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Mechanical and tribological properties of self-lubricating metal matrix nanocomposites reinforced by carbon nanotubes (CNTs) and graphene – A review

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Abstract

Rapid innovation in nanotechnology in recent years enabled development of advanced metal matrix nanocomposites for structural engineering and functional devices. Carbonous materials, such as graphite, carbon nanotubes (CNT's), and graphene possess unique electrical, mechanical, and thermal properties. Owing to their lubricious nature, these carbonous materials have attracted researchers to synthesize lightweight self-lubricating metal matrix nanocomposites with superior mechanical and tribological properties for several applications in automotive and aerospace industries. This review focuses on the recent development in mechanical and tribological behavior of self-lubricating metallic nanocomposites reinforced by carbonous nanomaterials such as CNT and graphene. The review includes development of self-lubricating nanocomposites, related issues in their processing, their characterization, and investigation of their tribological behavior. The results reveal that adding CNT and graphene to metals decreases both coefficient of friction and wear rate as well as increases the tensile strength. The mechanisms involved for the improved mechanical and tribological behavior is discussed.

Keywords: A. Metal Matrix Composites (MMCs); B. Wear; C. Mechanical properties; D. Powder processing; E. Self-Lubricating

1. Introduction

Composite materials are engineered or naturally occurring materials which contain two or more distinct constituents with significantly different chemical, physical and mechanical properties. The metal matrix composites (MMCs) reinforced by ceramic particles or fibers generally have superior properties, such as high specific strength and modulus compared to unreinforced alloys. In general, composites have the combination of properties of constituents, such as they inherit ductility and toughness of the matrix and high modulus and strength of the reinforcements [1]. In this regard, the applications of MMCs have been extended to use as structural materials in aerospace, automotive, marine and defense industries [2].

In MMCs, different metals or alloys of aluminum, magnesium, copper, or nickel are generally used as matrix materials. Among these matrix materials, aluminum alloys are the most widely used materials, both in research and industrial viewpoints [3, 4]. This is due to their outstanding properties, such as light weight, high strength, high specific modulus, low thermal expansion coefficient, and good wear resistance [5]. Depending on final desired properties of composites, different reinforcements are

used as reinforcement in aluminum matrix composites. Al_2O_3 , SiC, B_4C is among the very common particles which are used as reinforcement in aluminum matrix composites.

There are different methods to synthesize particle reinforced MMCs. Generally, these methods are classified into: (1) Solid state processing; (2) Liquid state processing; and (3) Vapor deposition. Two common methods which are applied to fabricate aluminum matrix composites reinforced by particulate reinforcements in large scales are powder metallurgy method (solid state) and casting (liquid state). In literature, a considerable amount of work has been devoted to producing aluminum matrix composites by using powder metallurgy and to study their mechanical properties [6-10]. The basic route in the powder metallurgy (P/M) technique, in which all materials remain in the solid state, is mixing the powders, compacting, and sintering of the compacted part to achieve the least possible porosities and the highest possible density. In liquid state methods, the matrix is in the state of a liquid. There are different methods of liquid state processing to produce metal matrix composites. These methods include stir casting [11], pressure infiltration [12], pressureless infiltration [13] and squeeze casting [14]. Ünlü et al. [15] investigated and compared the mechanical properties of aluminum matrix composites reinforced by Al_2O_3 and SiC which are produced both by powder metallurgy method and casting. The results show that the mechanical and tribological properties of composite which are produced by casting are higher than P/M method. In addition, casting is also a more industrial compatible technique to produce composites in large scales.

Among various reinforcements, recent emerging material, carbonous materials, is found to have many favorable attributes such as high thermal conductivity, low coefficient of thermal expansion, high damping capacity and good self-lubricant property[4]. The considerable amount of research has been made to study the influences of embedding graphite particles into the metal matrix on the tribological properties of aluminum alloys [16-18]. Metal matrix composites embedded by graphite or carbon fibers have self-lubricating behavior since graphite act as a solid lubricant [19]. In this regard, solid lubricant as reinforcement tends to decrease the friction coefficient of MMCs and improve tribological properties of self-lubricating composite compared to composites reinforced by ceramic particles. The graphite size, which is commonly used in MMCs fabrication and obtaining desired mechanical and self-lubricating properties are in the micron range [1, 4, 20-25].

Generally, size of reinforcement influences the mechanical properties such as strength, ductility and fracture of self-lubricating MMCs. By increasing the reinforcement size, tensile strength and ductility decrease simultaneously. MMCs reinforced by larger particles are susceptible to formation of defects such as cracking during mechanical testing which results in a premature failure of the composites. Therefore, it is expected to have superior properties when the reinforcement size is in the nano range. It is expected that by reducing the particle size in MMC's to the range of nanosize, some of the limitations such as poor ductility and elongation, poor machinability, and reduced fracture toughness of MMCs can be solved. The reason for significantly improved mechanical properties is due to the combined effect of Orowan strengthening and grain refining mechanisms and high temperature creep resistance properties could make metal matrix nanocomposites (MMNCs) very attractive, especially when lightweight metals such as Al or Mg are used as the matrix material. The strengthening mechanisms which are involved in enhancing mechanical and tribological properties of nano particle reinforced metal matrix composites have been discussed carefully elsewhere [4, 26]. To briefly discuss, there are a few common different mechanisms that have been suggested to enable increased strength in metal composites; (1) Orowan strengthening from dislocation bowing by reinforced particles, (2) Hall–Petch strengthening from grain refinement, (3)

Forest strengthening resulting from the Coefficient of Thermal Expansion (CTE) mismatch between matrix and particles, and (4) Taylor strengthening by modulus mismatch between matrix and particles. MMNCs will benefit from the Orowan mechanism only if a dispersed second phase of nanometer size could be attained. Hall–Petch strengthening will in general one of the mostly active mechanisms which improve strength of MMNCs by incorporation of nano particles. Usually, the addition of nano particles refines grains or confines grain growth. CTE and modulus mismatch are considered to be negligible when compared with Orowan and grain refinement in many recent observations [4, 23, 26, 27].

Theoretically, it has been shown that the nano sized particles can result in dramatic improvements in light of the very low proportion to the quantities they are added. However, the strength achieved in most solidification processed and powder metallurgy nanocomposites are still below the conventional monolithic alloys, which can be caused by possible nanoparticles agglomeration, in capability of solidification front in capturing nanoparticles (pushing off by growing dendrites), and porosity. Therefore, before these materials can be introduced as an excellent candidate for conventional metals and alloys, these challenges in synthesis and processing need to overcome. Gravity casting techniques are prone to porosity, which could be one cause for low yield strength in nanocomposites. However, techniques such as Squeeze Casting can eliminate porosity. Another factor is the degree to which the particles agglomerate and fail to achieve uniform dispersion. As far as ultrasonic dispersion is concerned, de-agglomeration depends on how much power can be transmitted to the melt. Therefore, processing technique is one of the factors which need to be considered in pursuing advances in MMNC's [4].

Recently, research has been focused on nano-sized carbonous materials, such as carbon nanotubes (CNTs) and nano-graphite or graphene [1] in order to attain enhanced mechanical, electrical, and tribological properties. For instance, carbon nanotubes have been demonstrated to exhibit ultrahigh strength and modulus, and also have anisotropic electrical conductivity; when included in a matrix, they could pass on significant property improvements to the resulting nanocomposites. Thus, the application of nanotechnology to materials science and engineering opens up new opportunity and research direction for the development of novel smart metal matrix nanocomposites. Carbon nanotubes and graphene possess exceptional mechanical strength as well as excellent electrical and thermal conductivities, and their incorporation in metallic matrices leads to composites with higher mechanical, electrical, and magnetic properties. This has been led to an increasing interest in incorporating carbon nanotubes and graphene in MMCs to be the most effective reinforcing fillers in synthesizing self-lubricating composites for structural engineering and functional devices [28-30]. Table 1 summarizes a list of research on MMCs containing carbon nanotubes and graphene. Carbon nanotubes and graphene in aluminum and copper alloys based composites enhanced their strength and tribological properties. The carbon nanotubes and graphene were observed to reduce the grain size in aluminum alloys, resulting in an additional higher strength. The incorporation of carbon nanotubes and graphene also increases the effectiveness of pure aluminum.

Table 1 Research reported in literature on metal matrix/nanocarbonous (including carbon nanotube and graphene) composites

Matrix	Reinforcement	Process	Properties	Ref.
Aluminum	Graphene	Powder metallurgy technique	Increasing the tensile properties by embedding 0.3 wt.% graphene while graphene particles were pulled out from the fracture surface	[31]
Aluminum	Graphene	Mixing AA2124 powder and graphene platelets and then cold compaction at 525 MPa pressure	Increasing the hardness and decreasing the relative density. On the other hand, there is an optimum point that wear rate is minimized.	[32]
Copper	nanographite	Powder metallurgy technique where nanographite were dispersed in ethanol and then copper introduced to solution, after that drying the powder and using 450 MPa pressure to make composite	Copper/nano-graphite exhibit better tribological properties than copper/micro-graphite. Also increasing the volume fraction of reinforcement tends to improve tribological properties	[30]
Aluminum	CNT	Synthesizing by hot press and hot extrusion.	No nanotubes damaged. No reaction products at the interface between the matrix and carbon nanotubes. No significant effect of annealing on the strength, while the strength of the pure aluminum matrix decreases with annealing time.	[33]
Aluminum	CNT	Sintering the mixture of aluminum and CNTs powders in a carbon mold under 50 MPa pressure	No change in elongation while there is a significant improvement in tensile strength.	[34]
Aluminum	MWCNT	High energy ball milling, cold compaction and hot extrusion were employed to synthesizing composite.	Wear resistance and hardness of composite significantly increased while COF decreased.	[35]
Chromium	MWCNT	Cr/MWNT coatings were electrodeposited from electrolytes	Wear rate of composite decreased in compared with unreinforced chromium	[36]

Extensive researches have been done on polymer matrix composites reinforced by graphene in the last two decades. Almost all of these researches have shown that by adding graphene as reinforcement to a polymer matrix, the properties of polymer matrix composites tremendously improved [37-42]. Tang et al. [37] have shown that by adding 1 wt.% graphene as reinforcement to polyvinyl alcohol, the tensile strength and tensile modulus increased by 178% and 139%, respectively. Additionally, only a very low volume fraction of graphene platelets (GPLs) increases the dielectric constant of polymer matrix composite. It has been also reported that by combining GPLs and BaTiO₃ and incorporating them into polymer matrix, the highest dielectric constant was reached without sacrificing the low dissipation factor [38]. Graphene based materials provide high thermal conductivity enhancement as well as the advantage

of improving barrier properties in comparison with polymer matrix composites reinforced by CNTs [39, 43]. Although, numerous polymer matrix nanocomposites reinforced by graphene nano-platelets and CNTs have been studied, still the challenge of adding nano carbonous materials into a metallic matrix and synthesize a fully uniform and dispersed structure is remained open to researchers [41, 44-47]. At the same time, another important processing issue in metal composite fabrication is the low interfacial strength between the CNTs and the matrix. In CNT/polymer nanocomposites, CNTs and polymer interact a molecular level while in the case of CNT/metal these types of bonding are not available.

It is expected that by adding graphene nano-platelets to a metal matrix, the mechanical and tribological properties would be enhanced. In literature, only limited researches on metal matrix composites reinforced by graphene are available [10, 32, 48-54]. To the authors' knowledge, so far, aluminum matrix composite reinforced by graphene has successfully produced only by powder metallurgy method [48, 49]. Recently, Wang et al. [48] have shown that by adding 0.3 wt.% Graphene nano-sheets to the aluminum matrix, the tensile strength of composite increased by about 62%. However, Bartolucci et al. [49] have shown that the tensile strength and strain at failure of aluminum matrix composites reinforced by 0.1 wt.% graphene platelets are less than its pure aluminum matrix. In addition to these two researches, Chen et al. [50] have produced magnesium matrix composite reinforced by graphene nano-platelets. They employed a novel method combining liquid state ultrasonic processing and solid state stirring to fabricate the composites. By using this novel method, they reported that the graphene nano-platelets (GNPs) could be dispersed uniformly into magnesium matrix. The results showed that the micro hardness of magnesium matrix composite reinforced by GNPs has been increased by 78% compared to that of pure Mg prepared under the same processing condition. They have also shown that the GNPs show an excellent strengthening effect on the magnesium matrix composite [50, 51, 55-58].

Although the emerging research interest in smart materials such as self-lubricating composites inspires both academia and industry that the combination of these carbonous materials and metallic matrices could potentially create composites that have high thermal and mechanical properties as well as exceptional wear resistance, there is still a need of understanding the nature, processing, and tailoring of these composites. This review initially covers a brief introduction of various nano-reinforcements potentially are used in self-lubricating nanocomposites. Then, it addresses the current progress of research in self-lubricating nanocomposites followed by discussing the effect of these reinforcements on tribological properties.

2. Properties of carbonaceous nanomaterials

2.1. Carbon Nanotubes (CNT)

If a sheet of carbon atoms is rolled, the carbon nanotubes will forms with a diameter of 1–2 nm which is called single-walled carbon nanotubes (SWCNT). Other types of carbon nanotubes are double- and multi-walled nanotubes with diameters ranging from 4 to 20 nm which are formed by rolling 2 or more carbon sheets as they are schematically shown in Figure 1. CNT has unique mechanical and physical properties as well as lubricant nature. Owe to these characteristics, it would be a promising candidate as reinforcement in a metallic matrix to enhance the properties of materials such as inherent stability at high temperature, high strength and stiffness, superior electrical and thermal conductivity and improved performance of metals in industrials components.

The results of the Brenner potential in predicting the modulus value of CNTs reveal that the modulus value of CNT is 1060 GPa that it is very close to in-plane graphing [59]. A study was employed using Molecular Dynamic (MD) simulation with a universal force field to measure the stiffness value of SWNTs and this turns into a 1 TPa stiffness value for SWNTs [60]. Researchers have developed several methods to measure elastic modulus and strength of CNTs and one of the key techniques is measuring the amplitudes of thermal vibration of nanotubes. This method measured an average value of 1.8 TPa for elastic modulus with a large scatter in the results ranging from 0.4 to 4 TPa. Using a similar technique, the modulus of 1.25 TPa for MWNTs grown by laser ablation was achieved [61].

Although SWNTs have superior mechanical properties, but yet they are not employed extensively as a reinforcement because they are costly to be produced and purified. On the other hand, MWNTs are easier to produce. MWCNTs are composed of number of centered layers. One of the drawbacks of using MWNTs is the susceptibility of the inner tubes to be pulled-out of the outer tube by tensile stresses. This phenomenon is usually referred as “telescoping effect” or “telescopic extension of multiwall carbon nanotubes”. In-situ TEM measurements on nanotubes demonstrated that this phenomenon occurs at ultra-low-friction state which revealed no wear or fatigue on the atomic scale. Telescoping failure and the higher probability of defects in MWNTs make them less favorable compared to SWNTs. However, still the strength measured in MWNTS is much higher than that of high strength metals such as steel [62, 63].

2.2. Graphene

Graphene is two-dimensional single atomic carbon sheet of sp^2 -bonded in which atoms densely packed in a honeycomb lattice. Graphite, the most common form of carbon, is stack of several graphene sheets along the c-axis with an interlayer spacing of 0.34 nm. The bonding between carbon atoms is very strong while there are weak van der Waals interactions among the layers. In terms of thermodynamics, it was thought that exfoliation of layered graphite to freestanding atomic layer would not be possible [53]. However, to date different approaches have been developed for synthesizing graphene in large quantities, including thermal evaporation of silicon carbide [64, 65], chemical vapor deposition (CVD) of graphene on metal carbides or metal surfaces [66, 67], and wet chemical synthesis of graphene oxides followed by reduction [68, 69].

The yield strength predicted for a single graphite layer using MD simulation has reached an extreme value of 0.912 TPa [70]. Another study employed quantum mechanical approach revealed that the elastic modulus for armchair graphene and zigzag graphene are 1.086 and 1.05 TPa, respectively [71]. The Young's modulus and intrinsic tensile strength of graphene monolayer were experimentally tested by using nano-indentation of the atomic force microscope (AFM). The Young's modulus and intrinsic tensile strength obtained using these techniques are 1.1.02 TPa and 130 GPa, respectively [72]. By using the same method, mechanical properties of graphene bilayer and trilayer have been determined where Young's modulus is 1.04 and 0.98 TPa and intrinsic tensile strength is 126 and 101 GPa, respectively [73]. These supreme mechanical properties of graphene along with extreme thermal conductivity ($5000 \text{ W m}^{-1} \text{ K}^{-1}$) [74], and super charge-carrier mobility ($200,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) [75] makes them an attractive material for researchers in the last decade to employ them as reinforcement into a metal matrix. The graphene has a plate shape; dispersion in any kind of matrices is easier in comparison with CNTs. Hence, the graphene too is a good substitution for CNTs as reinforcement for metal matrix composites [31].

Although graphene is defined as graphite single layers, graphene nanoplatelets (GNPs) or graphene nanosheets (GNSs) which are short stacks of platelet-shaped graphene sheets with an average thickness of the 5-100 nanometers are very common in the fabrication of metal matrix composites. Since, graphene in its single layer form cannot be easily be stable in the free State, usually GNPs are used as reinforcement and then the sheets are exfoliated to achieve a single layer dispersed graphene in a matrix. This inexpensive material possesses good thermal conductivity, electrical conductivity, mechanical strength and more surface area than the expensive carbon nanotubes (CNTs).

3. Self-lubricating nanocomposites

During relative motion of two surfaces, different types of wear mechanisms, including adhesive wear, abrasive wear, delimitation wear, erosive wear, fretting wear, fatigue wear, and corrosive/oxidative wear may occur. The nature of wear mechanisms can be understood by studying the worn surfaces of materials. At low loads and sliding speeds, abrasion is the dominant wear mechanism while at higher loads, the wear mechanism changes to delamination. Presence of grooves on the worn surfaces of the composites in the sliding direction at low normal loads shows that the abrasion wear mechanism becomes dominant.

Graphite is well-known reinforcement for metal matrix composites which acts as solid lubricant and makes the composite as self-lubricating composites [1, 4, 22, 30, 76, 77]. When graphite is embedded into a metal matrix, the friction and wear behavior of metal/graphite composite significantly improves compared to unreinforced metal which lead to their increased industrial applications where tribological properties are dominant. Damage accumulation will be reduced in the presence of graphite particles and hence decrease the wear rate of metal matrix insignificant extent. MMCs reinforced by graphite particles or fibers are potential structural materials for aerospace and automotive owe to their excellent tribological properties.

Among many alloys, aluminum based composites are extensively used in various industries because of high strength to weight ratio, superior tribological properties, and good corrosion resistivity. The explanation for the superior tribological properties of aluminum/graphite composites can be explained by the wear mechanisms which occur in these systems. Aluminum alloys have low yield stress and deforms extensively during sliding contact while graphite particles in aluminum/graphite composite improve the deformation and fragmentation of the surface and sub-surface by providing a continuous film of graphite on the contact surfaces after short running-in period. The graphite film hinders direct metal to metal contact and hence prevents seizure. Despite of good tribological behavior of metal/graphite composite, poor mechanical properties is the disadvantage in the graphite reinforced MMCs. These composites sometimes have lower mechanical properties than unreinforced alloy [1, 78]. In addition, graphite has a reverse effect on electrical conductivity when copper alloys are reinforced by micron sized graphite due to be hindering effect of particles in the continuous copper matrix network, though it has a moderate electrical conductivity. Another feature that causes to reduce electrical conductivity of copper-graphite composite is the poor interface bonding between copper and graphite particles which leads to more electron scattering [30]. Due to these shortcomings in using graphite as reinforcement in metals, incorporation of nano-sized carbonous materials sought to be promising.

In general, it is desirable in terms of mechanical properties to have matrix grain size in the range of nanometer to achieve enhanced hardness, yield strength, and tribological properties such as wear

resistance and friction coefficient [79]. Using nano size particles as reinforcement also enhances both Young's modulus and tensile strength of composites as well as improving tribological performance. Due to the fact that nanocarbonous materials have superior physical and mechanical properties, they have recently employed as a novel reinforcement for self-lubricating metal matrix nanocomposite. Superior properties of MMNCs reinforced by carbon nanomaterials is due to metallurgical factors, such as Hall–Petch effect by grain size refinement, Orowan looping and dislocation generation resulting from thermal mismatch between the matrix and reinforcements [53]. Previous studies revealed that the MMCs with smaller size reinforcements exhibit lower coefficient of friction and wear rate, thus, it was concluded that the MMNCs have excellent tribological properties rather than metal matrix micro-composites, as it is also experimentally confirmed and the results are presented in Figure 2 [1, 80, 81]. More specifically, the composite reinforced by nano particles (graphene) has lower COF than the composite reinforced by micro particles (graphite). Also, the hardness of composites reinforced by graphene is found to be higher than the composite reinforced by CNTs [4]. Worn surface observation suggested that the dominant wear mechanism for non-reinforced pure Al specimen has been delaminating wear accompanied by some adhesive wear mechanism. However, worn surfaces of the nano-particle reinforced composites were smoother and the total depth of deformations were smaller, grooves were finer than the unreinforced aluminum alloy matrix specimens [82, 83]

There is a great challenge in introducing carbonous materials to metal matrices. Generally, molten aluminum is not able to wet carbonous materials, such as carbon fibers (CFs), graphite particles, carbon nanotubes (CNTs) and graphene where the contact angle of molten aluminum with graphite is between 140° and 160° [84]. The reason for high contact angle between carbonous materials and molten aluminum is due to the high surface tension of aluminum in liquid state. The surface tension of molten aluminum and carbon nanotubes are 955 mN/m and 45.3 mN/m, respectively [85]. The very high value of the surface tension of molten aluminum compared to carbon nanotubes makes synthesis of aluminum matrix composite reinforced with carbonaceous materials a challenging task. One of typical way to improve wetting behavior of molten aluminum on carbonous materials is by forming metallic coatings, such as copper and nickel on reinforcements to reduce its contact angle, as shown in Figure 3 [86, 87]. The formation of Al_3Ni , Al_3Ni_2 and $CuAl_2$ as an intermetallic compound plays a key role in achieving good wettability between aluminum with copper and nickel [28, 88]. In the following sections, self-lubricating metal/CNT and Metal/ Graphene (single layer or nanoplatelets) nanocomposites have been introduced and their mechanical properties are discussed.

3.1. Metal-CNTs nanocomposites

The shear lag models, used in the case of conventional fiber reinforced composites, have also been applied to CNT composites. The stress is transferred to the fiber (σ_f) through the interface and is related to the shear stress (τ_{mf}) between the fiber and matrix given by:

$$\frac{l_f}{D_f} = \frac{\sigma_f}{2\tau_{mf}} \quad (1)$$

Where l_f and D_f are the length and diameter of the CNT, respectively. Carbon nanotubes with a larger aspect ratio assist larger load transfer and hence efficient utilization of reinforcement. For a critical length l_c , the value of σ_f becomes equal to the fracture strength of CNTs. For nanotube lengths $l < l_c$, the fracture strength of the composite is given as:

$$\sigma_c^{Frac} = V_f \sigma_f^{Frac} \left(\frac{l}{2l_c} \right) + V_m \sigma_m^{Frac} \quad (2)$$

Dislocation generation by thermal expansion mismatch between matrix and CNTs and also Orowan looping mechanism play an important role in strengthening of aluminum/CNTs composites [89]. To achieve the theoretical value of strength prediction, uniform dispersion of CNTs in the matrix during synthesizing is a great challenge [90]. Many attempts have been done in synthesizing CNTs reinforced metal matrix composites using traditional liquid casting or powder metallurgy processes which have not shown promising results [91-93]. In case of liquid mixing due to the poor wetting properties of CNTs and graphene with liquid aluminum and the difference in their densities, CNTs and graphene immediately float to the surface without being mixed. In P/M route and hot pressed sintered aluminum CNT composites, CNTs cause a tremendous amount of residual stress in aluminum matrix which cause a lower the tensile strength in the non-annealed composites than that of pure aluminum. Additionally, the bonding obtained through using these techniques is usually weak, which causes the load transfer ineffectiveness.

The potential energies of interaction between two parallel infinitely long carbon nanotubes of the same diameter can be simplified greatly by assuming only van der Waals interactions in graphitic systems. For a pair of parallel carbon nano tubes at a distance of about 0.315 nm the cohesion energy has been calculated to be about 37kT nm⁻¹. So, an agglomerated CNT bundle needs about 120 kT nm⁻¹ since three tube-tube contacts have to be broken to separate a tube from a bundle. On the other hand, carbon-carbon bond energies lie about 190 kT. From this standpoint, it can be concluded that the strong cohesion and very small difference between carbon- carbon covalent bond energy, and CNT-CNT van der waals energy, dispersing nanotubes is a difficult task which needs careful considerations. In polymer matrix composites, generally, ultrasonication aqueous media combined with an appropriate surfactant is a technique to de-agglomerate CNTs. However, the task becomes more sophisticated in metals processing. Although high intensity ultrasonication during liquid metal processing has been employed in synthesizing nanocomposites, but still no experimental or simulation data is available in CNT reinforced nanocomposites confirming the complete and successful dispersion of these reinforcements in a metallic matrix [94-97].

In late 1990s, Kuzumaki et al. [33] fabricated aluminum/5 vol.% CNT and aluminum/10 vol.% CNT composites by mixing aluminum powders and carbon nanotubes in ethanol followed by hot pressing and extrusion. However, no improvement in tensile strength of aluminum/5 vol.% CNT and aluminum/10 vol.% CNT was reported in comparison to pure aluminum due to the poor dispersion of nanotubes in aluminum matrix. Different factors, such as type of nanotubes, functionalization of nanotubes, nanotube contents, matrix materials, and milling times can significantly affect the dispersion of CNTs in a metallic matrix. Generally, to achieve a uniform dispersion of CNTs in the final composite, a homogenous distribution of CNTs in the powder mixture at the starting stage is an important factor [53]. Uniform dispersion of CNTs in matrix tends to increase the hardness of composites compared to unreinforced pure aluminum. If only a small amount of CNT is embedded into a metal matrices, the hardness of the composite increases due to the fact that the micro voids of metal matrix will be filled by CNTs. Beyond a specific CNT volume percentage, the excess CNTs which were not able to fill the micro voids will be agglomerated with the aluminum particles. This agglomeration interrupts the complete sintering and leads to the formation of defects which ultimately results in gradual reduction in hardness [98, 99].

Deng et al. [29] have investigated the physical and mechanical properties of aluminum AA2024/MWNT nanocomposites synthesized by cold isostatic press followed by hot extrusion. During synthesis, AA2024 powders and MWNTs were first mixed in ethanol using ultrasonic mixing. Then, ethanol evaporated and the dried mixture ball milled and finally cold isostatic pressing and hot extrusion at 450 °C were performed. Figure 4a shows the changes in relative density and Vickers hardness with CNTs content. By increasing the weight percentage of CNTs up to 1 wt.% of MWNT, the relative density and hardness of the nanocomposites increase. It can be clearly seen that the relative density of nanocomposite containing 2 wt.% MWNT sharply dropped which could be due to the formation of nanotubes clusters. As shown in Figure 4b, tensile strength and elongation of aluminum/CNT composites depict the same trend as the relative density. The AA2024/1 wt.% MWNT exhibit maximum elongation and tensile strength and there is a drop in mechanical properties at high amount of reinforcements.

Noguchi et al. [100] developed a two-step method in which initially CNTs were uniformly dispersed in an elastomer matrix; and in the second step, the elastomer matrix was displaced by Al. Low-energy ball milling in a Turbula mixer followed by hot compaction, sintering and HIPing was another route which was used to homogenize the mixture Al/CNTs and eliminate CNT clustering. Esawi et al. [101], showed the effect of mixing time and mixing speed on CNT cluster size using dry mixing in a Turbula mixer. He et al. [102] were also developed an in-situ route to synthesize CNTs inside the aluminum matrix by using a three-step process including deposition-precipitation, reduction, and chemically vapor deposition route. They have reported that the first step involves producing a Ni(OH)₂-Al precursor through a deposition-precipitation route. In their method, Ni nanoparticles were used as catalyst on which the CNTs will grow. Therefore, by uniform distribution of Ni nanoparticles on the surface of Al powders, a uniform distribution of CNTs could be guaranteed. Cha et al. [103] used another technique to mix CNTs and matrix material in a solution. In their fabrication process which basically involves molecular-level mixing of CNTs and the matrix material, a suspension of reinforcements, and dissolved metal ions in ethanol or water is prepared. Then the suspension was dried and oxidized metal powders forms. Finally, the metal oxides are reduced under control atmosphere to achieve a dispersed CNT reinforced metal matrix composite.

3.2. Metal-graphene nanocomposites

Similar to aluminum/CNT composites, in order to achieve the full potential of graphene nano sheets (GNSs) as reinforcement, a homogeneous distribution of GNSs in the aluminum matrix along with maintaining the structural integrity of the GNSs is essential. Aggregated graphene behaves no differently than particulate graphite platelets. The ultrahigh surface area that can be obtained in a 2D graphene sheet is lost when these sheets are clustered [54, 104]. In this regard, the main challenge in fabricating metal/graphene nanocomposites is to find an approach to fully disperse these sheets or exfoliate the single sheets of GNS to graphene monolayer. Although, exfoliation of GNSs in polymeric matrices has been studied extensively and successful results have been achieved, but yet in a metallic matrix it has been remain as the key challenge. The main obstacle in achieving a highly exfoliated structure in metal-graphene nanocomposites is the high difference between carbon and metals surface energies. This high surface energy difference does not let metal to easily wet the graphene sheets and fall them apart. Several investigations have been carried out to find the best route to synthesize a fully dispersed and homogenous graphene reinforced metal matrix composite including electrochemical deposition, metal evaporation, and hydrogen reduction of metallic salts-graphite composite [105-107]. However, so far fully exfoliated

graphite flakes have not been obtained. Additionally, the difficulty in large-scale synthesis of these composites becomes an obstacle in their production.

The powder metallurgy (P/M) route which is recently widely developed for the fabrication of aluminum/CNT composites can be considered as an applicable pathway to fabricate aluminum/GNS composites. Graphene oxide (GO) nanosheets have hydroxyl and epoxy groups on its surface which helps to have better dispersion in solvents and form more stable solutions than graphene. That makes the GO nanosheets a more favorable reinforcement rather than GNSs [108]. Figure 5 shows the main difference between graphene and GO. Wang et al. [31] reported a route in synthesizing aluminum/GO composites using powder metallurgy in which four steps are involved:

(1) Exfoliating GO into several-layered or single-layered nanosheets by sonicating GO aqueous dispersion in deionized water.

(2) Aluminum flakes surface modification through ball milling followed by introducing a hydrophilic membrane on the surface of the aluminum flakes such as PVA.

(3) Adsorption and reduction of GO nanosheets by adding the powder slurry of modified aluminum flakes in deionized water to the GO aqueous dispersion. The mixed slurry color changes from brown to transparent during stirring. Heating the aluminum/GO composite powders decompose the hydrophilic membrane and reduce the GO nanosheets to GNSs, until finally aluminum/GNS composite powders is obtained.

(4) Compacting and consolidation of aluminum/GNS composite powders. Consolidation can be achieved by sintering in an argon atmosphere followed by hot extrusion.

Bastwros et al. [10] used graphite and exfoliated it to graphene in nitric acid and sodium chlorate solution. The intercalated graphite was achieved through sedimentation and finally the intercalated graphite was exfoliated to monolayer or few-layer graphene oxide using ultrasonication. Then, they employed ball mill to mix Al6061 powder and graphene at different milling times. The composites were then synthesized by hot compaction in the semi-solid regime of the Al6061. Ghazaly et al. [32] have synthesized the aluminum graphene at different weight percentage (0.5, 3 and 5 wt.%) by employing powder metallurgy technique. Aluminum powder with the graphene nanosheets were mixed in a high energy ball mill which resulted in the formation of nonuniform particles of aluminum covered by graphene layer and disappearance of the graphene nanosheets. A combination of cold compaction and hot extrusion at $\sim 0.45T_m$ (305 °C) were employed to synthesize aluminum/graphene self-lubricating nanocomposite. The results show a decrease in density of nanocomposites by increasing the amount of graphene as shown in Figure 6. Also, the variation of hardness with increasing the amount of graphene is presented in Figure 6. As there was an expectation that addition of graphene up to 3 wt.%, the hardness of nanocomposites reinforced by graphene increase when compared to unreinforced alloy. The 3 wt.% graphene reinforced composite produced a 47.5% increase in hardness over the base AA2124 alloy. By further increasing the amount of graphene (more than 3 wt.%) nanocomposite, the hardness reduces but the hardness values are more than the unreinforced alloy and less than the composite with reinforcements between 0.5 and 3 wt.%. By comparing Figure 4 and Figure 6, it is obvious that aluminum/graphene has superior mechanical properties than aluminum/CNT.

To achieve individual graphene sheet, according to many available literatures, another method is attaching molecules or polymers to graphene sheets to decrease aggregation [52, 109]. Xu et al. [54]

suggested a combined technique in which a solvent, graphene oxide, and an inorganic nano particle was incorporated to obtain graphene as individual sheets inside a metal matrix. The inorganic nanoparticles sits in between graphene sheets and prevent re-agglomeration of them. . After the solvent is dried, dispersion of isolated graphene sheets can be obtained having graphitic stacks with inorganic nanoparticles in the interlayer spacing.

Aluminum/0.3 wt.% graphene nanocomposite was also synthesized by using a slurry based process followed by sintering and hot extrusion [31]. The normalized tensile strength and ductility of Al/GNS composites in comparison with unreinforced pure matrix is shown in Figure 7a. The tensile strength of aluminum/GNS nanocomposite is 249 MPa. The nanocomposite showed a significant improvement (about 62%) in tensile strength compared to that of unreinforced aluminum matrix (154 MPa). Higher tensile strength of nanocomposite compared to unreinforced aluminum demonstrate that GNSs have a dominant role for increasing mechanical properties and reveal that the GNSs have a good potential to be used as reinforcement in aluminum matrix composites to improve the mechanical properties. Generally, GNSs could contribute to the strength improvement by grain size refinement, dislocation strengthening and stress transfer. GNSs hinder grain growth and hence result in having grain refinement and also provide resistance to the dislocation movement during thermal processing and plastic deformation, therefore, the tensile strength of aluminum matrix increase in presence of GNSs. Another important reason of high tensile strength in presence of GNSs is the load bearing ability where the graphene tolerate a substantial part of load during plastic deformation. The fracture surface is shown in Figure 7b. Inasmuch as the fracture strength of a perfect single layer graphene is 125 GPa, the theoretical strength of the aluminum/graphene is 500 MPa by using the rule of mixtures for aluminum/0.3 wt.% GNSs. The experimental result shows that strength of aluminum/0.3 wt.% GNSs is 250 MPa. The less strength in experimental than theoretical study is due several parameters, such as to different alignment of GNSs along the tensile direction, processing parameters, microstructure, and weak interfacial bonding of the aluminum/GNSs [49].

Latief et al. [110] have synthesized aluminum/ graphene composites using different percentages of exfoliated graphite nano-platelets particles by employing powder metallurgy method in order to study the physical and mechanical properties of nanocomposites. As observed in Figure 8, the results revealed that the Vickers hardness (Figure 8a) and compression strength (Figure 8c) increases with increasing graphene content up to 5 wt.% in the pure aluminum matrix while the density (Figure 8b) decreases with increasing graphene content. This can be expressed by following equation [111]:

$$\lambda = \frac{4(1-f)r}{3f} \quad (3)$$

Where λ , f and r are respectively the distance between the reinforcement particles, particles volume fraction, and the particle radius (particle is assumed spherical). The shear stress for all tested alloys can also be calculated according to the following equation:

$$\tau_0 = \frac{Gb}{\lambda} \quad (4)$$

where, τ_0 , G and b is the shear stress, the shear module, and the Burger's vector, respectively [112]. According to Equation (3), it can be concluded that the distance between the graphene particles decreases by increasing their amount in the composite. Based on Equation (4), the shear stress that is

required to move dislocations between the graphene particles will increase when the distance between the reinforcement particles decreases which results in an increased yield stress of materials [113, 114]. In addition, the reinforcements are an obstacle that causes to lock dislocation movement extremely in metal matrix through dispersion strengthening mechanism. This mechanism can increase the mechanical properties of aluminum matrix composites reinforced by graphene [111].

Bartolucci et al. [49] have compared the mechanical properties of aluminum/MWNT and aluminum/GNP composites. Figure 9 depicts the hardness of self-lubricating aluminum nanocomposites at different samples after hot isostatic pressing and extrusion. It is obvious from the results of hardness that the aluminum reinforced with 1.0 wt.% MWNT exhibits the highest hardness than aluminum/0.1 wt.% graphene and pure aluminum. Figure 10 shows a comparison between the tensile strengths of composite reinforced by MWCNT and graphene and also effect of fabrication method including extruded and pressed composites. The metal matrix composite reinforced by nanotubes show the higher strength than reinforced by graphene. For extruded composites, the tensile strength of aluminum/nanotubes was about 12 percent greater than the pure aluminum while the aluminum/graphene showed about 18 percent lower tensile strength as compared to the pure aluminum. In addition, Figure 10 shows the average strain-to-failure of the samples where the results revealed samples reinforced by nanotubes and graphene displayed the lowest ductility.

4. Tribological behavior of self-lubricating nanocomposites

Tribology is an investigation on wear and friction performance of materials. It is the science of interacting surfaces in relative motion [115]. When under an external load, two materials are in contact with each other, the asperities of two surfaces come into close contact and during movement, deterioration of the surfaces occurs which is known as wear. During the sliding process of softer materials against harder materials, atoms will be taken off from the softer one and these atoms tend to locate themselves in the asperities of harder surface. As results, a cold welding occurs in contact surface and interatomic junctions across the interface forms. By continuing the sliding process, fracture can take place at the junctions and causes the detachment of the fragments from adhering asperities. Friction force causes shear at interatomic junctions during movement of two surfaces under an applied force. Archad [116] expressed a formula for wear of materials that describes the volume of wear loss (V) of materials due to adhesive wear:

$$V = c \frac{PL}{H} \quad (5)$$

where c , P , L and H are wear coefficient, applied load, sliding distance, and hardness of the softest contacting surfaces, respectively.

Usually to avoid friction and consequently deterioration of material under wear, liquid or solid lubricants are employed. However, in cases such as high vacuum environment, high-speed conditions, high applied loads, and very low or high temperatures, liquid and grease type lubricants are undesirable. In such a tribological systems the common liquid and grease type lubricants do not show desired performance or durability [53]. Another approach is replacing the liquid and grease type lubricants with solid lubricant coatings that they are used to decrease coefficient of friction and wear rate. The coatings are applied on the surface of materials by depositing via chemical or physical vapor deposition techniques

to form a coating layer [117, 118]. The disadvantages of solid lubricant coating are limited lifetime, difficulty in replenishment, oxidation and aging-related degradation, and poor adhesion. Therefore, to avoid the drawbacks of both the liquid and grease type lubricants and the solid lubricant coatings, embedding carbonous materials in the metal matrix seems promising.

Generally, metal matrix composites have lower coefficient of friction (COF) compared to unreinforced matrix [1, 4, 35, 119-121]. Furthermore, adding ceramic particles to the metal matrices lead to an increase in wear resistance of the matrices [1, 4, 121-124]. The main reason for increasing of wear resistance of metal matrix composite is attributed to low friction coefficient of metal matrix composite compared to the unreinforced metals. For conventional metal matrix composites, the reinforcements act as load bearing components at contact surface which tend to protect the surface from ploughing during sliding. Generally, the hardness of reinforcement greatly affects the wear loss and hence, the wear volume of MMCs. The wear loss of MMCs depend on several intrinsic properties such as the reinforcements dispersion state, distribution of reinforcement, size of reinforcing particles, and interfacial bond between matrix and particles [53]. When bonding between matrix and reinforcement is poor, the hard ceramic particles are easily pulled out from MMCs and then they will be trapped between the sliding surfaces and act as third body abrasives and help to increase worn surface damage and wear rate. Among the composites, composites reinforced by carbonous materials show better tribological properties compared to composites reinforced by ceramic reinforcements, such as SiC and Al₂O₃ due to the lubricative nature of carbonous materials that make them a potential reinforcement for self-lubricating composite. The conventional self-lubricating composites are embedded by graphite particles or carbon fibers [1].

The main reason for significant decrease in COF and wear rate is due to formation of a lubricant film between the contact surfaces because of presence of carbon-based solid lubricant in the MMCs. Thus, the lubricant film prevents direct contact between sliding surfaces and reducing wear [78]. In addition, due to the presence of lubricant film which prevents direct contact, the transfer of atoms from the asperities of softer surface to the asperities of harder surface will be reduced that hence, it leads to decrease in cold welding of atoms of softer materials with atoms of harder materials during sliding and then subsequent fracture of atomic junctions [30]. As noted before, although the graphite particles in the metal matrix improve the tribological performance, it tends to reduce the mechanical properties of the composites. Hence, recently, the nano solid lubricants are used as the dominant reinforcement for the metal matrices in self-lubricating composites. This is because the metal matrix composite reinforced by nano solid lubricant have excellent self-lubricating behavior with low coefficient of friction and wear rate as well as high mechanical properties [125].

4.1. Effect of CNT

Carbon nanotubes (CNTs) are one of the important solid lubricants that is increasingly employed in novel self-lubricating nanocomposite materials because of the superior properties of CNT including electrical, optical, and mechanical properties and have a key role in enhancement of wear resistance and reduction in COF of metal matrix composite reinforced by CNTs. Metal matrix nanocomposites reinforced by CNT is an excellent candidate for industrial applications because of its excellent mechanical properties, lightweight and superior tribological properties. It is expected that the utilization of Multi-Wall Carbon Nano Tubes (MWCNTs) in the composites will increase the industrial applications due to their reasonable cost. Thus, there have been many investigations to develop MMNCs reinforced with MWCNTs using various fabrication routes [1]. Previous investigations exhibit superior tribological

properties of metal/CNTs composites as a result of the reduction in wear rate and the coefficient of friction (COF) due to the lubricating nature of CNTs. CNTs form a lubricant film between contact surfaces during sliding.

It has been reported that when there is a strong bonding between functionalized MWNTs and an epoxy matrix the outer shells of the tube remain embedded in the matrix following pull-out [126, 127]. However, in case of metal matrix, the MWCNTs are attached by a very weak van der Waals forces, there is no direct observation that support the telescopic extension of multiwall carbon nanotubes occurs. The weak bonding between CNT-metal led to easily slide or roll between the contact surfaces and minimize a direct contact between the surfaces, thus results in decrease in friction coefficient of the composite. The improvement in wear resistance is attributed to the role of CNTs as spacers that prevent direct contact between rough surfaces [28]. Generally, several material parameters, such as amount of reinforcements, size of reinforcement, and spatial distribution have direct effect on tribological properties of self-lubricating metal/CNTs composites [1, 128].

Zhou et al. [129] fabricated Al-Mg/MWCNT composite by using a preform with MWCNT and then using pressureless infiltration method to infiltrate molten metal into the MWCNT preform. Embedding the MWCNT into Al-Mg alloy increases the hardness of the composites compared to unreinforced aluminum alloy. Furthermore, by increasing the MWCNTs volume percentage, the hardness of composite initially increases. Then, further increase in MWCNTs content has an adverse effect and decreases the hardness as shown in Figure 11a. The effect of volume fraction of CNTs on the friction coefficient and wear rate of the composite is shown in Figure 11b. It can be seen that although hardness has an optimum value with increasing volume fraction of CNTs in the composite, yet the coefficient of friction decreases even at high CNT content. At high amount of CNTs the direct contact between the metal surfaces is hindered which ultimately results in better tribological properties. The favorable effects of CNTs on tribological behavior of composites depend on their excellent mechanical properties, well dispersity in the composite and the efficiency of CNTs as reinforcement. X-ray diffraction (XRD) analysis of contact surface reveals that the wear particles are mainly aluminum oxide. During wear process, laminated oxide films formed at contact surfaces that they subsequently broke up and flaked off due to low adhesion between the oxide films and aluminum matrix. While the oxide particles which form at contact surface are harder than the aluminum matrix and are able to increase abrasive wear. As the aluminum matrix gradually wear out during sliding process, the CNTs which was initially embedded in the matrix now are pulled out and exposed on the contact surface and form a lubricant film on worn surface. Those solid lubricating films significantly reduce the adhesive wear cause by oxide particles compared to unreinforced aluminum.

Choi et al. [125] have shown that the wear loss and coefficient of friction decrease with increasing the CNTs content. However, beyond a critical amount, 4.5 vol.% CNTs in their investigation, the wear rate and friction coefficient increase as shown in Figure 12. The deteriorated wear properties in the composites at high volume of MWCNTs may be associated with the presence of voids and cracks due to the very high amount of CNTs which could act as a source of delamination. As shown in Figure 13(a) and (b), grooves and material delamination were observed on the worn surfaces for pure aluminum and aluminum reinforced by 1.5 vol.% MWCNT. This would confirm that micro-ploughing and delamination are the two main dominant wear mechanisms of pure aluminum and aluminum/1.5 vol.% MWCNTs. By increasing the volume percentage of MWCNTs to 3 and 4.5 vol.%, less deep grooves can be observed and the surface is much smoother compared to the one it was observed in Figure 13(c) and (d). The surface of

aluminum reinforced by MWCNTs of 6.0 vol.% demonstrates rougher worn surface than aluminum/4.5 vol.% MWCNTs. At this amount of CNT the debris are able to easily separate out from the surface which can be justified by presence of pores as shown in Figure 13(e). This can be the main reason for increasing of wear loss and COF at high volume percentage of MWCNTs. Furthermore, They confirmed that the coefficient friction of aluminum composite significantly decreased by adding CNTs where COF is 0.35 and 0.06 for pure aluminum and aluminum/4.5 vol% MWNT, respectively, under an applied load of 30 N and a sliding speed of 0.12 m/s. In addition, there is a variation between COF and volume content of CNT where COF decrease by increasing the amount of CNTs. The effect of applied load and sliding velocity on coefficient of friction and wear loss was also investigated and it is shown in Figure 14. The investigations have shown that the COF and wear loss increases with increasing normal load for aluminum/5 vol. % MWNT composite at sliding speed of 0.12 m/s. However, the coefficient of friction was still lower than 0.1. At higher load, severe wear is the dominant mechanism which results in increasing friction coefficient and wear loss and subsequently severe surface damage. On the other hand, the coefficient of friction and wear loss slightly decreases with increasing sliding speed at an applied load of 30 N as shown in the Figure 14.

4.2. Effect of graphene

High strength, lightweight and lubricating nature of graphene made it suitable as reinforcement for self-lubricating ultrahigh strength metal matrix nanocomposites. As this is fairly a novel material and it is difficult to uniformly disperse in metals as well as its complex microstructure, there are only a few studies which investigated the tribological properties of graphene in a metallic matrix. Ghazaly et al. [32] who have investigated the effect of weight percentage of graphene on mechanical properties, also studied its effect on wear rate of self-lubricating AA2124 aluminum alloy matrix nanocomposites. The results showed that self-lubricating composite reinforced by 3 wt. % graphene has better tribological properties under dry wear test compared to unreinforced and other amount of graphene reinforcements as shown in Figure 15. SEM micrographs of worn surfaces of unreinforced aluminum alloy and Al/graphene nanocomposites are shown in Figure 16, which clearly demonstrate the presence of longitudinal grooves in all samples. In addition, by comparing the worn surfaces, it is obvious that the scratches, craters, and delamination of AA2124/3wt.% graphene composite is less than that of unreinforced alloy. Thus, unreinforced alloy and AA2124/3wt.% graphene composite are in the severe and mild wear regime, respectively. Shallow parallel grooves and ridges formed on the worn surface of AA2124/0.5 and 5 wt.% graphene nanocomposite due to microploughing. Thus, the dominant wear mechanism is severe plastic deformation of the matrix that results in high wear rate. Entrapped debris between delaminated surfaces was observed at high magnification on worn surfaces of unreinforced AA2124 alloy while there is no wear debris on the worn surfaces of nanocomposite as illustrated in Figure 17. Alumina fragmented films or strain hardened particles are the two main sources of debris. This debris is from the heavily milled consolidated powders which were detached under the load during the wear test. By comparing the worn surfaces at high magnifications, it is obvious that the surface of nanocomposite containing 3% graphene is smoother than that of unreinforced alloy and the composite reinforced with 5wt.% graphene. Furthermore, the surface of AA2124/3 wt.% graphene composite was covered by lubricant films that results in reducing friction and wear due to the soft nature of the lubricant film. Conversely, deep grooves and severe damage exist on the worn surfaces of AA2124/5 wt.% graphene composites which delaminated in the direction of sliding that explains the significant increase in wear rates and weight loss.

Inasmuch as copper has good electrical and thermal conductivities and graphite has lubricious nature, copper/graphite composites have variety of application in industries. Conversely, the mechanical properties of copper composites decrease in the presence of graphite reinforcement. To solve the impact of micro sized graphite particles, Rajkumar et al. [30] employed powder mixing, compaction and microwave sintering methods to synthesize copper nanocomposite reinforced by nano-graphite (NG) particles with an average particle size of 35 nm to form copper/5–20 vol% NG nanocomposites. The graphite particles were coated with copper using electrodeless plating method. The nano graphite particles have not been exfoliated in this investigation and cannot be considered as single or “few layer” graphene sheets. Table 2 compares the physical properties, such as relative density and electrical conductivity and also mechanical properties, such as hardness of sintered copper/graphite composite and copper/nano-graphite composites. As comparison shows, the nanocomposites had better hardness and electrical conductivity compared to microcomposites. As stated earlier, the volume percentage of nano-particles has an effect on physical and mechanical properties of self-lubricating composites. The amount of nano-particles is also influence the relative density as shown in Table 2. The relative density increases with increasing the volume percentage up to 15 vol.% of nano-graphite due to ability of nano particles to fill up the porosity cavities. When the nano-graphite amount is increased over 15% volume fraction, the relative density and hardness reduced due to the reduction in the distance between particles, which consequently facilitate nano particles agglomeration.

Table 2 Comparison of physical and mechanical properties between micro graphite particles and nano graphite particle [30]

Composite	Relative Density (%)	Hardness (HV)	Electrical Conductivity (%IACS)
Cu-15% Gr	92.3±0.13	72±1.6	65±1.5
Cu-5% NG	95.82±0.14	94±1.9	79.8±1.9
Cu-10% NG	96.41±0.13	90±1.0	72.4±1.0
Cu-15% NG	96.92±0.12	81.5±1.6	70±1.3
Cu-20% NG	88.42±0.15	56±2	38.7±2.5

Figure 18 illustrates the variation of normal load and sliding speed with coefficient of friction at different volume percentage of graphite particles. Figure 19 shows the variation of wear rate with normal load for copper based composites reinforced by micro and nano graphite particles. Results revealed that, at constant volume fraction, embedding nano-particles decreases the coefficient of friction and wear rate compared to the composite reinforced by micro sized graphite particles. Higher hardness, lower porosity and finer microstructure are the reason for the improved wear resistance of nano-graphite reinforced composites. Further, the nano-graphite particles reinforced composites are more effective on the degree of self-lubrication compared to micron-size graphite particles reinforced composites. The amount of nano-graphite particles also influences the tribological properties of self-lubricating copper composites. The increase in volume percentage of nano-graphite up to 15 vol.% tends to decrease the wear rate and COF because of the formation of a uniform and continuous layer of solid lubricant film. This lubricant film

reduces the rate of deformation of the matrix and improves the tribological behavior. When the amount of reinforcement increases, the decrease in the COF is associated with increase in the availability and uniformity of lubricant layer. The lubricant layer causes to minimize the metal to metal contacts between the copper matrix composite and steel counter surface. In contrast, when the volume fraction of nano-graphite is more than 15 vol.%, a large amount of agglomeration was observed that tends to incomplete spreading of graphite at the contact zone, and hence, increases the wear rate. Increasing of COF at high volume fraction of nano-graphite is a result of increasing the deformation and fracture at the contact surface of copper matrix and increasing the amount of copper debris at contact surfaces.

It can be seen from Figure 18 and Figure 19 that the wear rate and coefficient of friction both increase with increasing applied load while the coefficient of friction decrease with increasing the sliding speed. Increasing normal load also increases the amount of copper wear debris at the contact zone and hence influences the rate of increase in the coefficient of friction with normal load. In these figures, it can be seen that the coefficient of friction of self-lubricating composite significantly decreases with increasing the sliding speed up to 1.77 m/s because of formation of uniform lubricant film. By increasing the sliding speeds beyond 1.77 m/s, the coefficient of friction slightly increase or become constant for 5 and 10-15 vol.%, respectively. This is due to peel off of the self-lubricating film on the contact surface at high sliding speed. Furthermore, sliding speed does not affect the coefficient of friction of copper/nano-graphite with high amount of nano-graphite content due to the contact surface that is uniformly covered with the highly adherent graphite layer. As shown in SEM micrograph of worn surface at different sliding speed, at constant normal load, in Figure 20, the lubricant film on copper/15 vol.% nano-graphite is not continuous at lower sliding speed. While increasing the sliding speed, a lubricant layer uniformly form on the surface of composite that decreases COF as a direct result of a decrease in direct surface to surface contact. However, a gradual increase in COF was observed for 20 vol.% of nano-graphite composite by increasing the sliding speed that it leads to lower mechanical properties such as hardness due to increase in temperature at the interface. Further, it tends to more grain fracture during sliding. This phenomenon is more intensive at higher sliding speeds.

The mechanism of wear under normal loads suggested by Huang et al. [130]. Fine graphite particles form an adherent layer at the contact zone and under high normal loads; these nano-graphite from composite is squeezed out to the contact zone. Owe to their smaller size, nano graphite particles are able to penetrate deep inside the asperities of composite and counter surface during the sliding process. During sliding, the nano-graphite particles could have filled most of the asperities of the composite surface. So a graphite layer forms at the pin (composite) –disc (steel) interface. The layer formation process continues up to the formation of thick adherent graphite layer. In case of micron size graphite particles, they can also undergo similar process; however due to their larger size they are not able to penetrate into the very narrow grooves which formed during the wear process or gaps between the asperities of sliding contact easily.

Additionally, when the size of graphite particles comes down in the range of nanosize, at a same volume fraction, the mean free path between the graphite particles also decreases (Figure 21a) compared to same volume fraction of micron sized graphite particles (Figure 21b). This will cause smaller size asperities and also less space between the asperities compared to micro graphite reinforced composite (Figure 21d) during the wearing process which can be filled nano-graphite particles as shown in Figure 21c. The completely filled nanographite particles produce more uniform graphite layer that reduces the direct contact between the two wearing body and will cause reduction in the frictional coefficient. As

confirmed by SEM (Figure 21h and Figure 21f), nanocomposite reduces the wear debris size, as shown in Figure 21e when compared to microcomposites (Figure 21f)[30].

5. Conclusion

Carbon nanotubes (CNTs) and graphene have superior properties, including large aspect ratio, exceptional high Young's modulus and strength, and excellent electrical and thermal conductivity. These unique properties attract researcher to use them as reinforcement for metal matrix composites to enhance properties of composites and make them high strength, lightweight and self-lubricating. These favorable properties can be achieved only if the reinforcements are dispersed uniformly and not agglomerated in the matrix. Based on the literatures available, so far, a few studies have been conducted on metal matrix composite reinforced by CNTs and graphene because of difficulty in synthesizing and dispersing. Most researches were focused on fabrication and some of them have been investigated the mechanical properties and self-lubricating properties. Mechanical properties of composites were remarkably increased by adding CNTs and graphene. Important phenomena for increasing the mechanical properties of metal matrix composites reinforced by CNTs and graphene are dislocation generation by thermal expansion mismatch between the matrix and reinforcements, and also Orowan looping mechanism. Further, a few research papers are available on tribological behavior of self-lubricating nanocomposites reinforced by graphene and CNTs. A significant reduction in wear rate and coefficient of friction can be achieved in presence of CNTs and graphene up to a critical volume fraction. The reinforcements prevent a direct contact between two surfaces by forming a lubricant film between the contact surfaces. Beyond a critical volume fraction of CNTs and graphene, the wear rate and COF increases. At higher amount of reinforcement, the mechanical properties also decrease due to agglomeration that leads to the formation of some defects which ultimately decrease the tribological performance of the nanocomposites. Several material parameters, such as amount of reinforcements, size of reinforcement, and spatial distribution have an effect on mechanical and tribological properties. There is an optimum amount of reinforcement in the composites to have excellent mechanical and tribological properties.

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Figures

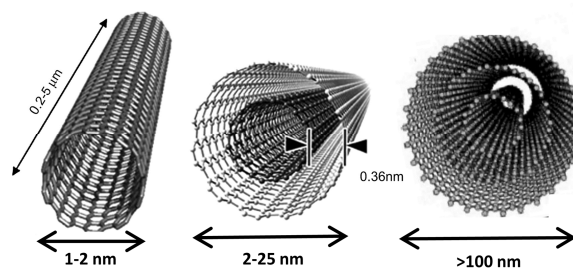
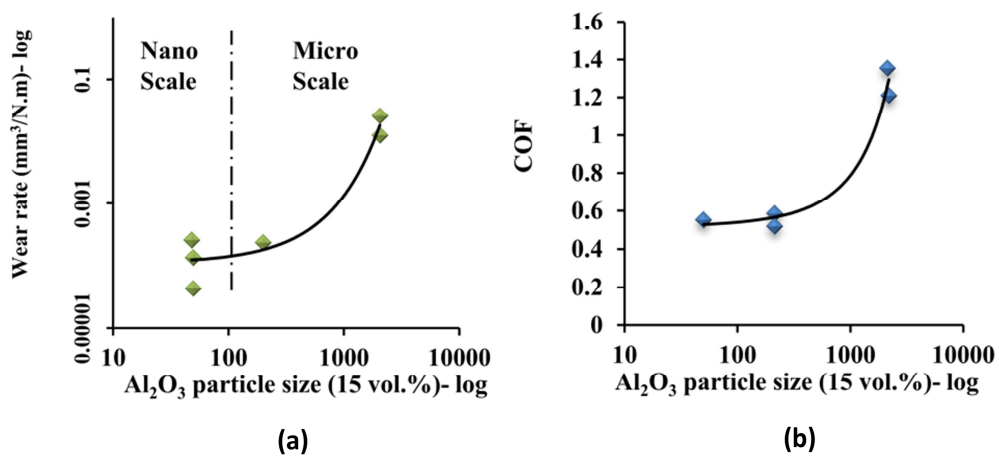


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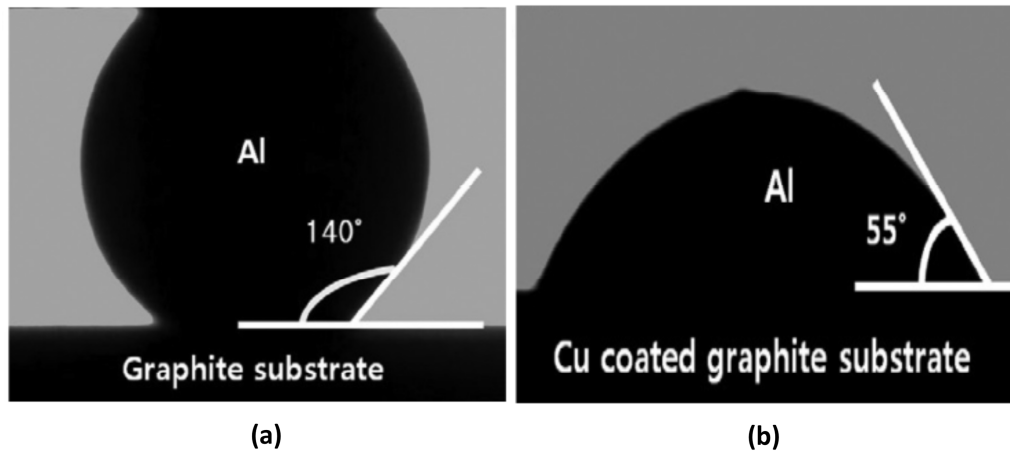


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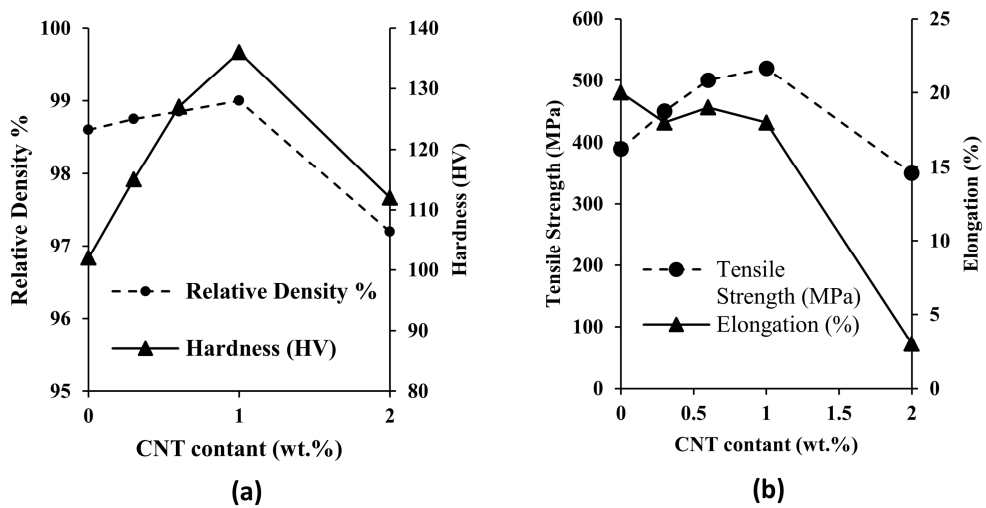


Figure 4 Variation of (a) relative density and hardness, and (b) tensile strength and elongation with carbon nanotube content for AA2024/MWNT nanocomposites[29]

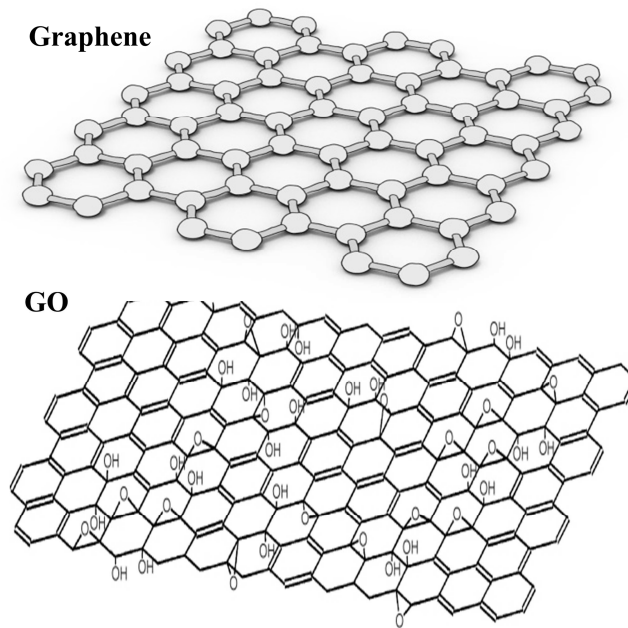


Figure 5 Surface structure of the graphene and GO

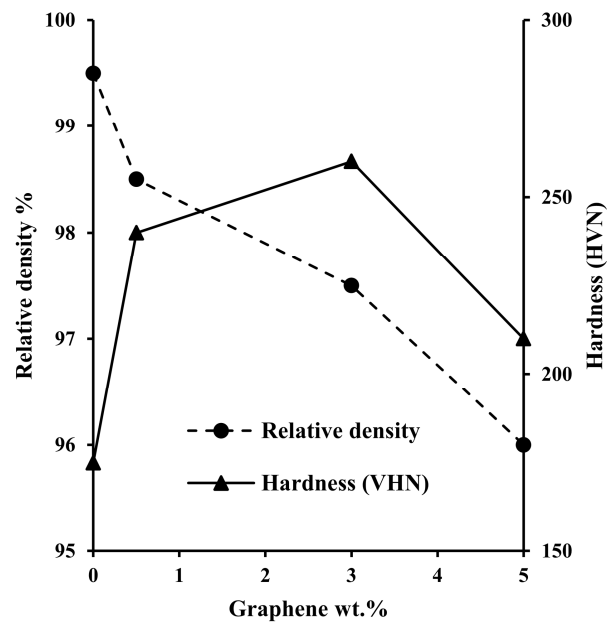


Figure 6 Relative density and hardness variations as a function of graphene addition to AA2124 nanocomposite[32].

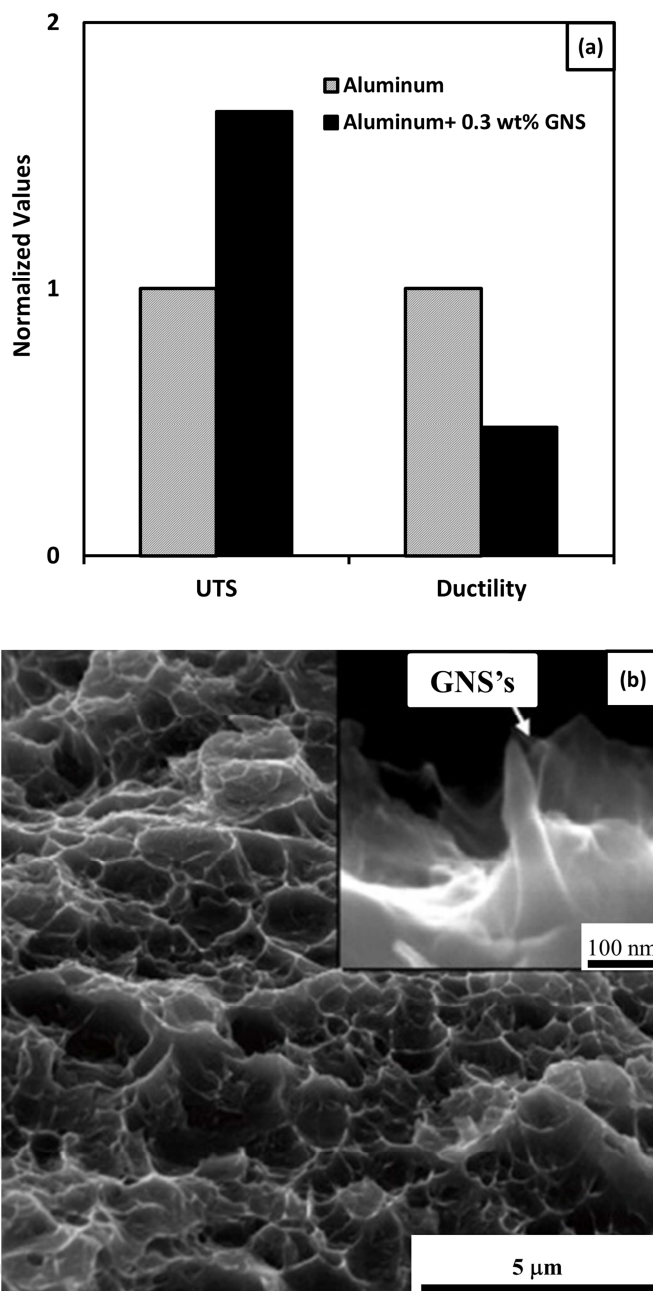
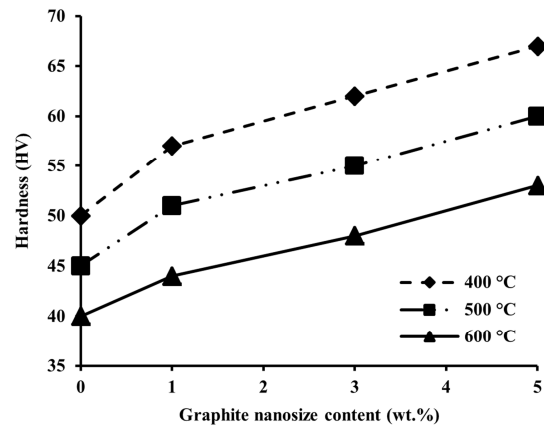
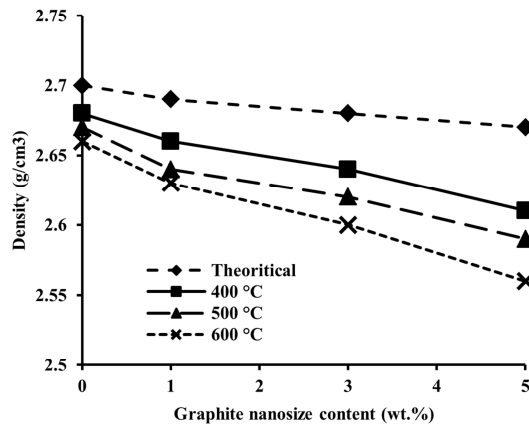


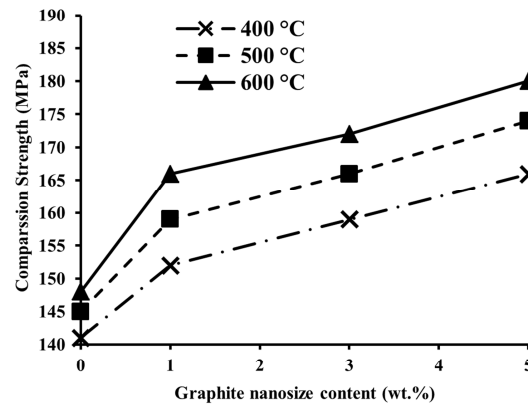
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(a)



(b)



(c)

Figure 8 Variation of (a) Vickers hardness, (b) Density, and (c) Compressive strength of aluminum alloys with exfoliated graphite nanoplatelets particles contents for GNP/Al composites at different sintering temperature[110].

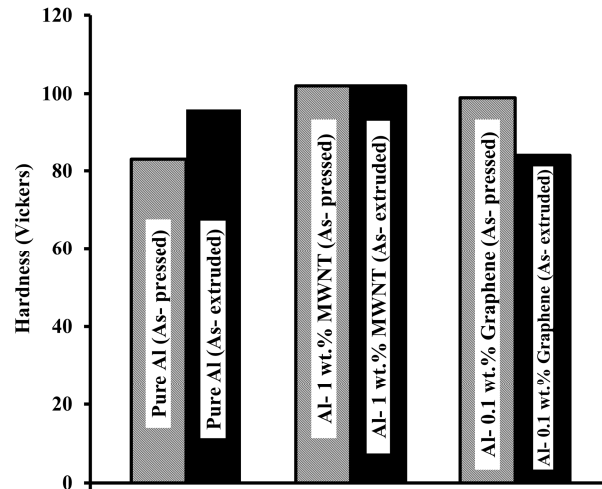


Figure 9 Vickers hardness data for the various materials and conditions[49].

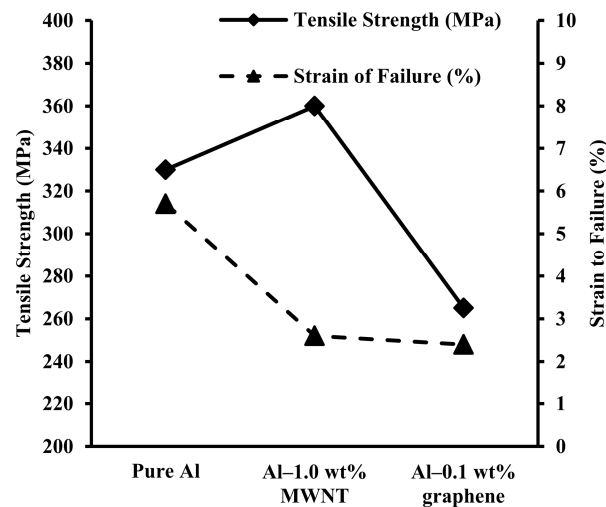
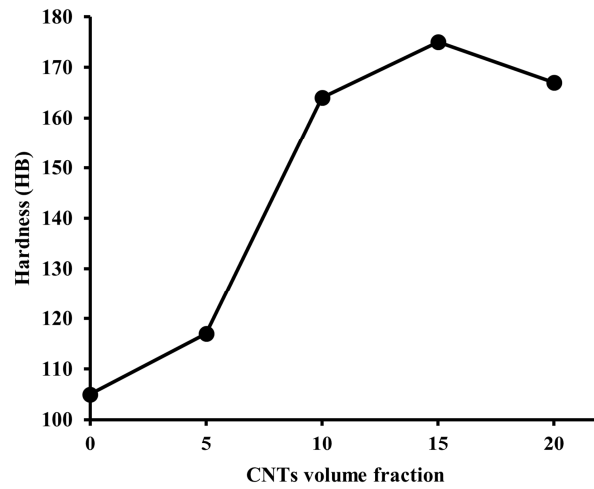
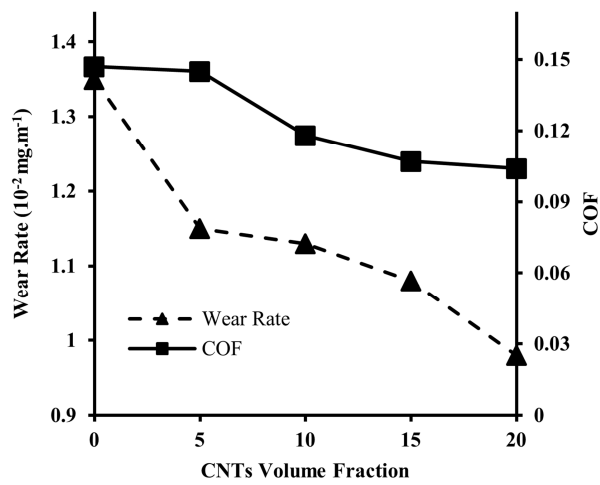


Figure 10 Ultimate tensile strengths and strain-to-failure of pure Al, Al-1 wt.% MWNT, and Al-0.1 wt.% graphene[49].

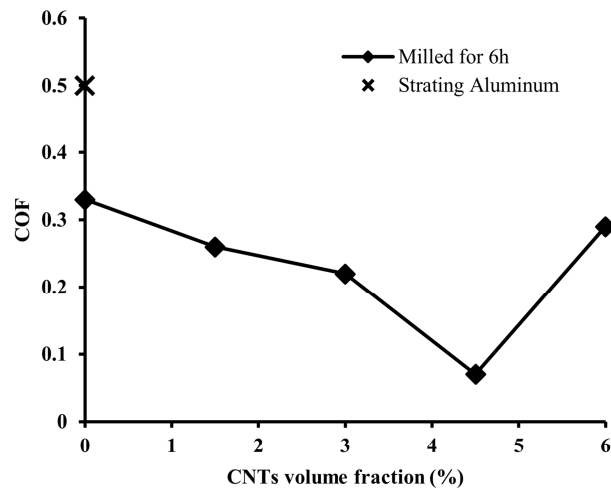


(a)

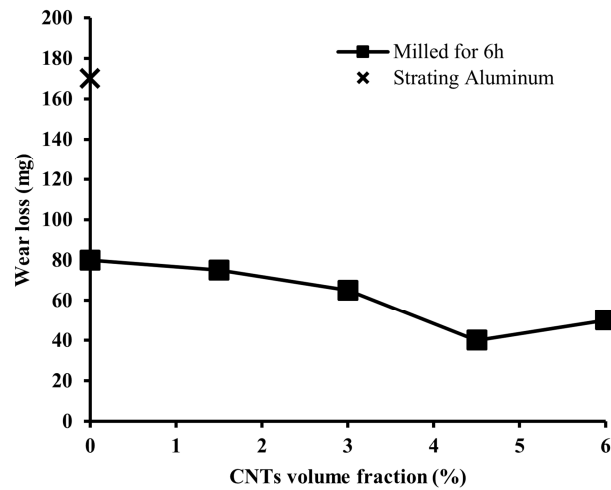


(b)

Figure 11 Variations of a) Brinell hardness (HB) b) wear rate and coefficient of friction with MWNTs content for melt-infiltrated MWNT/Al-Mg composites under an applied load of 30 N and a sliding velocity of 1.57 m/s [129]



(a)



(b)

Figure 12 Effect of volume fraction of CNT on friction coefficient and wear loss of Al/CNT nano composite at normal load 30N and sliding speed 0.12 m/s[125].

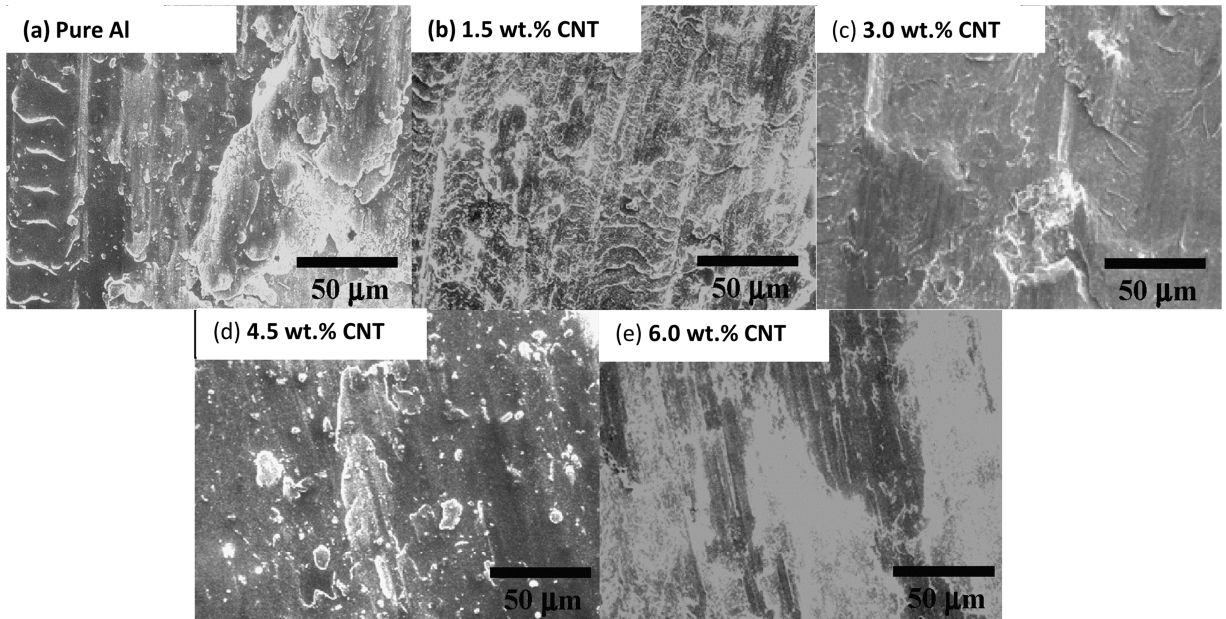


Figure 13 SEM images of wear tracks for (a) pure aluminum with a grain size of $\sim 150\text{nm}$ and composites containing MWCNTs of (b) 1.5 vol.%, (c) 3.0 vol.%, (d) 4.5 vol.%, and (e) 6.0 vol.%, respectively[125].

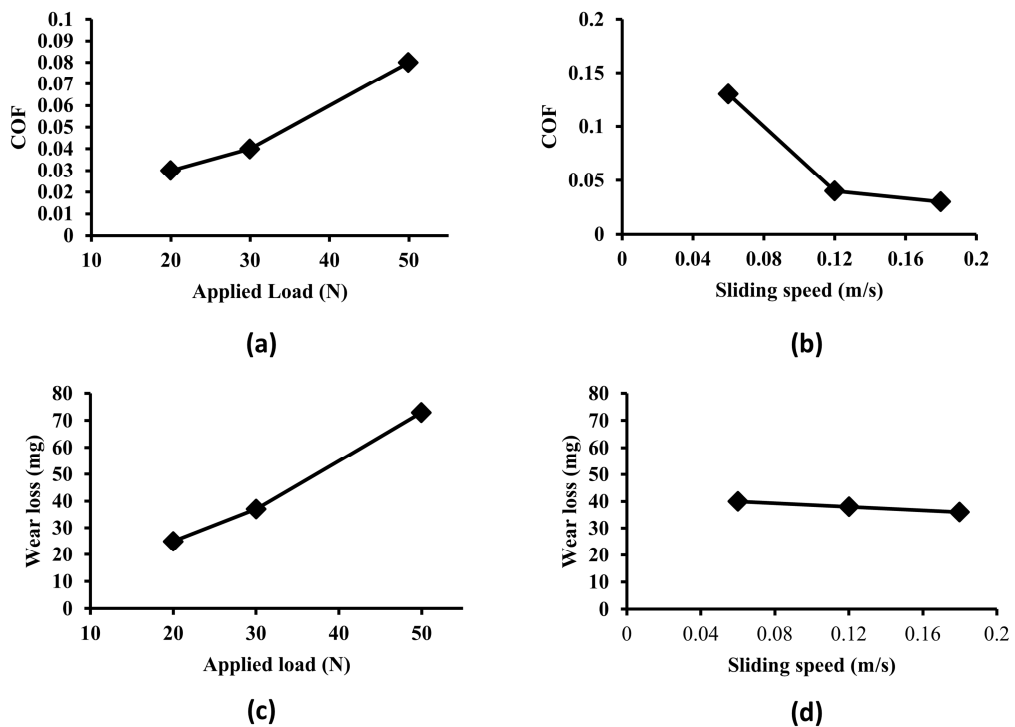


Figure 14 Variations of COF of 4.5 vol% MWNT/Al composite with (a) applied load at sliding speed 0.12 m/s and (b) sliding speed at applied load 30N. Variations of wear loss of 4.5 vol%

MWNT/Al composite with (c) applied load at sliding speed 0.12 m/s and (d) sliding speed at applied load 30N[125].

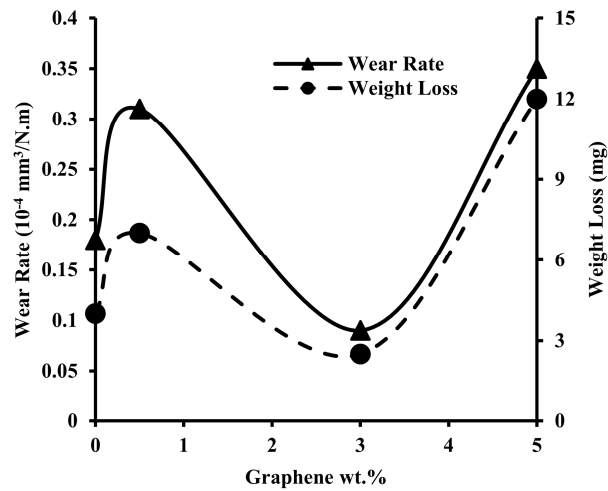


Figure 15 Wear rate and weight loss variation as a function of graphene content in AA2124 matrices[32].

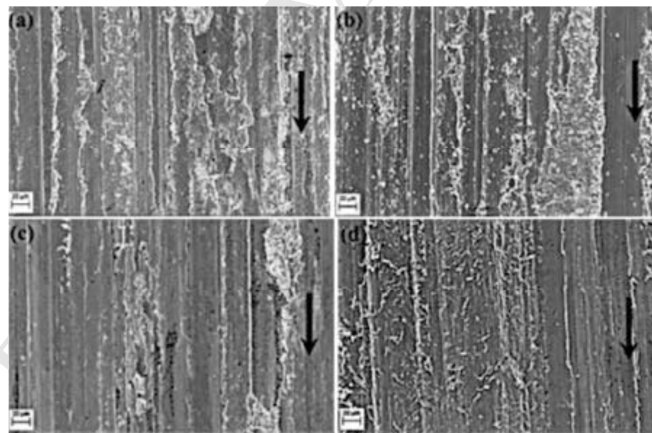


Figure 16 SEM micrographs of worn surfaces of AA2124 a) unreinforced, b) 0.5, c) 3 and d) 5 wt.% graphene nanocomposite[32].

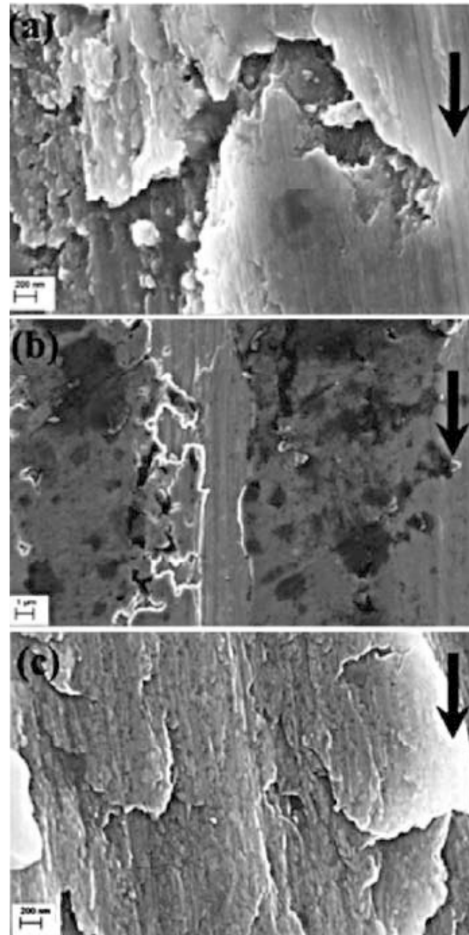
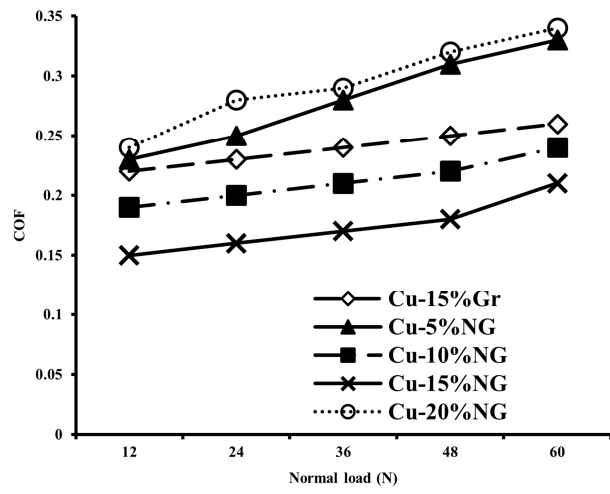
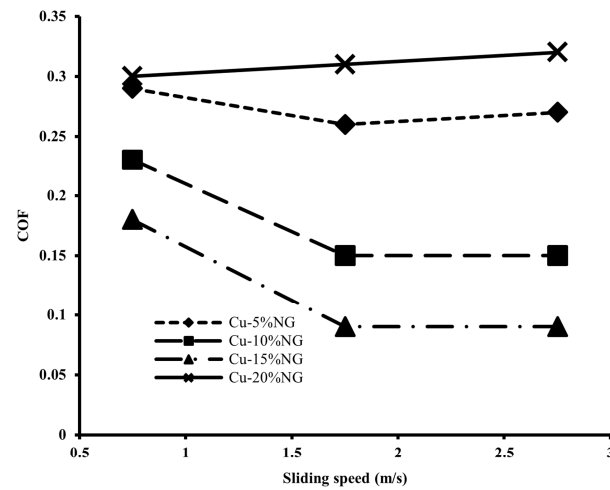


Figure 17 SEM micrographs for the worn surfaces of AA2124- a) 0, b) 3 and c) 5wt% graphene nanocomposite[32].



(a)



(b)

Figure 18 Variation of a) Coefficient of friction with normal load at sliding speed 0.77 m/s b) Variation of coefficient of friction with sliding speed at 36 N [30].

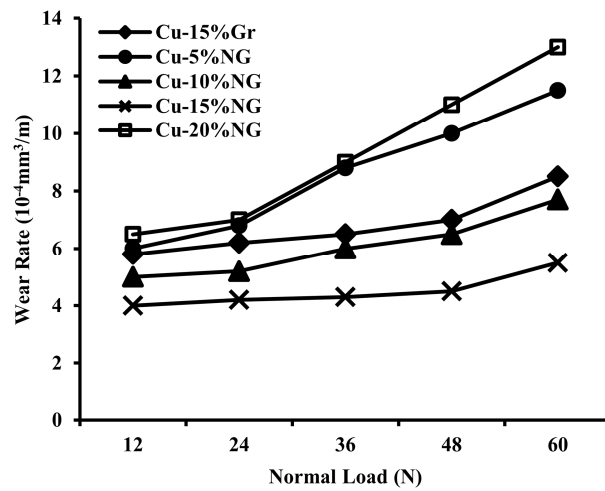


Figure 19 Variation of wear rate of composites with normal load at sliding speed 0.77 m/s[30].

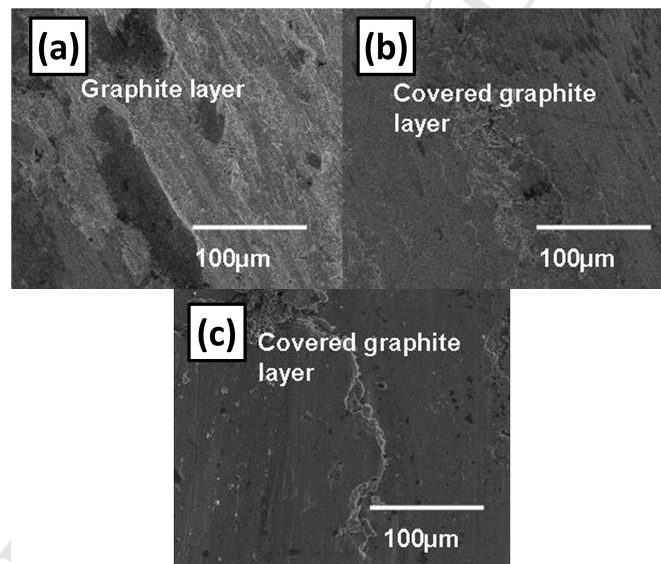


Figure 20 SEM image of worn surface of copper–15% nanographite composites at different sliding speeds at 36 N a) 0.77 m/s, b) 1.77 m/s and c) 2.77 m/s[30].

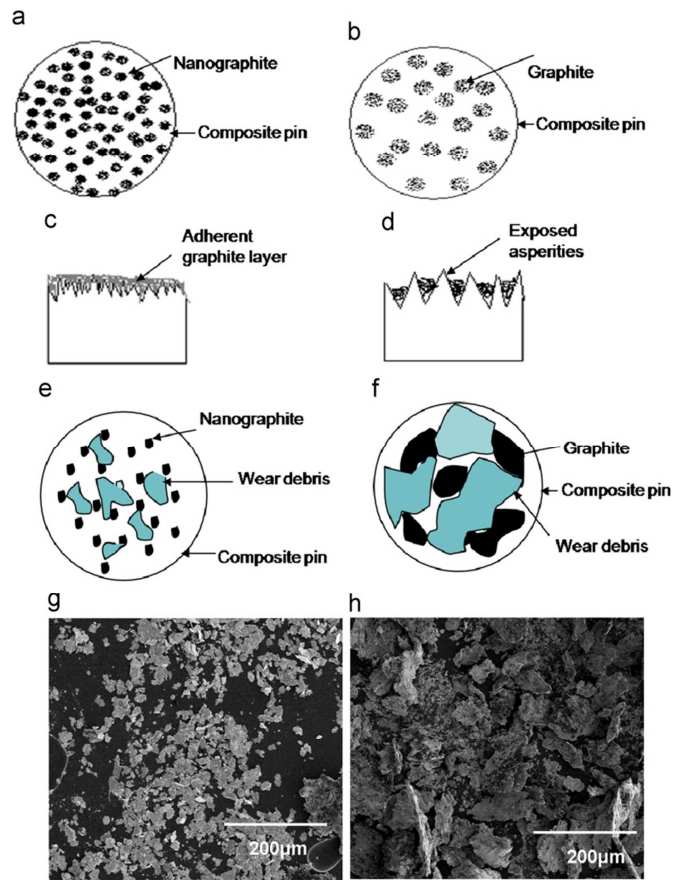


Figure 21 a) Distribution of nanographite in matrix, b) distribution of graphite in matrix, c) contact profile nanographite composite, d) contact profile of graphite composite, e) and f) conceptual wear generation model for nanographite and graphite reinforced composite respectively, g) and h) typical wear debris at 48 N and 0.77 m/s for copper–nanographite and copper–graphite respectively[30].