access from Metals) |

## Table Caption List

## Information Regarding Figures and Tables

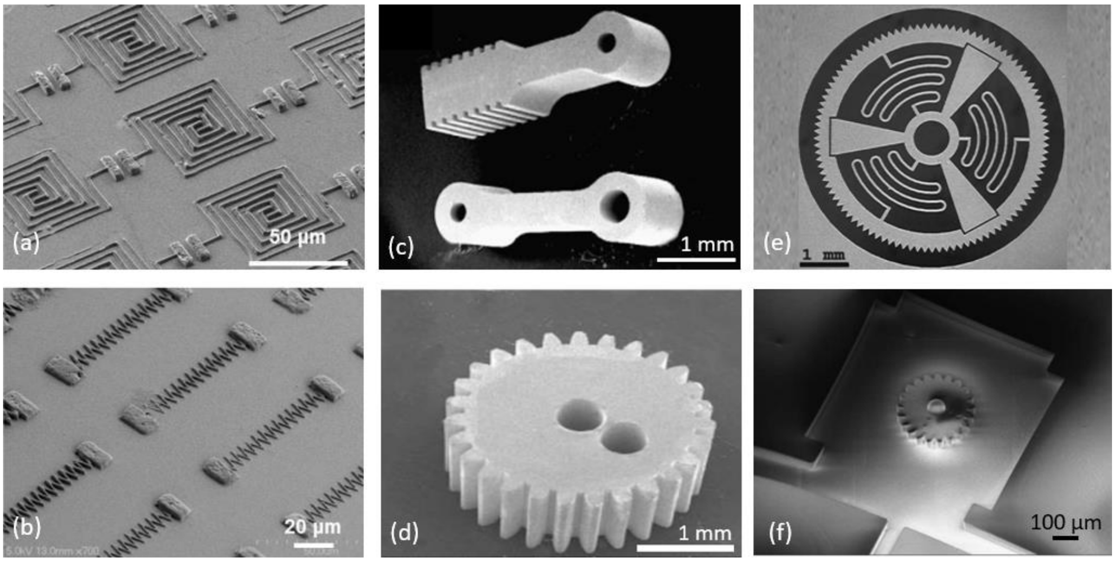


Figure 1: Micro-features as applications of micromachining of nanocomposites. (a - b) SEM images of MTA/MWCNT micro-capacitors and micro-resistors made by precise 3D printing (Reprinted from [12] Copyright 2016, with permission from Society of Photo-Optical Instrumentation Engineers (SPIE)); (c) SEM image of a stainless steel/Al2O3 piston and a linkage rod made by soft molding (Reprinted from [22] Copyright 2009, with permission from Acta Materialia); (d) SEM image of a stainless steel/titania micro-gear made by soft molding (Reprinted from [23] Copyright 2009, with permission from Elsevier); (e) Optical image of a Epoxy/SiO2 wheel and (f) SEM image of a epoxy/SiO2 micro-gearwheel made by UV-LIGA methods (Reprinted from [229] Copyright 2009 , with permission Elsevier)

MTA: thiol-acrylate; MWCNT: multi-walled carbon nanotube; SWCNT: single-walled carbon nanotube; PP: polypropylene

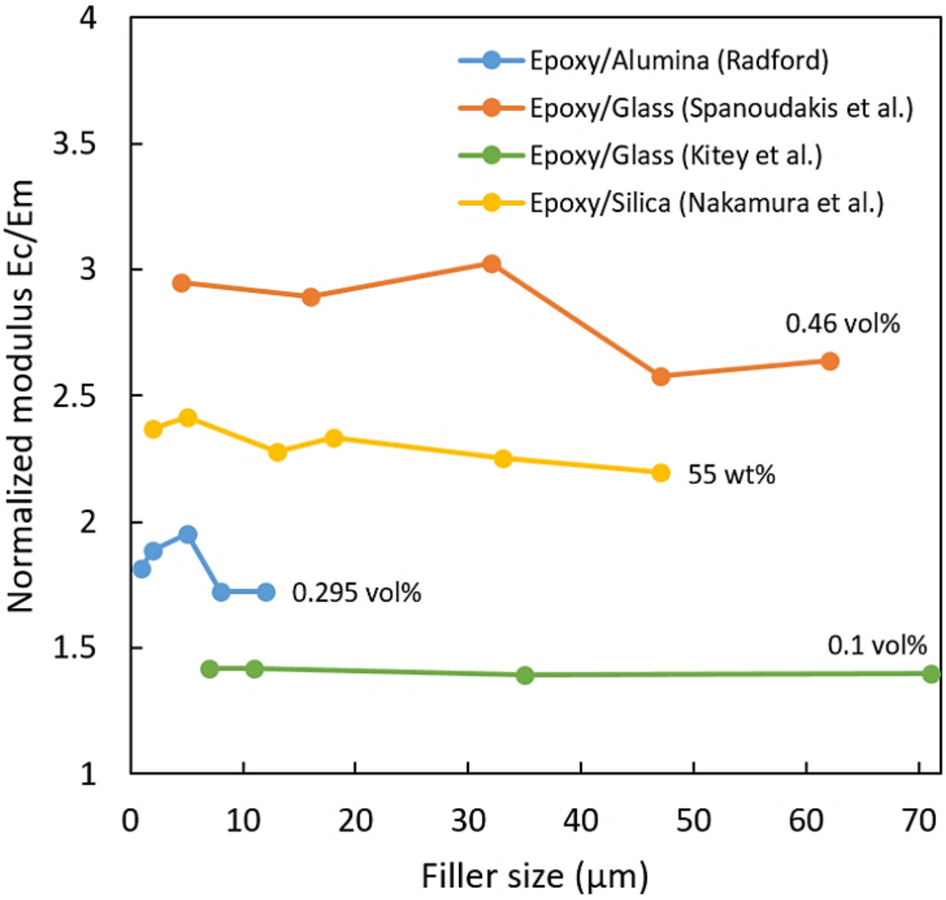


Figure 2: Normalized tensile modulus of epoxy-based composites as a function of micro-filler size (Adapted from [35-37], [230])

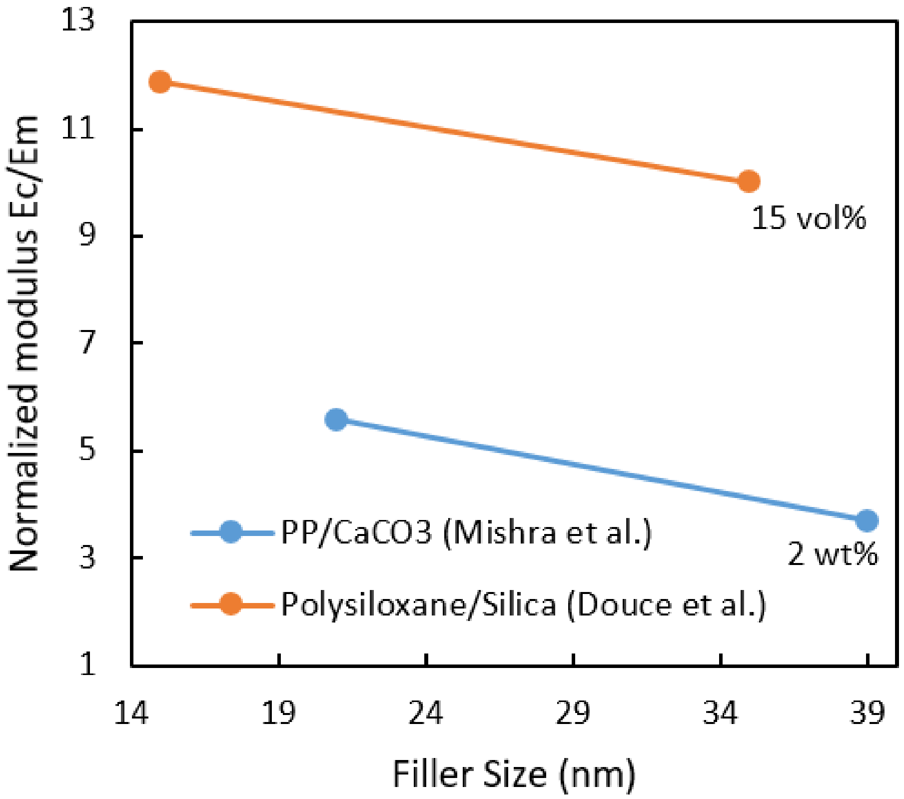


Figure 3: Normalized tensile modulus of polymer-based nanocomposites as a function of nano-filler size (Adapted from [231, 232])

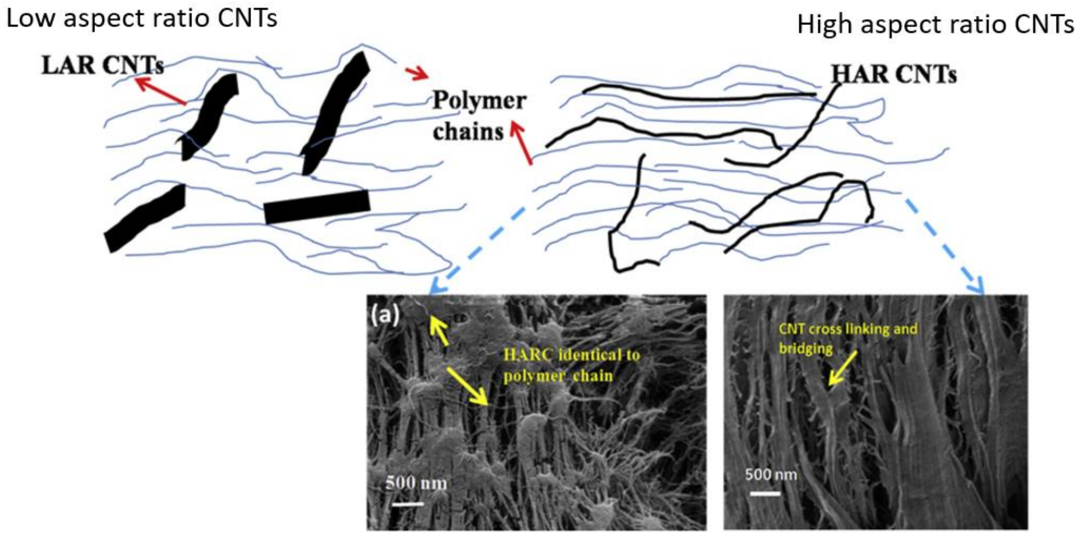


Figure 4: Graphical representation of the effect of CNT aspect ratio while interacting with polymer chains (Reprinted from [46] Copyright 2015, with permission from Elsevier)

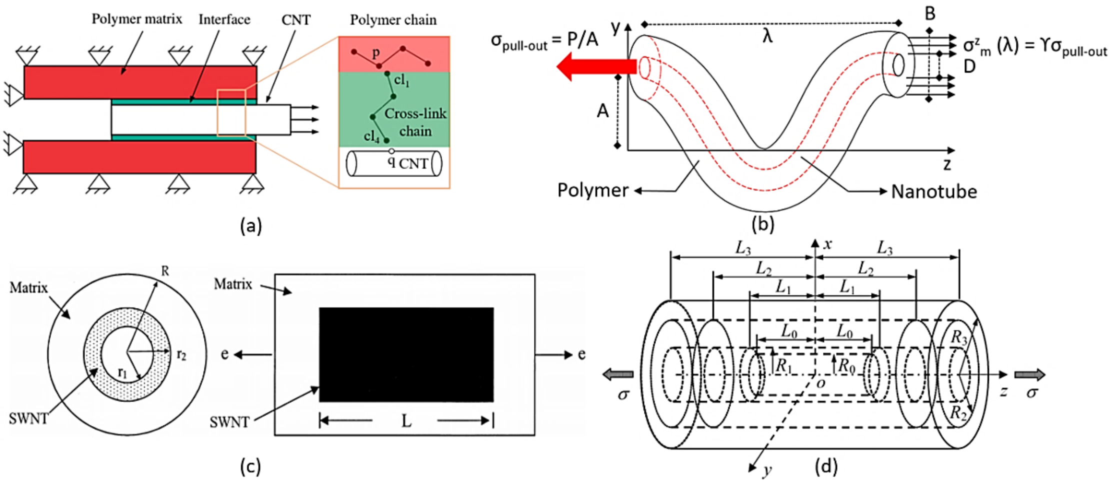


Figure 5: Theoretical models representing the micromechanical strengthening mechanism of polymer/CNT nanocomposites via the analysis of interfacial stress transfer with the consideration of various factors: (a) Effects of polymer matrix density, chemical cross-links and CNT defect (Reprinted from [47] Copyright 2006, with permission from Elsevier); (b) Effects of SWCNT waviness (Adapted with permission from [48] Copyright © 2011 Elsevier); (c) Effects of tube length and diameter (Reprinted from [49] Copyright 2004, with permission from Woodhead Publishing); (d) Effects of nanotube aspect ratio and fiber volume fraction (Reprinted from [54] Copyright 2006, with permission from Elsevier)

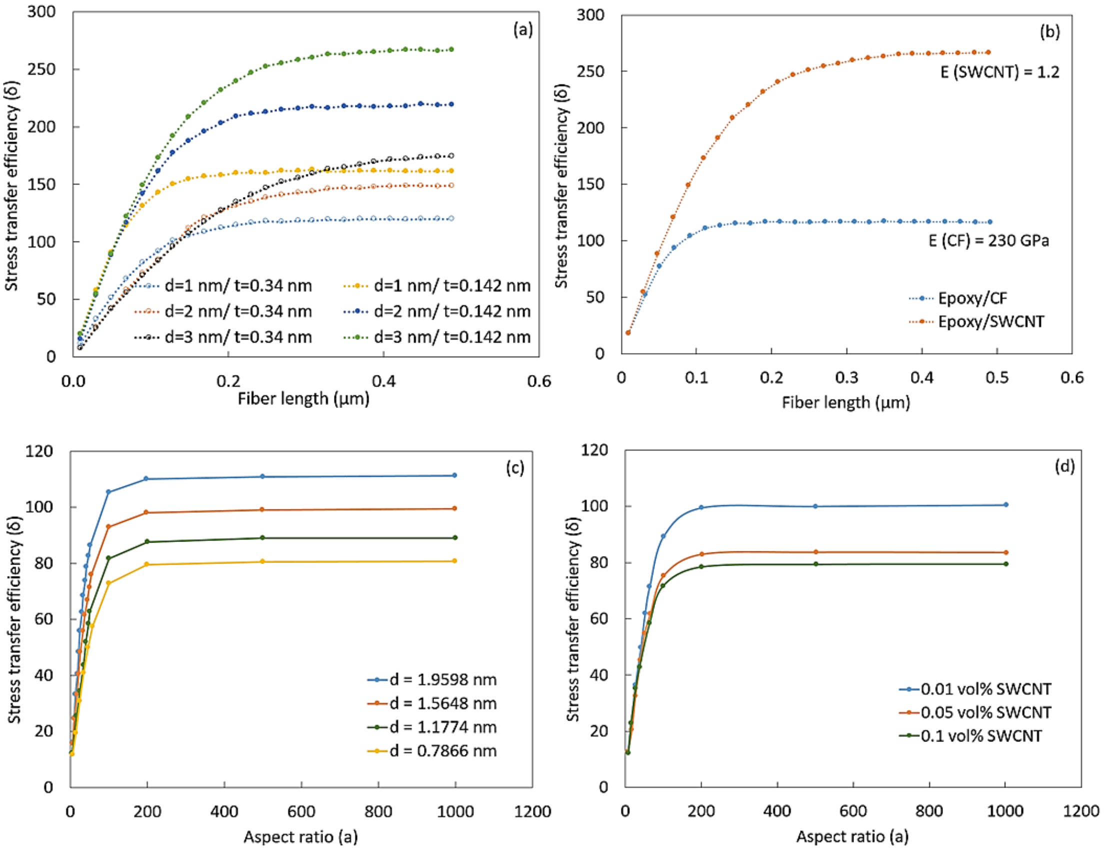


Figure 6: (a) Effect of SWCNT's diameter, thickness and length on the stress transfer efficiency (δ); (b) Effect of Young's modulus on the stress transfer efficiency (δ) (Assumed that CF had the same hollow structure with d= 3nm and t= 0.142 nm as SWCNT) (Fiber volume fraction $V_f$ ~ 0.17 % )(Reproduced from [49]); (c) Effect of nanotube diameter on stress transfer efficiency ($V_f$ = 0.1%); (d) Effect of nanotube volume fraction on stress transfer efficiency (d = 0.7086 nm) (Reproduced from [54])

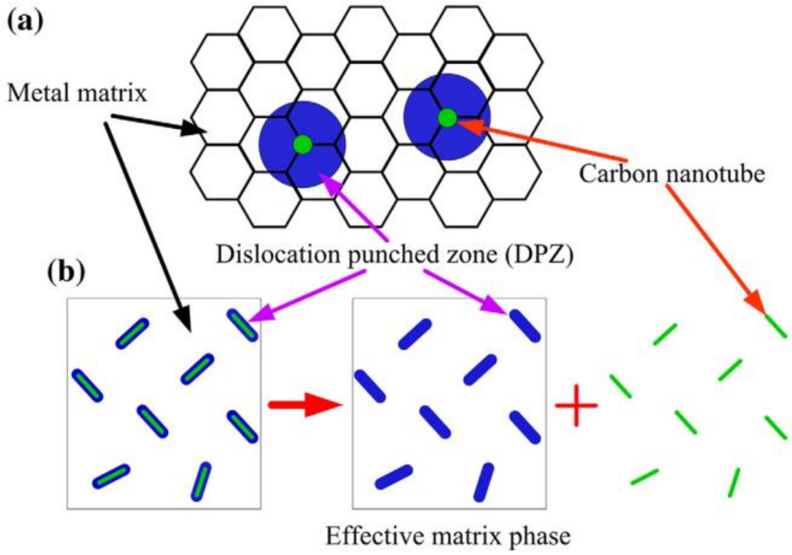
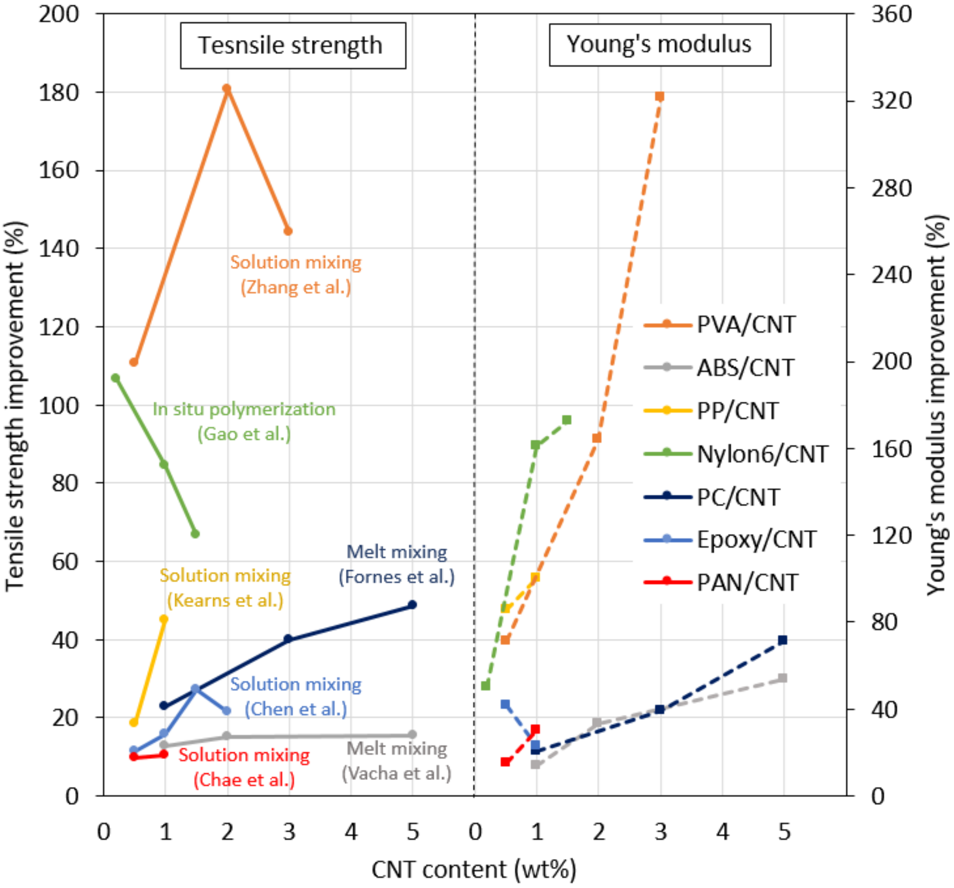


Figure 7: (a) Schematic of the microstructure of CNT-reinforced MMCs with DPZs. (b) The entire composite is decomposed into CNT and effective matrix phase, and the effective matrix is comprised of DPZs and pure metal matrix (Reprinted from [63] Copyright 2014, with permission from Elsevier)



(ABS: Acrylonitrile butadiene styrene; PAN: Poly-acrylonitrile; HDPE: High-density Polyethylene)

Figure 8: Mechanical properties of polymers reinforced CNTs with corresponding dispersion techniques (Adapted from [94, 96, 100, 233-236])

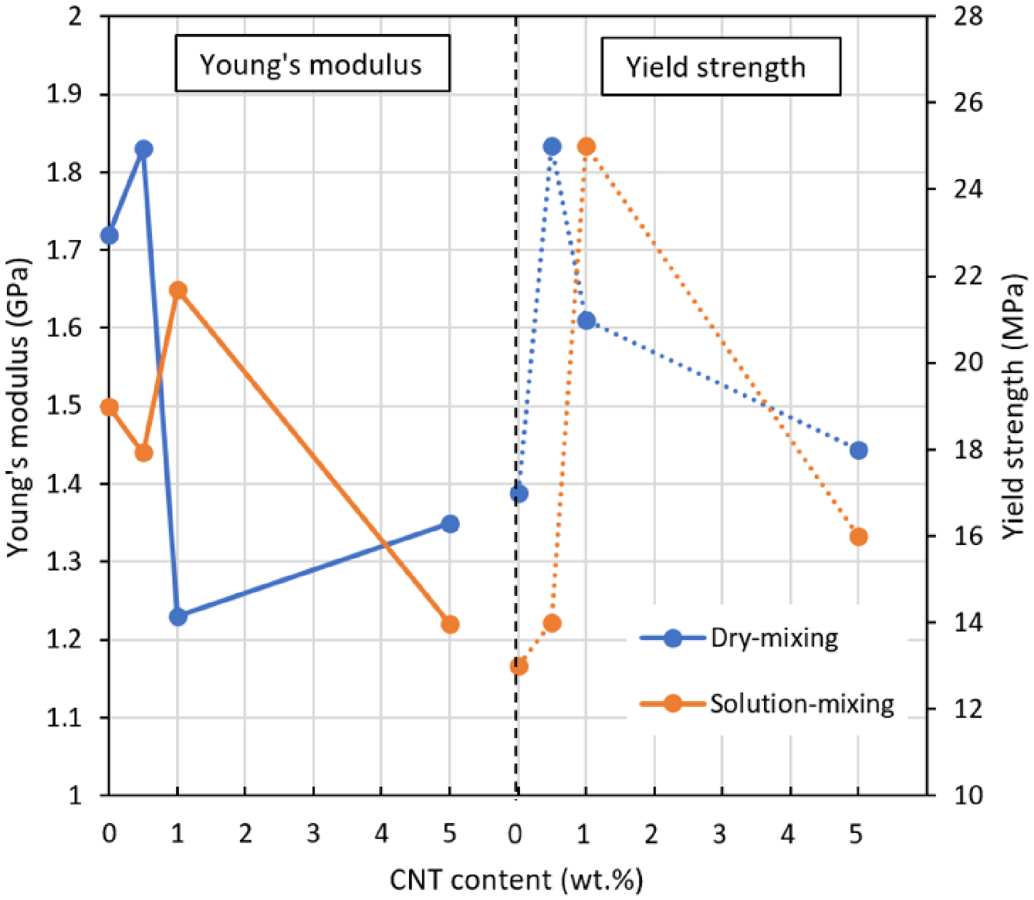


Figure 9: Effects of dispersion method on mechanical properties of CNT reinforced PMNCs (Reproduced from [98])

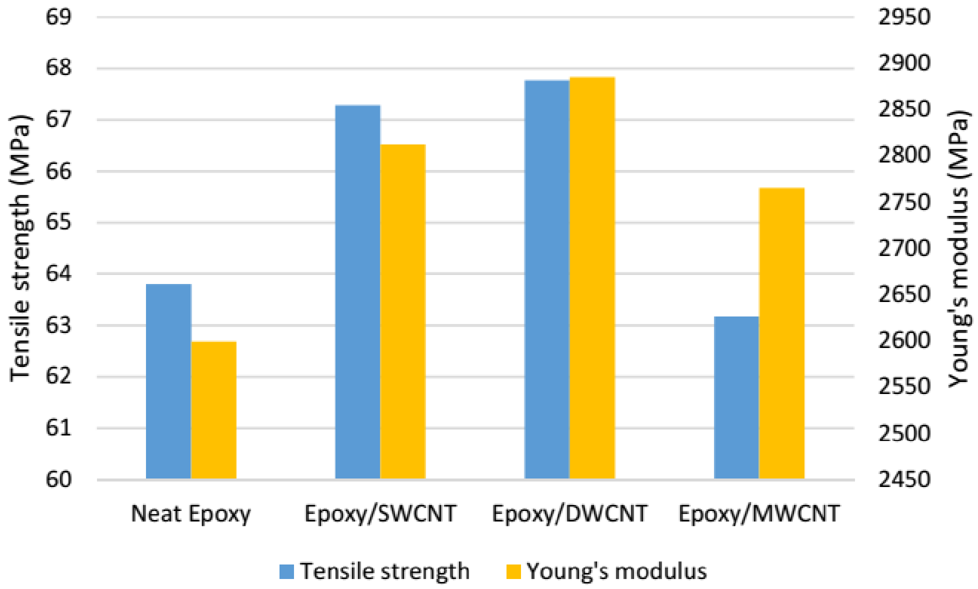


Figure 10: Effect of CNT structure on mechanical properties of polymer nanocomposites at a filler content of 0.3 wt.% (Adapted from [117])

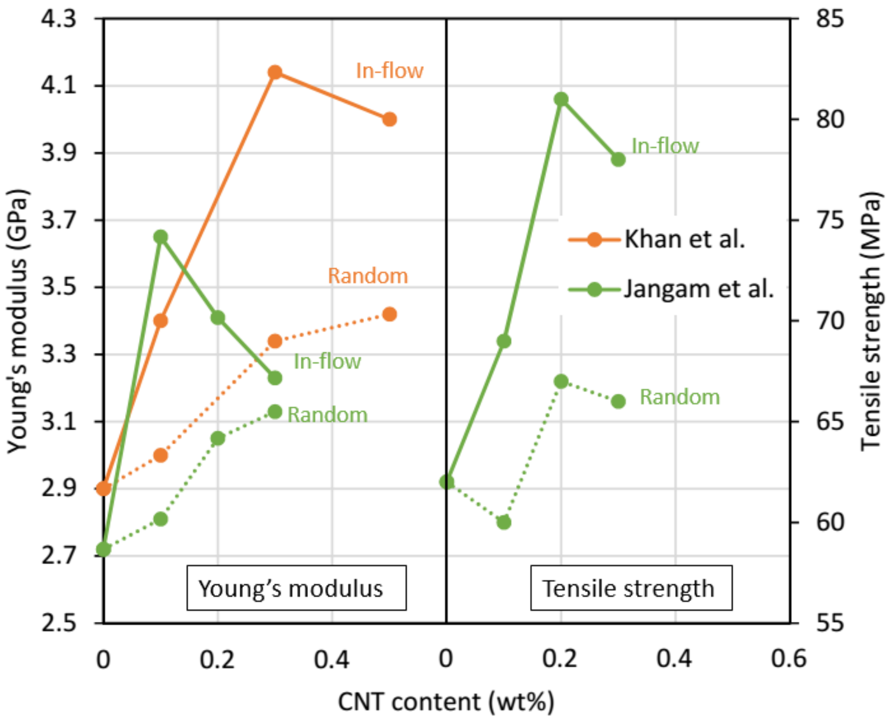
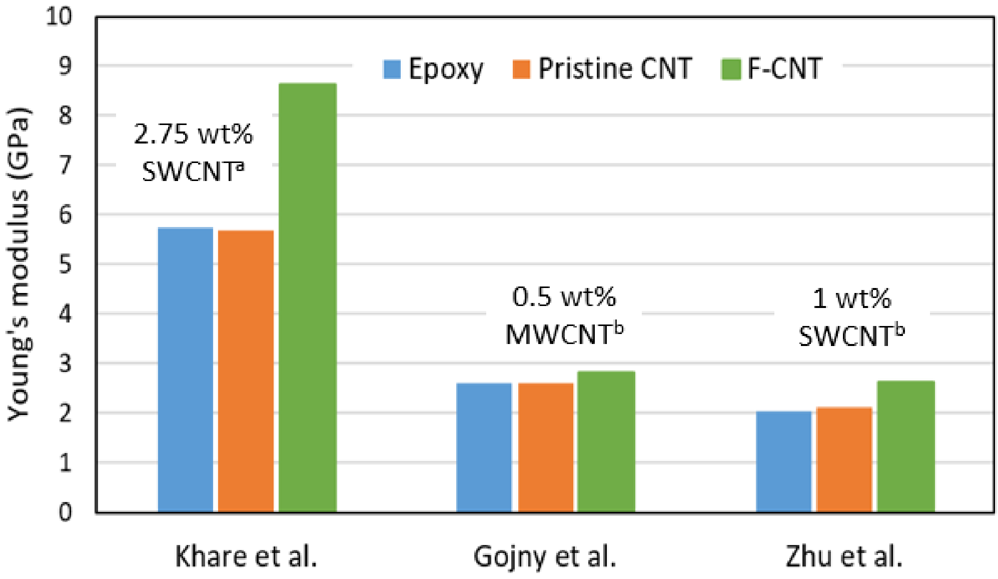


Figure 11: Effect of CNT alignment on mechanical properties of epoxy-based nanocomposites (Adapted from [237, 238])



[a] Simulation result ; [b] Experimental result

Figure 12: Effect of CNT functionalization on tensile modulus of CNT reinforced epoxy matrix nanocomposites (Adapted from [80, 108, 117])

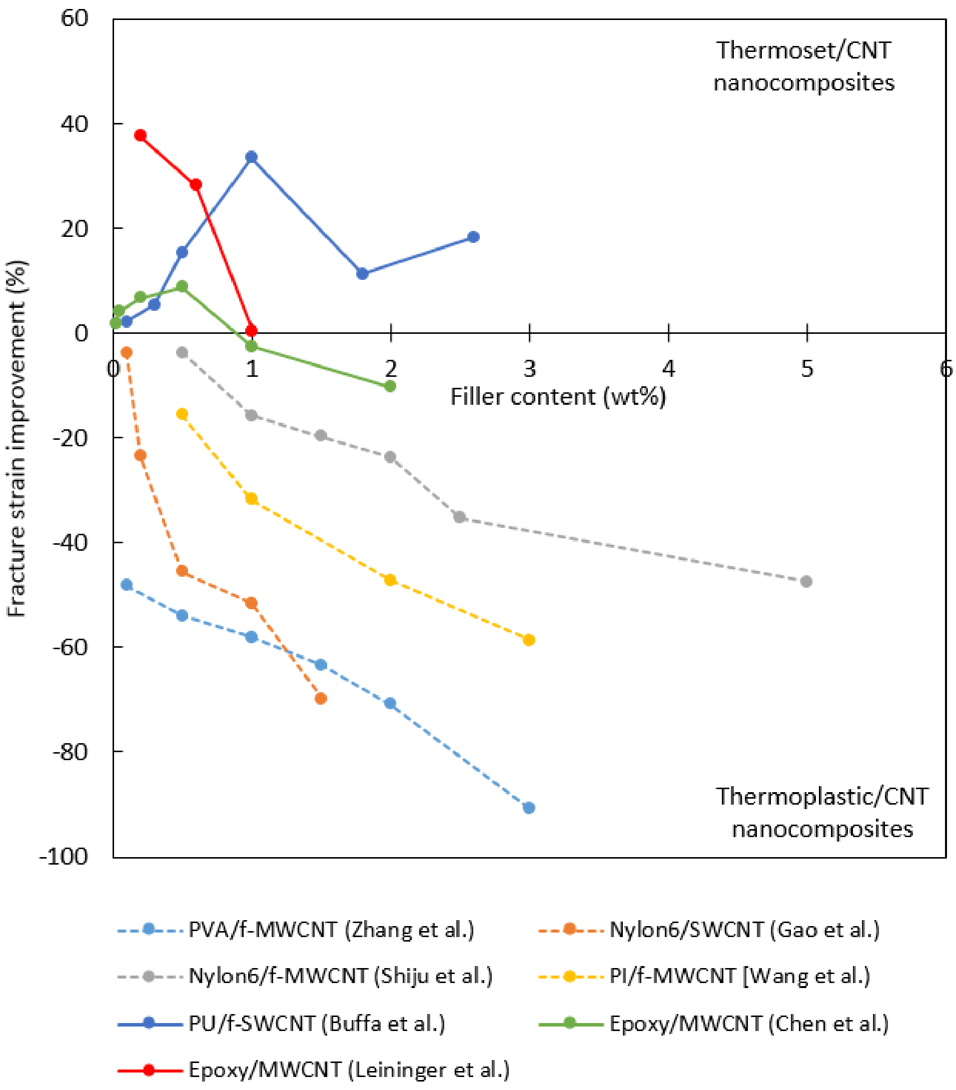


Figure 13: Fracture strain improvement of CNT based polymer nanocomposites as a function of filler content (Adapted from [94, 96, 109, 110, 239-241])

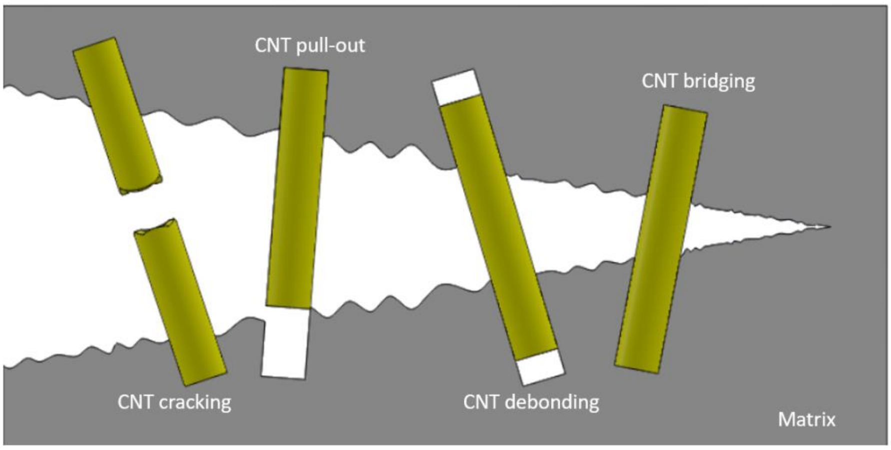


Figure 14: Schematic representing fracture toughening mechanism of CNT reinforced polymer nanocomposites (Reprinted from [113] Copyright 2017, with permission from Elsevier)

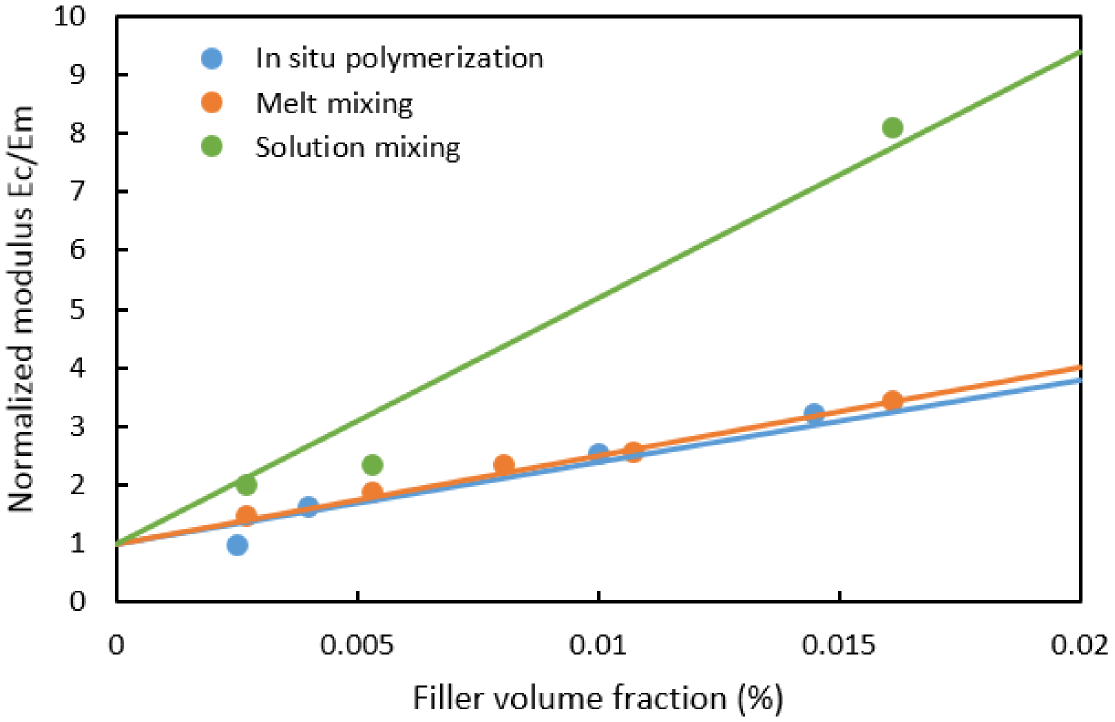


Figure 15: Normalized Young's moduli of TPU/graphene nanocomposites in different processing methods) (Adapted from [11])

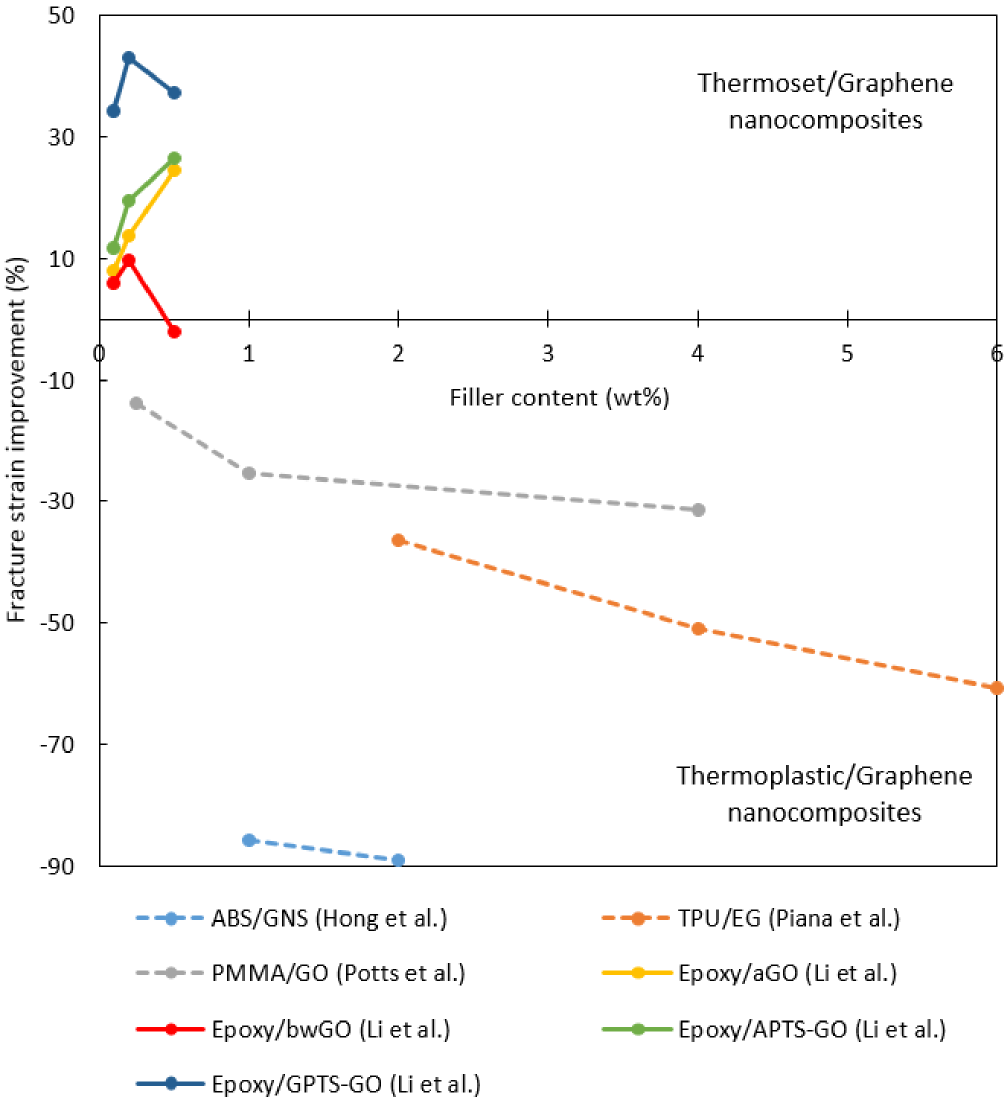


Figure 16: Fracture strain improvement of graphene-based PMNCs as a function of filler content (Adapted from [164, 242-244])

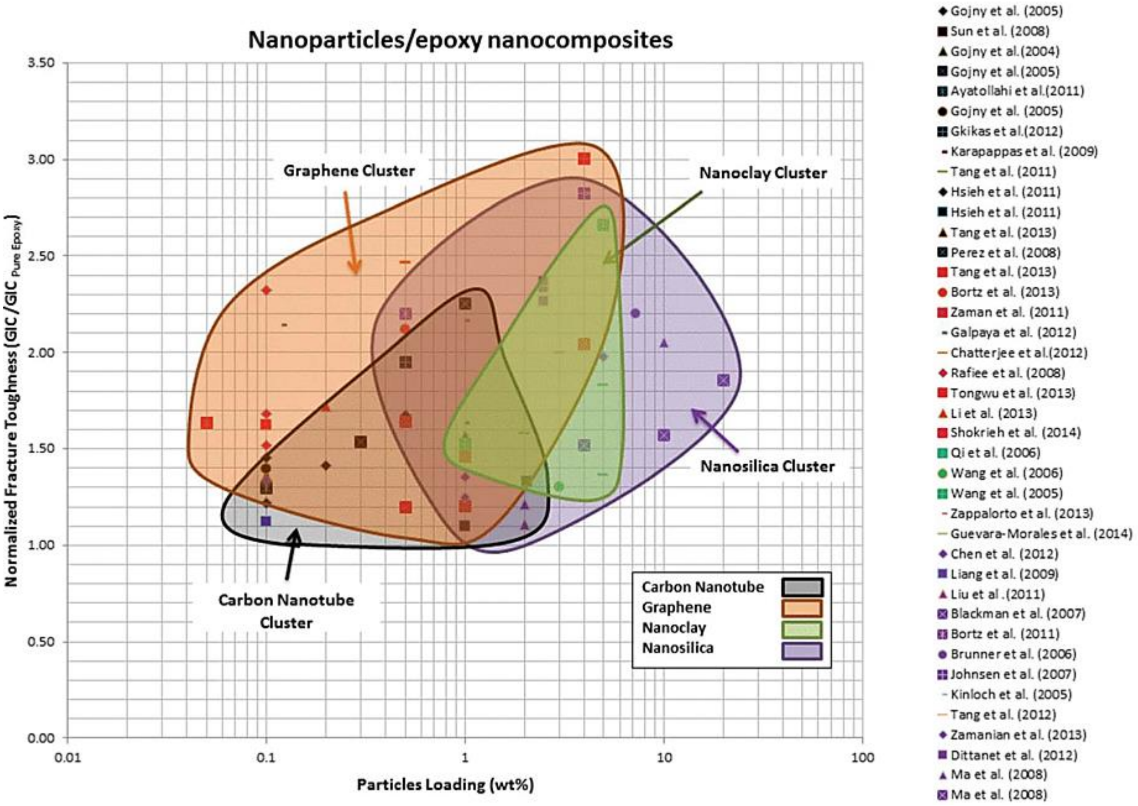


Figure 17: Comparative fracture toughness improvement of epoxy-based nanocomposites using different nano-fillers [165] (Open access from The Royal Society of Chemistry)

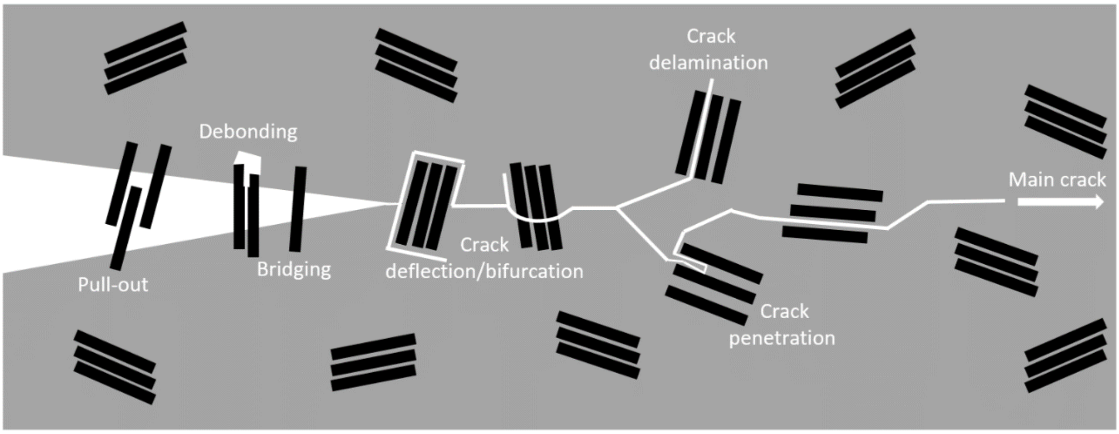


Figure 18: Schematic representing fracture toughening mechanism of CNT reinforced PMNCs (Adapted from [152] Copyright 2015 The Authors – Open access from Elsevier)

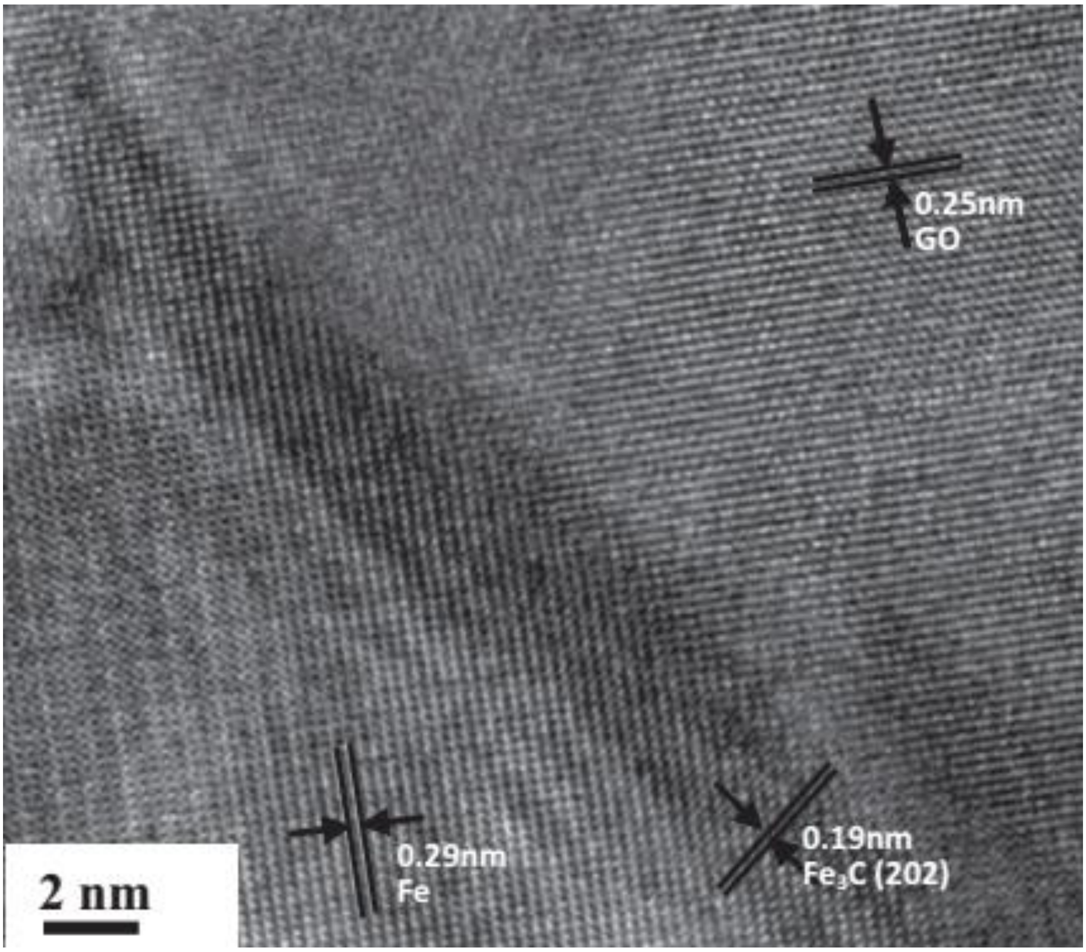


Figure 19: The formation of carbide when fabricating metal/GO (2 wt.%) nanocomposites (Reprinted from [180] Copyright 2014, with permission from Acta Materialia)

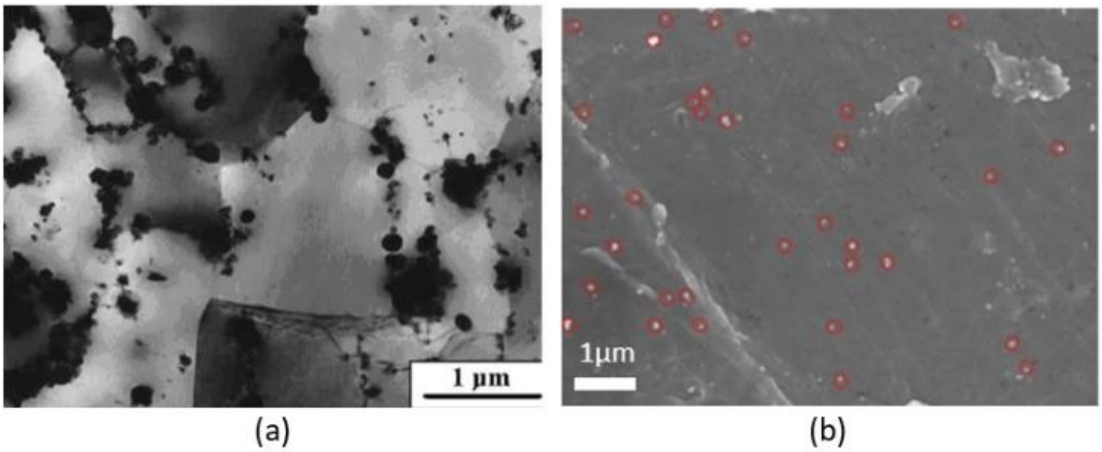


Figure 20: The agglomerations of ceramic nano-particles in Al matrices: (a) Al/$Al_2O_3$ 4 vol.% [217] and (b) Al/SiC 3 wt.% [216] (Open Access from Metals)

Table 1: Nanocomposite manufacturing techniques

| Nanocomposites | Manufacturing method | Ref. |
|---|---|---|
| Polymer matrix nanocomposites (PMNCs) | Melt mixing | [245] |
| | Solution mixing | [246] |
| | In situ polymerisation | [247] |
| | In situ formation | [248] |
| | Sol-Gel | [249] |
| Metal matrix nanocomposites (MMNCs) | Spray pyrolysis | [250] |
| | Infiltration | [251] |
| | Rapid solidification | [252] |
| | High energy ball milling and powder metallurgy (consolidation) | [253] |
| | Chemical vapour deposition | [254] |
| | Physical vapour deposition | [255] |
| | Colloidal suspension | [256] |
| | Sol-Gel and hot pressing | [257] |
| Ceramic matrix nanocomposites (CMNCs) | Powder processing (Compression, rolling, and extrusion) | [258] |
| | Polymer precursor | [259] |
| | Sol-Gel and consolidation | [260] |

Table 2: Nano-filler geometries

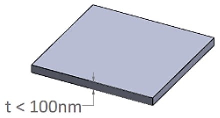
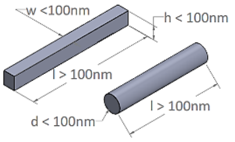
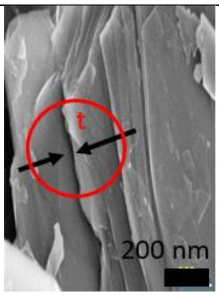
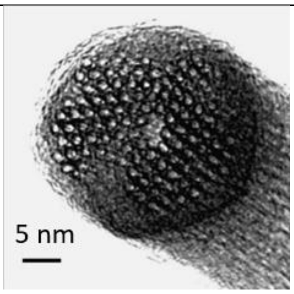
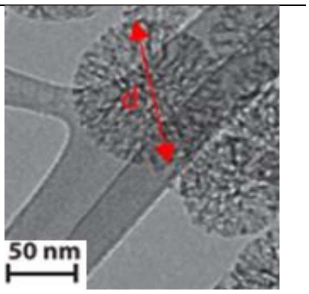
| 1D-Planar shape | 2D-Tubular shape | 3D- Spherical shape |
|---|---|---|
|  |  |  |
| at least one dimension ≤ 100 nm | at least two dimensions ≤ 100nm | all three dimensions ≤ 100nm |
|  |  |  |
| Graphene [261] | SWCNT (Reprinted with permissons from [262] Crown copyright © 2004) | Silica (Reprinted with permissons from [263] Copyright © 2016 Elsevier) |

Table 3: Fracture toughness of polymer reinforced CNT nanocomposites with the consideration of various factors

| Matrix | Factors | Variables | Filler content (wt.%) | Fracture toughness Improvement (%) | Ref. |
|---|---|---|---|---|---|
| Epoxy | - CNT structure<br>-Functionalization<br>- Filler content | SWCNT | 0.05 | 10.8 | [117] |
| | | | 0.1 | 23.0 | |
| | | | 0.3 | 12.3 | |
| | | DWCNT | 0.1 | 16.9 | |
| | | | 0.3 | 30.8 | |
| | | | 0.5 | 30.7 | |
| | | DWCNT-NH$_2$ | 0.1 | 18.5 | |
| | | | 0.3 | 41.5 | |
| | | | 0.5 | 43.0 | |
| | | MWCNT | 0.1 | 21.5 | |
| | | | 0.3 | 23.0 | |
| | | MWCNT | 0.1 | 24.6 | |
| | | | 0.3 | 30.8 | |
| | | | 0.5 | 29.2 | |
| Epoxy | - CNT diameter | Type A (20-30 nm) | 0.5 | 31.8 | [264] |
| | | Type B (30-40 nm) | | 26.4 | |
| | | Type C (40-50 nm) | | 24.2 | |
| | | Type D (50-60 nm) | | 21.2 | |
| Epoxy | - CNT length | 2091 nm | 0.1 | 46.6 | [265] |
| | | 1689 nm | | 43.3 | |
| | | 1332 nm | | 8.7 | |
| | | 992 nm | | 2.0 | |
| | | 503 nm | | -14.7 | |
| Epoxy | - CNT alignment<br>- Filler content | Random orientation | 0.05 | 6.2 | [237] |
| | | | 0.1 | 11.6 | |
| | | | 0.3 | 16.9 | |
| | | | 0.5 | 13.8 | |
| | | Aligned orientation | 0.05 | 23.3 | |
| | | | 0.1 | 36.1 | |
| | | | 0.3 | 53.0 | |
| | | | 0.5 | 31.1 | |
| Epoxy | | Three-Roll Milling | 0.1 | 58.3 | [266] |
| | | | 0.5 | 64.5 | |

| | | | | | |
|---|---|---|---|---|---|
| | -Processing technique | (TRM) | 1 | 68.5 | |
| | | | 1.5 | 60.0 | |
| | | | 2 | 57.7 | |
| | | HPH (High Pressure Homogenization) | 0.1 | 48.3 | |
| | | | 0.5 | 56.4 | |
| | | | 1 | 64.2 | |
| | | | 1.5 | 56.0 | |
| | | | 2 | 42.0 | |
| | | HPH + TRM | 0.1 | 82.6 | |
| | | | 0.5 | 72.5 | |
| | | | 1 | 78.5 | |
| | | | 1.5 | 78.0 | |
| | | | 2 | 74.0 | |

Table 4: Mechanical properties of CNT reinforced metal matrix nanocomposites

| Material | Tensile strength (MPa) | Young's modulus (GPa) | Hardness (GPa) | Elongation (%) | Reasons | Fabricating methods |
|---|---|---|---|---|---|---|
| Al/MWCNT 1wt.% [139] | 521.7 (35.7%) | 102.2 (41.3%) | 0.136 (30.8%) | 17.9 (-5%) | Good interfacial bonding, homogenous distribution, high elastic behavior of CNT, integrality of Al | Powder metallurgy cold isostatic press, hot extrusion |
| Al/MWCNT 0.5wt.% [138] | 130 (9.2%) | 60 (20%) | -- | 18.7 (-25.8%) | Good distribution of CNT due to rolling process | Mechanical mixing, hot rolling |
| Al/(Si) MWCNT 10wt.% [143] | 83.1 (4%) | 120.4 (78%) | -- | 8.8E-4 (-54%) | Strengthening by retained CNT and nano-crystalline structure | Thermal spray, plasma spraying (SPS) |
| Al/(Si) CNT 10wt.% [267] | -- | 125 (39%) | 2.1 (141%) | -- | Good distribution of CNTs due to Si-coating and dispersing method | Spark plasma sintering (SPS) |
| Al/(Ni) CNT 5wt.% [132] | 213 (52%) | -- | 3200 (113.3%) | -- | Homogenous dispersion, CNT structure retention, strong interfacial strength due to molecular mixing in in-situ CVD synthesis as compared to ball milling | Ball milling |
|  | 398 (184%) | -- | 6.5 (333%) | -- |  | In situ CVD synthesis |
| Cu/MWCNT 20 vol.% [137] | -- | 106.5 (108%) | 1.304 (72%) | -- | Homogenous distribution and reduction of MWCNT agglomerations | SPS and electroless deposition |
| Cu/MWCNT 10 vol.% [136] | 196 (45%) | 135 (95%) | 1.75 (207%) | -- | Homogenous distribution of CNT | SPS, cold rolling |
| Cu/(Ni) CNT 12 vol.% [268] | -- | -- | $21.5^a$ (111%) | -- | Highest mechanical reinforcement at 12 vol.% of CNT | Mechanical mixing and hot pressing |
| Cu/(Ni) MWCNT 0.75wt.% [140] | 279 (76%) | -- | 1.383 (54%) | -- | CNTs resist plastic deformation, thermal expansion mismatch, homogenous distribution, good interfacial bonding, high hardness of Ni | Ball milling and hot pressing |
| Cu/MWCNT 0.5 vol.% [269] | 307.4 (81.6%) | -- | 106 (14.7%) | 38.44 (55%) | Well dispersed of MWCNTs<br>Stable interface blocks dislocation | Ball milling and hot pressing |
| Ni-P/MWCNT 0.21wt.% [270] | -- | 665.9 (303%) | 28.9 (331%) | -- | Strengthening effects due to MWCNT presence | Electroless deposition |
| Ni/MWCNT 0.1wt.% [271] | 1140.7 (14.2%) | -- | 4.824 (74.5%) | 7.69 (-29.7%) | High quality dispersion and integrity of MWCNT due to surfactant treatment | SPS |

| | | | | | | |
|---|---|---|---|---|---|---|
| Mg-Zn/CNT<br>1 vol.% [142] | 321<br>(13.6%) | 52<br>(42.5%) | -- | 17<br>(-54.5%) | Homogenous, single distribution of CNTs, no reaction at interface, grain refinement | Melting and solidification |
| Mg/(Si)MWCNT<br>5 vol.% [134] | 296<br>(44.4%) | -- | 1.569<br>(100%) | 1.3<br>(-74%) | High wettability of MWCNTs due to Si-coating leads to good distribution, bonding strength | Ball mill and hot pressing |
| Mg/CNT<br>1.3wt.% [272] | 210<br>(9%) | -- | 46$^b$<br>(2.2%) | -- | The coefficient mismatch of thermal expansion and elastic modulus of Mg-CNT | Melt deposition and hot extrusion |
| Mg/(Ni) MWCNT<br>0.3wt.% [133] | 237<br>(38.6%) | -- | 0.54<br>(41%) | -- | Improved adhesion of Mg-(Ni)MWCNT due to Ni coating leading to $Mg_2Ni$ intermetallic formation, grain refinement | Powder metallurgy, microwave-assisted sintering |
| Mg alloy/MWCNT<br>2wt.% [273] | 297<br>(6.1%) | -- | 0.8<br>(0.37%) | 9.2<br>(-36%) | Strengthening mechanism, thermal mismatch of CNT-Mg alloy | Ball mill, hot compact |

$^a$: HRB; $^b$: HR15T

Table 5: Mechanical properties of graphene reinforced polymer matrix nanocomposites

| Material | Tensile Strength (MPa) | Young's modulus (GPa) | Reasons | Fabricating methods |
|---|---|---|---|---|
| EP/GNP 0.3 wt.% [274] | 64.4 (12.6%) | 2.16 (30%) | The high temperature in low viscosity system, low concentration of Gr leading to low agglomeration and uniform distribution of Graphene | Solution mixing |
| EP/APTS-GO 0.2 wt.% [244] | 81.2 (16%) | 3.3 (32%) | Uniformly distributed APTS-GO, strong interfacial stress | Solution mixing |
| EP/NH$_2$-GNP 4 wt.% [275] | 66 (1.5%) | 3.4 (17%) | GNP aggregation, poor GNP dispersion | Solution mixing |
| EP/EGS 3 wt.% [276] | 41 (20%) | 3.7 (25%) | High aspect ratio and uniformly distributed EGS, good interfacial adhesion of EP-EGS | Solution mixing |
| EP/RGO 0.2 wt.% [277] | 52.6 (-0.8%) | 3.1 (7%) | Weak interfacial bond of EP-RGO, RGO agglomeration, RGO curvature | Solution mixing |
| EP/GNP 0.1 wt.% [278] | 78 (40%) | 3.7 (31%) | The high specific surface area of GNPs, strong matrix-filler adhesion/interlocking due to wrinkled surface of GNPs | Solution mixing |
| EP/GNP 0.3 wt.% [279] | 70.4 (23%) | 1.28 (47%) | High effect of Sodium Dodecyl Sulphate (SDS) as solution on GNP dispersion, effect of surfactants | Solution mixing |
| ABS/CO(OH)$_2$/ GNS 4 wt.% [242] | 43.2 (50%) | -- | High modulus GNS, homogenous dispersion | Melt mixing |
| PMMA/GO 1 wt.% [243] | 70.9 (22%) | 4.39 (28%) | High aspect ratios and good dispersions of GO at low loading, wrinkled platelets | In situ polymerization |
| PC/FLG 1 wt.% [280] | 60 (10%) | 1.45 (26%) | Optimizing the aspect ratio of the graphene flakes | Solution mixing |
| HDPE/GNS 3 wt.% [281] | 47 (77%) | 2.033 (87%) | High specific area and flat-structure of GNS, the mechanical interlocking of HDPE-GNS, enlarging the interphase zone | Melt mixing |
| LDPE/RGO 5 wt.% [282] | -- | 10.1 (60.7%) | Homogeneous distribution of exfoliated carbon sheets in functionalized PE | Solution mixing/ melt compounding |
| TPU/EG 10 wt.% [164] | 9 (-84%) | 0.091 (237%) | Homogenous distribution, HS crystallization, EG hindering the amorphous phase, weak interfacial interaction of TPU-EG, EG agglomerations | Melt mixing |
| PVA/GNS 3 wt.% [162] | 43.2 (122%) | 1.186 (155%) | Homogenous distributed Gr and strong interfacial interaction of PVA-GNP. GNP restricting polymer chain movements | Solution mixing |

APTS-GO: amino-functionalized – graphene oxide; EGS: Graphene stack; FLG: Few layer graphene sheets; EG: Expanded graphite; GO: Graphene oxide; RGO: Reduced graphene oxide; GNP: Graphene nano-platelet; GNS: Graphene nano-sheet; GNP: Graphene nano-platelet

Table 6: Mechanical properties of graphene reinforced metal matrix nanocomposites

| Material | Tensile Strength (MPa) | Young's modulus (GPa) | Hardness (GPa) | Elongation (%) | Reasons | Fabricating methods |
|---|---|---|---|---|---|---|
| Al/RGO<br>0.3wt.%<br>[172] | -- | 90.1<br>(18%) | 1.59<br>(17%) | -- | High quality of RGO, uniform dispersed of RGO, strong interfacial bonding of Al-RGO | Compacting and hot pressing |
| Al-Mg-Cu/Gr<br>0.3wt.%<br>[283] | 454<br>(25%) | 72<br>(-1%) | -- | 11.8<br>(7%) | Evenly distributed of Gr, good interfacial bonding, Gr structure retention | Ball milling, hot isostatic pressing (HIP) and extruding |
| Al/GO<br>15 mg ml$^{-1}$<br>[284] | 192<br>(-7%) | -- | -- | 28<br>(50%) | Dynamic recrystallization of matrix phase due to shear deformation, heat from FSP and grain refinement | Friction stir processing (FSP) |
| Al/GNS<br>1 wt.%<br>[285] | 248<br>(68.7%) | -- | -- | 8.3<br>(-52%) | Homogenous distribution of GNFs, no metallurgical at interfaces of Al-GNF | Blending, cryo-milling, degassing and hot extrusion |
| Cu/FLG<br>3 wt.%<br>[286] | -- | -- | 0.46<br>(39%) | 16<br>(-71%) | Disruption of Gr layers due to Cu deformation by rolling, Gr partly hinder grain growth | Rolling |
| Cu/RGO<br>2.5 vol.%<br>[184] | 335<br>(30%) | 131<br>(30%) | -- | -- | Strong interfacial bonding of Cu-RGO | Compacting and SPS |
| Cu/GO<br>0.5 gL$^{-1}$<br>[287] | -- | 137<br>(30%) | 2.5<br>(96%) | -- | Uniform distribution of Gr, grain size refinement | Pulse reverse electrodeposition |
| Cu/(Ni)GPL<br>0.8 vol.%<br>[288] | 245<br>(42%) | -- | -- | 9<br>(-67.8%) | Good dispersion of GPL and strong interfacial bonding Cu-GPL (Ni), covalent interaction of Ni-GPL | Solution, sonication, SPS |
| Cu/(Ni)GNS<br>1 vol.% c | 320<br>39.1%) | 132<br>(61%) | -- | -- | Homogenous dispersion of GPL and strong interfacial interaction of Cu-GPL due to Ni coating | Solution, sonication, SPS |
| Cu/GNP<br>1.3 wt.%<br>[289] | 485<br>(107%) | 104<br>(21%) | -- | 9<br>(-64%) | Homogenous dispersion of GNPs due to pre-coating of Cu on GNPs | Electroless plating, SPS tensile |
| Cu/RGO<br>0.3 wt.%<br>[290] | 308<br>(41%) | 109<br>(12%) | -- | -- | Less structural damage on RGO, randomly oriented of RGO benefit the load transfer of nanocomposite | Hot pressing |
| Ni/GO<br>0.12 wt.%<br>[174] | -- | 252.76<br>(51.6%) | 6.85<br>(278.4%) | -- | Compact interfacial bonding of Ni-GO, homogenous dispersion of GO | Electrodeposition |
| Ni/Gr<br>0.05 gL$^{-1}$<br>[179] | -- | 240<br>(70%) | 4.6<br>(20%) | -- | High interaction of Ni-Gr, Gr preventing Ni dislocation | Electrochemical deposition |
| Mg/Ti/GNP<br>0.18 wt.%<br>[188] | 230<br>(8.5%) | -- | -- | 14<br>(27%) | High specific area and adhesion of Gr | Semi powder metallurgy |
| Mg-Al-Sn/<br>GNP 0.18 wt.%<br>[291] | 269<br>(14%) | -- | -- | 10.9<br>(-34.7%) | High specific surface area, superior nano-filler adhesion and two-dimensional structure of GNPs | Semi-powder metallurgy |

| | | | | | | |
|---|---|---|---|---|---|---|
| Mg/GNP 0.3 wt.% [173] | 246 (32.2%) | 13.84 (131%) | 55HV (34%) | 16.9 (74.2%) | High specific surface area of GNPs, uniform dispersion | Compaction, sintering, extruding |

Table 7: Mechanical properties of ceramic nano-fillers reinforced polymer matrix nanocomposites

| Material | Tensile Strength (MPa) | Young's modulus (GPa) | Reasons | Fabricating methods |
|---|---|---|---|---|
| PP/MMT 15 wt.% [292] | 38 (13%) | 3.36 (100%) | Well-dispersed MMTs | Melt mixing, injection molding |
| PP/MMT 10 wt.% [199] | 41.6 (38.7%) | -- | The intercalation of MMT layers in the matrix due to the presence of MAPP compatibilization | Melt mixing, injection molding |
| PP/MMT 2 wt.% [203] | 32 (18%) | 0.9 (82%) | The persistence of silicate layers, modification of PP structure, partially immobilized polymer segments | Melt mixing |
| PA6/MMT 7.2 wt.% [204] | -- | 5.7 (107%) | High molecular weight and integrity of matrix | Melt mixing |
| SPU/30B 7 wt.% [205] | 21 (-53.3%) | 0.024 (220%) | Well-dispersed and delaminated of Cloisite 30B regardless processing methods, good interaction of PU-clay, preferable solution mixing due to PU and surfactant degradations from melt mixing, larger improvements of the stiffness of SPU due to higher fraction of soft segment | Solution mixing<br>Melt mixing |
| HPU/30B 7 wt.% [205] | 34 (-41.3%) | 0.134 (168%) | | |
| SPU/30B 7 wt.% [205] | 7 (-66.7%) | 0.0193 (168%) | | |
| HPU/30B 7 wt.% [205] | 15 (-66%) | 0.119 (95%) | | |
| EVA/ Cloisite Na 3 wt.% [201] | 25.9 (-8.8%) | 0.0135 (10.7%) | The dominant effect of exfoliation on tensile properties of nanocomposites, high interaction between EVA and Cloisite 30B | Melt mixing, compression molding |
| EVA/ Cloisite 20A 3 wt.% [201] | 25.8 (-9.2%) | 0.0249 (104.1%) | | |
| EVA/ Cloisite 25A 3 wt.% [201] | 26.2 (-7.7%) | 0.022 (80.3%) | | |
| EVA/ Cloisite 30B 3 wt.% [201] | 30.7 (8.1%) | 0.0228 (86.9%) | | |
| EVA/ Nanofile 757 3 wt.% [201] | 27.6 (-2.8%) | 0.0116 (-4.9%) | | |
| EVA/ Nanofile 15 3 wt.% [201] | 26.7 (-6%) | 0.024 (96.7%) | | |
| EVA/ Somasif ME100 3 wt.% [201] | 24.5 (-13.7%) | 0.0124 (1.6%) | | |
| EVA/ Somasif MAE 3 wt.% [201] | 25.1 (-11.6%) | 0.021 (72.1%) | | |
| EP/SMC 2 wt.% [200] | 57 (25%) | 3.3 (10%) | Homogenous distribution and high exfoliation of SMCs | Solution mixing |
| EP/$SiO_2$ 3.72 vol.% [206] | -- | 3.96 (12%) | Less agglomeration, well dispersed and high aspect ratio of Silica at low loadings | Solution mixing |
| EP/$SiO_2$ 4 wt.% [293] | 42.3 (30.57%) | -- | Optimal interaction of EP-SiO2 at 4 wt.% | Solution mixing |
| EP/$SiO_2$ 13.4 vol.% [207] | -- | 3.85 (30%) | High modulus, well dispersion and no agglomeration of Silica | Sol-gel mixing |
| EP/$SiO_2$ 20 wt.% [294] | -- | 3.97 (26.4%) | High stiffness of silica | Solution mixing |
| EP/$SiO_2$ 20.2 wt.% [295] | -- | 3.85 (30%) | Well-dispersed silica | Solution mixing |

MMT: montmorillonite; SMC: Silane-modified clay; SPU: Soft polyurethane; HPU: Hard polyurethane; EVA: Ethylene-vinyl acetate

Table 8: Mechanical properties of ceramic nano-particle reinforced metal matrix nanocomposites

| Material | Hardness (GPa) | Tensile strength (MPa) | Yield strength (MPa) | Elongation (%) | Reasons | Fabricating methods |
|---|---|---|---|---|---|---|
| Al/SiC 3 wt.% [216] | 0.55 (39.5%) | 164.4 (29.3%) | 145 (31%) | 165.3 (-32.8%) | Homogenous distribution of SiC, effects of Orowan strengthening, grain size refinement (yield strength), mismatch of thermal expansion coefficients between Al and SiC (CTE). Porosities lead to a reduction of ductility. SiC hindered metal matrix dislocation that increasing hardness | Powder metallurgy (PM), ultrasound-assisted stirring and planetary agitation, hot compressing |
| Al/SiC 6.5 vol.% [219] | -- | 807 (26.5%) | 610.8 (9.5%) | 59 (-91.8%) | Homogenous dispersed SiC, grain size refinement, Orowan strengthening | Mechanical alloying, cryo-milling, HIP consolidation |
| Al/SiC 1.25 vol.% [222] | -- (69%) | -- (109%) | -- | 3.6 (-40%) | Homogenous distribution of SiC and CTE strengthening at low loading, residual stresses, high dislocation density and grain size reduction increased hardness, agglomeration and porosity at a high loading of SiC. Hard particles reduced ductility. | Mechanical alloying, ultra-sonication |
| Al/SiC 2 wt.% [228] | -- | 301 (12%) | 156.3 (43.4%) | 3.4 (26%) | Uniformly dispersed SiC, CTE, high dislocation density, restriction of dislocations | Mechanical alloying, ultra-sonication |
| $Al/Al_2O_3$ 4 vol.% [217] | 66.6 (109.5%) | 245.5 (67%) | 183.7 (138.6%) | 12.8 (-51.7%) | Evenly distributed Al2O3 Orowan strengthening and grain size refinement effects at below 4 vol.% of loading | PM, wet mixing, cold isotropic pressing (CIP) and sintering |
| Al alloy/SiC 10 wt.% [227] | 87.2 BHN (9%) | 265 (12.3%) | 257 (16.8%) | 18.2 (-6.2%) | The composite density was reduced, SiC hindered alloy dislocations, uniform distribution of SiC and their high wettability due to Mg coating | Stir casting |
| Al alloy/SiC 10 wt.% [226] | 78 BHN (28%) | 188 (0.53%) | -- | 2.4 (-22.6%) | Insufficiently homogenous distribution of SiC due to improper stirring led to the low improvement of tensile strength, hard | PM, mechanical mixing |

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|---|---|---|---|---|---|---|
| | | | | | SiC hindered dislocations that contributed to hardness improvement | |
| Al alloy/SiC 6 wt.% [296] | 116 BHN (62%) | 267 (29%) | -- | 3.6 (-41%) | SiC possessed advanced hardness, strong interface of Al alloy - SiC | Liquid metallurgy |
| Mg alloy/SiC 1.5 wt.% [224] | -- | 199.3 (90%) | 71.7 (73%) | 20 (135%) | Grain size refinement, strong bond of Mg/4Zn-SiC, SiC clusters, no reason for ductility improvement | MA, ultrasonic cavitation |
| Mg/SiC 3 vol.% [225] | 0.58 (180%) | 288 (21.5%) | -- | 6 (-40%) | Grain size refinement, dispersion hardening, strain hardening due to extrusion, dislocation hindering | PM, ball milling |
| Mg/SiC 1.84 wt.% [223] | 0.42 (10%) | 203 (18%) | 157 (25.6%) | 7.6 (31%) | Hard SiC led to a high constraint of matrix deformation that improved hardness, dislocation density, internal stresses formation due to thermal expansion mismatch, Orowan strengthening and the mismatch of elastic reinforcing phase - plastic matrix phase contributed to tensile properties increments, the activation of non-basal slip and SiC-Mg interfacial integrity led to ductility improvement | MA, pressing, microwave hot sintering |
| Mg/$SiO_2$ 10 vol.% [297] | 1.03 (75%) | 251 (32%) | 225 (60.7%) | 4 (-69%) | Uniform distribution of SiO2, grain size refinement, Mg2Si, MgO formations during FSP | FSP |
| Al/TiC 10 wt.% [221] | 44.17 HV5 (143.6%) | -- | 189 (136%) | 5 (-83.3%) | Strong TiC particles, grain size refinement | In situ |