

Review Article

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Recent development in graphene-reinforced aluminium matrix composite: A review

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Abstract: Considerable attention has been given to graphene as a reinforcement material for metal matrix composite (MMC) because of its great potential for use in the automotive and aerospace industry. In general, the difficulty in achieving optimally improved properties can be attributed to poor wettability, agglomerations, and non-uniform distribution of reinforcement in the MMCs. Therefore, in terms of structural integrity, interfacial bonding, and its strengthening mechanism are important to achieve a high performance composite, which makes it imperative to discuss the integration of graphene into the alloy. The reinforcement mechanism of graphene-reinforced aluminium alloy has been evaluated in a limited number of studies, and this article examines current publications in this area. This article outlines three key topics related to the key challenges of graphene as a reinforcement material, the strengthening mechanism of graphene in a metal matrix, and the factors limiting the properties enhancement. Lastly, future works and recommendations addressed are summarized. The review presented aims to benefit to a wide range of industries and researchers and serve as a resource for future scholars.

Keywords: graphene, aluminium, strengthening mechanisms, metal matrix composite, interfacial reaction

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1 Introduction

Aluminium alloy has been used in various structural applications mainly in the aerospace and automotive industries because of its excellent properties, such as light-weight, excellent corrosion resistance, and high thermal resistance [1–5]; however, it has limited strength. The benefits of being lightweight are that it can facilitate a major increase in performance and fuel consumption. However, its limited strength has gained the attention of researchers who have sought to incorporate reinforcements such as boron [6,7], alumina (Al_2O_3) [7–9], silicon carbide (SiC) [10–14], graphene [15–19], and carbon nanotubes (CNTs) [20–23] to develop an excellent metal matrix composite (MMC). CNTs and graphene [10] are the most used carbon materials as compared to other reinforcements. CNT was discovered in 1991 by Iijima [24] and was followed by the discovery of graphene, a new carbon class, by Geim et al. in 2004 [9].

Graphene has been the subject of considerable attention since Novoselov et al. [9] discovered it and successfully isolated it from graphite through the tape stripping route. Graphene is a two-network carbon nanomaterial that forms a unique honeycomb lattice structure through sp^2 hybridisation. This material has excellent physical and chemical properties because it is harder than diamonds and 100 times stronger than steel with a great Young's modulus (1 TPa), high thermal conductivity ($5.3 \text{ kW}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$), and excellent ultimate tensile strength (UTS) (130 GPa) [9]. The density of graphene is also estimated to be as low as $1.06 \text{ g}\cdot\text{cm}^{-3}$ [25] which makes it extremely light. These characteristics have led to the selection of graphene as an effective reinforcement for MMC.

In comparison to CNTs, graphene has a better improvement in terms of dispersion and bonding with the matrix. The properties of graphene and CNT are listed in Table 1 [26–28]. The specific surface area of graphene and CNT are 2,630 and $1,315 \text{ m}^2\cdot\text{g}^{-1}$, respectively. Graphene has higher specific surface area and a lower tendency to twist allowing

Table 1: Physical and functional properties of graphene and carbon nanotubes [26–28]

Properties	Young's modulus	Ultimate tensile strength	Thermal conductivity	Density	Specific surface area
Unit	TPa	GPa	$\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	$\text{g}\cdot\text{cm}^{-3}$	$\text{m}^2\cdot\text{g}^{-1}$
Graphene	1	130	5,300	1.06	2,630
CNT	0.9	63	3,000	1.3–1.4	1,315

it to avoid dispersion, which results in effective load transfer and an increase in strength and stiffness [29–32]. As reported in ref. [33], even with the same amount of graphene and CNT, CNT has an issue with agglomeration because of its higher aspect ratio as compared to graphene. Graphene also has a lower density than CNT, 1.06 and $1.3\text{ g}\cdot\text{cm}^{-3}$, respectively. Research on graphene has grown rapidly over the last decade due to these interesting characteristics.

However, proper dispersion of graphene without damaging the structure is the main challenge that needs to be addressed to achieve enhanced performance [32]. The insolubility of graphene in the matrix is due to the Van der Waals attractive forces and the stacking of pi between the graphene lamellae [32,34]. The weak bonding between the matrix and graphene makes it difficult to bond. Hence, enhancing the interface bonding between aluminium and graphene is important to attain good performance of the composite.

Other factors, such as fabrication technique, materials of matrix dispersion, and particle geometry also play significant roles in the dispersion of the graphene in a metal matrix [35]. The formation of carbide and interfacial interactions also contribute to the strengthening of its efficiency or stress transfer [36]. Carbide formation can be attributed to Gibb's free energy thermodynamically. The conversion of sp^2 to sp^3 bonded carbon acts as a favourable site for carbide formation, which may positively or adversely affect the final composite properties. Therefore, many researchers have attempted to combine several processing techniques to obtain homogenous graphene dispersion in the composite.

In general, many studies have highlighted the effect of graphene reinforcement on the mechanical properties of Al matrix, particularly its tensile strength, hardness, and flexural strength [8,25,37]. However, limited studies have discussed the effects of graphene reinforcement in Al matrix on the strengthening mechanism to predict the behaviour of graphene dispersion. The suggested strengthening mechanism for graphene-aluminium matrix composite (AMC) include grain refinement, Orowan strengthening mechanism, stress load transfer, and thermal expansion mismatch.

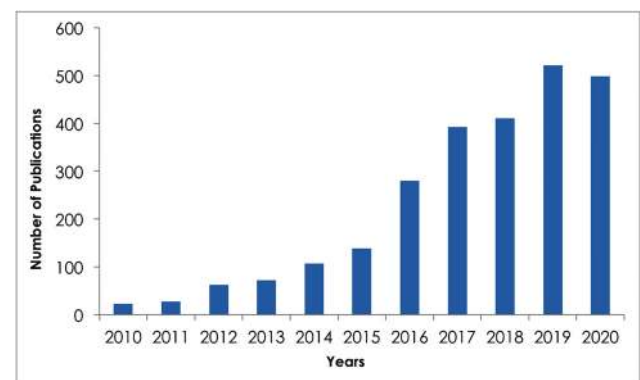
Based on a search on the Web of Science databases, a significant trend can be observed among research papers published with the related keywords of graphene in the field of AMC within the last ten years as illustrated in Figure 1. The increasing trend indicates the interest in scientific research of graphene as a potential reinforcement material in AMC.

This field of research requires systematic study to provide a clear pathway for graphene in AMC fabrication. Therefore, this review article will summarise the brief description of graphene and its derivatives, and the challenges on graphene as reinforcement. Then, it will highlight the strengthening mechanism and its effects on the distribution and dispersion of graphene. Finally, this article summarises the factors that limit the properties enhancement of graphene in AMC.

2 Graphene as reinforcement

2.1 Derivatives of graphene

Graphene has several derivatives, which have been used in published literature, including single-layer graphene, graphite, graphene nanoplatelets (GNPs), graphene oxide (GO), and reduced graphene oxide (rGO). These derivatives

**Figure 1:** Number of publications of Graphene in AMC between years 2010–2020 based on Web of Science database.

related directly to carbon lattice and functional groups, have many layers and crystallographic structures, as shown in Figure 2. For several decades, graphene has been known as the basic structural unit of bulk graphite and is physically unstable. In 2004, Geim et al. successfully performed the fabrication of graphene using the micromechanical cleavage method. This method involves separating the single layer of graphene from graphite by using scotch tape.

2.2 Challenges of graphene

The challenges of using graphene as reinforcement include poor dispersion, low interfacial bonding, carbide formation, and low structural integrity [39]. Poor dispersion is caused by the variations in the bonding properties of graphene and Al. Al has metallic bonding, while graphene has Van der Waals and covalent bondings [40–42]. The agglomeration issue is also prone to occur due to the strong interplanar Van der Waals forces interacting between graphene sheets in a metallic matrix, which has a detrimental effect on the mechanical properties. The agglomeration affects the composite, causing significant porosities and premature failure of the composite. They have a great tendency to wrinkle or form clusters because of Van der Waals forces. These clusters are source of pores, cracks, or pinholes that may cause premature failure of the composite [31].

Several studies have used the MMC on graphene dispersion, but these studies are still in the early stages and non-homogeneous dispersion remains the main concern. The most common processing method used to fabricate graphene in AMC is powder metallurgy (PM) [21–23] because of the simplicity of the process followed by a secondary processing method. Other commonly used secondary processing methods include hot rolling, stir casting, and ultrasonication to obtain high-density composite with minimal porosity [24–27]. To evaluate the graphene dispersion in the Al matrix, Bastwros et al. [43] used PM accompanied by hot compaction in a semi-solid system. At 1 wt% of graphene and 60 min ball milling times, they recorded an improvement in flexural strength by 47%. Meanwhile, Niteesh Kumar et al. [44] used the same direction of PM route followed by hot extrusion technique to obtain uniform dispersion and excellent interfacial bonding between Al and graphene. Their result shows an increment of 11.8% in hardness and 11.1% in UTS as compared to pure Al alloy.

Fadavi Boostani et al. [45] manufactured Al-SiC, which encapsulated graphene nanosheets through non-contact ultrasonic to avoid cluster. This method effectively avoided the agglomeration of nanoparticles and increased the dispersion of the graphene sheets. Baig et al. [36] produced GNP-reinforced Al composite through ball milling and sonication method. The 0.3 wt% of GNPs/Al samples demonstrated a maximum improvement in hardness

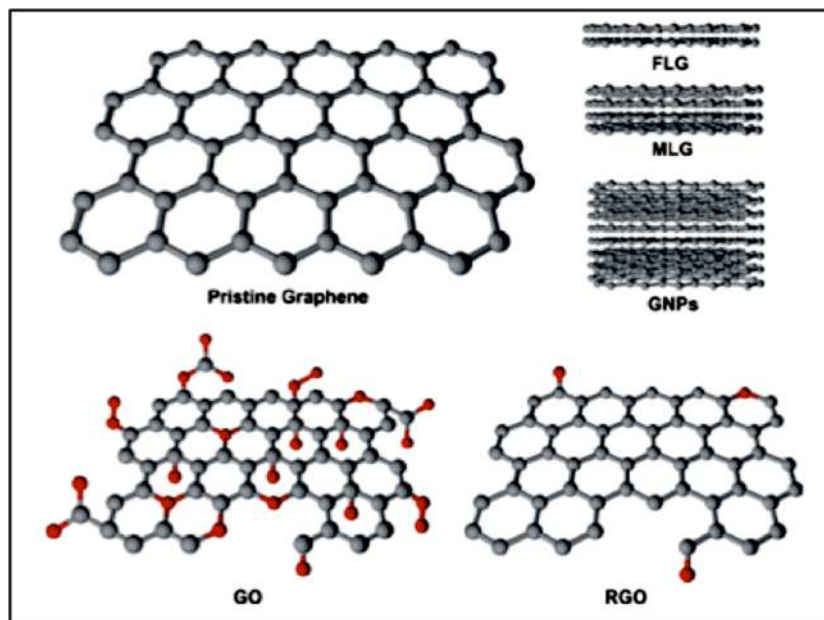


Figure 2: Graphene derivatives where grey sphere refers to carbon atom and red spheres refer to functional groups such as hydroxyl ($-\text{OH}$), alkoxy ($\text{C}-\text{O}-\text{C}$), and carboxylic acid ($-\text{COOH}$). Reproduced from ref. [38].

(35.61%) and a decrease in wear rate (76.68%) as compared to pure Al at 300 rpm milling. Studies on mechanical properties have focused predominantly on the consistency of the composite interface. The consistency of the interface quality relies on wetting, structural integrity, and interface reaction as well as carbide formation [46].

Structural integrity has a significant effect on strengthening efficiency. Generally, the fabrication technique includes PM, casting, sonication, and additive manufacturing. A common route in graphene AMC fabrication is PM because of its ability to disperse homogeneously and potential for scale-up production. However, the drawback for this method is the major detrimental effect on the structural integrity that will affect the mechanical properties because of the strong relationship between structural damage and strengthening efficiency [47]. The effect of ball milling time on the graphene structure in AMC have been reported in several published studies [43,48–50]. The findings show the intensity ratio of D-band to G-band (I_D/I_G) increased from 1.1 to 1.4 from prolonged ball milling time, which indicates high defect in the graphene structures [51,52]. Meanwhile, Shao *et al.* [52,53] compared GO and GNP with Al5083 and found that GNPs have a lower defect in structure as compared to the GO based on (I_D/I_G) which are 0.31 and 0.84, respectively. Therefore, they concluded that GO was more prone to damage in the structure. Composites reinforced with GO or RGO are notably more prone to interfacial reactions between Al and carbon than those reinforced with GNP.

The interfacial reaction between graphene and Al creates the aluminium carbide (Al_4C_3) formation because of the low Gibbs free energy $-196 \text{ kJ}\cdot\text{mol}^{-1}$ at 298 K [39]. The reaction between carbon and Al is thermodynamically

a spontaneous reaction that makes it difficult to control the interface reaction within graphene-reinforced AMCs. Interestingly, the Al_4C_3 phase effects of graphene in AMC mechanical characteristics are still under discussion.

Several authors have suggested that the occurrence of Al_4C_3 enhanced the interfacial bonding between graphene and Al. Jiang *et al.* [39] reported the nucleation and growth mechanism of Al_4C_3 in their present work. They discovered that nucleation begins at the open edge and defect site of graphene because of high chemical reactivity. The growth of Al_4C_3 was controlled by its crystal structure characteristics. The Al_4C_3 particles are located along or at an angle to the Al grain boundary, and several orientation relationships between Al_4C_3 and Al were observed, as shown in Figure 3. This relationship demonstrated that altering the preferred orientation of the Al matrix can enhance interfacial bonding.

Meanwhile, Xiong *et al.* [54] reported that they found a small amount of carbide formation that provides an interface between the matrix and reinforcement for wetting and bonding. This may be effective in providing shear resistance and serving as anchors to avoid serious damage to the graphene structure. In the analysis, the effect of Al_4C_3 on the strengthening effect has been summarised based on several factors. First, the size of the Al_4C_3 diameter with a mean diameter of 30 nm is larger than the GNP thickness that contributes to the change in the mechanical bonding to strong chemical bonding. Second, the well-distributed Al_4C_3 in near contact with GNP provides an anchor influence between the Al matrix and GNP. This anchoring effect tends to improve the interface load transfer [55]. Thus, it can also be concluded that this sub-product has a positive effect on strength enhancement.

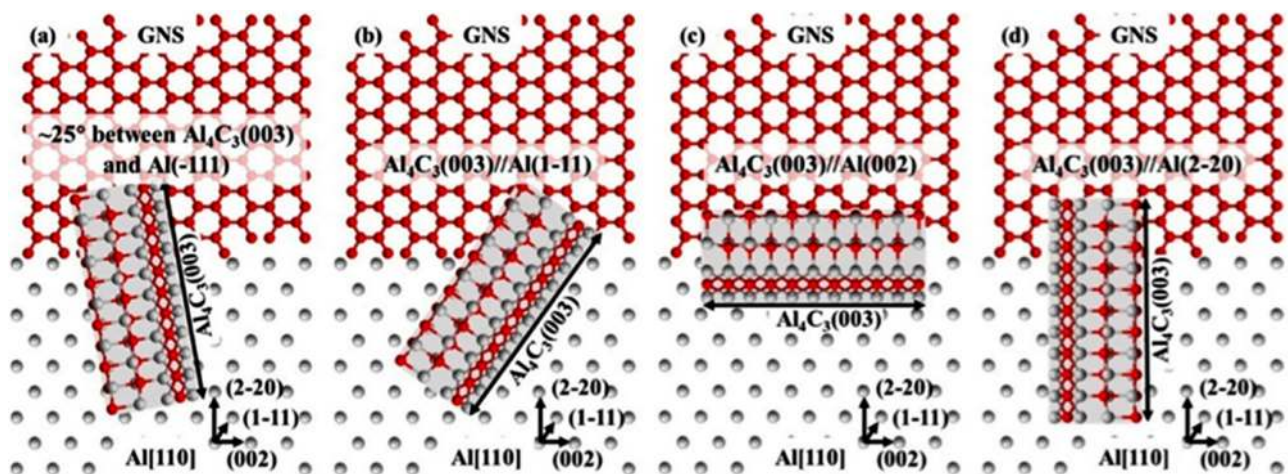


Figure 3: The nucleation and the growth mechanisms of Al_4C_3 where (a) $\sim 25^\circ$ angle between $Al_4C_3(0003)$ and $Al(-111)$; (b) $Al_4C_3(0003)//Al(1-11)$; (c) $Al_4C_3(0003)//Al(002)$, and (d) $Al_4C_3(0003)//Al(2-20)$. Reproduced from ref. [39].

Other reports have also suggested that Al_4C_3 formation might decrease mechanical properties because of the induction of microcracks, resulting in the premature failure for Al/graphene composite [40,56–58]. Etter et al. [58] reported that Al_4C_3 is related to surface defects of the carbon fibres and their presence in the structure or edges at graphitic plane will degrade the fibres, thereby reducing the mechanical properties [57,59]. Carbide has hydrophilic characteristics that are sensitive to moisture contact that will induce the fatigue crack growth rate to accelerate and as a consequence, reduce yield strength [58]. Hence, they suggested two methods to overcome the carbide formations in which the reinforcement is coated with an inert layer. The inert layer coat will turn into a diffusion barrier between the carbon and Al. The second method is to introduce another element to the Al alloy to reduce the solubility of carbon atoms in Al alloy, such as Silicon (Si) because Si is a known element to reduce the carbide formation.

Bartolucci et al. [57] reported that carbide formation was not beneficial to graphene reinforcement. They mentioned that carbide reduces strength and causes a decline in mechanical properties. They also suggested that graphene was prone to carbide formation because of the higher surface area and 2D geometry. The 2D geometry can be attributed to the lower and upper surfaces available for severe defects during milling and prismatic planes or edges of graphene also can be a favourable site for defects and reaction with the matrix.

The fabrication method can reduce carbide formation because of the processing condition as reported by other researchers. Saboori et al. [40] compared two fabrication techniques to evaluate the carbide formation on 0.5 and 1.0 wt% of GNP/Al via hot rolling technique and conventional press sintering method. They found that no significant carbide formation occurred because of the insufficient level of defects in the GNP/Al composites, which may be attributed to the low sintering temperature, size, crystallinity, and low percentages of the GNP.

In summary, the occurrence of Al_4C_3 enhancing the interfacial bonding between Al and graphene is still under debate. Some reports have mentioned that Al_4C_3 improved the load transfer capacity of graphene and provided a strengthening effect that contributed to the enhancement in strength. The formation of Al_4C_3 is also related to the integrity of graphene in which the worse the integrity of the graphene, the more likely the formation of Al_4C_3 . Meanwhile, other reports have found that Al_4C_3 causes premature failure because of the nucleation of microcrack and acts as detrimental interfacial phases. Nevertheless, the parameter affecting Al_4C_3 formation is

well worth of addressing and optimizing in future works to attain enhanced properties because they may have positive or negative consequences on the strength at certain conditions.

3 Strengthening mechanism in graphene-Al composite

This section will address the strengthening mechanisms that describe the strengthening effect of graphene in AMC. In general, two key features of the reinforcement strengthening system can be identified, namely, indirect strengthening from reinforcement and direct strengthening from metal matrix to reinforcement. Direct strengthening can be accomplished through hard reinforcement in the soft matrix. The applied load is moved from the matrix to the reinforcement because of the hard reinforcement in the matrix, which increases the composite resistance to plastic deformation during external loading [61]. Meanwhile, indirect strengthening is related to thermal expansion mismatch between the higher thermal expansion coefficient (CTE) of the matrix and lower CTE of reinforcement. As the temperature increases, the thermal stresses result in the formation of dislocation at the matrix/reinforcement interface. The increase in dislocation density contributes to an improvement in the strength of the composite material. An increase in the reinforcement and decrease in the particle size of the reinforcement material have also been found to contribute to an increase in the dislocation density and an increase in strength [1].

In the case of AMC, the strengthening mechanism can be attributed to the following: (1) Orowan looping of nanoparticles, (2) load transfer from Al matrix to graphene, (3) dislocation strengthening through the CTE mismatch, and (4) grain refinement strengthening. Strengthening of the graphene MMC depends on the processing method, interface quality, dispersion at grain boundaries, and their intrinsic characteristics such as grain size, aspect ratio, and geometry. Hence, understanding the strengthening mechanism that affects the composite properties is important.

3.1 Orowan looping of nanoparticles

Orowan looping predicts the effect of interference caused by the smaller size uniform particles within the dislocation path. It is not significant if the particle is microsize because the interparticle spacing is large. However, if

nano-sized particles are used as reinforcement, such as GNPs, the Orowan looping mechanism provides a dominant strengthening effect. Generally, the nano-size of the reinforcement responds to the dislocation results in Orowan looping [54].

The graphene nanoparticles play a role as a barrier that hinders the dislocation motion and avoids the dislocation pile-up. Hence, the dislocation loops that have been successfully generated pass through the particles and create enough back stress that drives the dislocation motion because they tend to bend, which results in the formation of semicircular shapes in graphene addition. This mechanism is known as Orowan looping in which the back stress helps in improving the composite strength. The strength gained through this mechanism can be formulated as in equation (1)

$$\Delta\sigma_{\text{Orowan}}(\text{MPa}) = \frac{\alpha G b}{d_p \left[\left(\frac{1}{2} f_v \right)^{\frac{1}{3}} - 1 \right]} \ln \frac{d_p}{2b}, \quad (1)$$

where α is the Taylor factor, G and b are shear modulus and the burger factor of the matrix, respectively, d_p is the average distance between the reinforcements, and f_v is the volume fraction.

Apart from particle size, uniform dispersion plays a significant role in the strengthening mechanism. The graphene particles need to disperse homogeneously within the microstructure to effectively hinder the movement of dislocation. However, the graphene content will agglomerate at a certain threshold, leading to mechanical properties deterioration that needs to optimise the number of additives [32,45,62].

A major analysis and discussion on the topic raised by Xiong *et al.* [54] indicated that GNPs and Al_4C_3 play important roles in the Orowan looping mechanism because of its nano-size. They investigated the strengthening effect from the interfacial reaction in different volume fractions

(0.3, 0.6, 0.9, and 1.2 vol%) of GNP-reinforced Al alloy via spark plasma sintering. The interfacial reaction changed from mechanical bonding to a strong chemical bonding because the Al_4C_3 phases are tightly locked into the Al matrix, which then acts as an anchor, as can be seen in Figure 4. The figure shows TEM images of the interface in GNP/Al composite that indicate the nano-sized Al_4C_3 phase with a mean diameter of 30 nm. Based on the results, they proposed that Al_4C_3 and GNP should be considered as a whole in calculating Orowan strengthening because of three main reasons: (1) the Al_4C_3 are tightly linked to GNP, (2) their sizes are within nanoscale, and (3) Orowan strengthening is independent of mechanical property of the nanoparticle. Thus, it can be concluded that a well-distributed Al_4C_3 has an anchor effect between GNP and Al matrix that causes the interfacial bonding to change from mechanical to strong chemical bonding. The measurements in Orowan looping calculation must also be treated as a whole because of their nanoscale size.

Orowan strengthening mechanism has also contributed the most in strengthening the composite as reported by Bisht *et al.* [32]. They prepared 0.5, 1, 3, and 5 wt% of GNP/Al by spark plasma sintering method at a pressure of 50 MPa and at 550°C with a holding time of 40 min. The increment of yield strength and tensile strength by this method has reached up to 84.5 and 54.8%, respectively, with the addition of 1 wt% of GNP. The TEM micrograph result shows that GNP was properly dispersed up to 1 wt %, while increase in the GNP content causes agglomeration, which eventually reduces the strength and ductility. Because of the low temperature and short time for consolidation, they discovered that Al_4C_3 is not present. In comparison to other mechanisms, the Orowan strengthening mechanism is the most dominant in the reinforcing effect of GNP in the Al matrix. The yield strength calculated through the Orowan looping mechanism matches that in the experimental data as shown in Figure 5. This

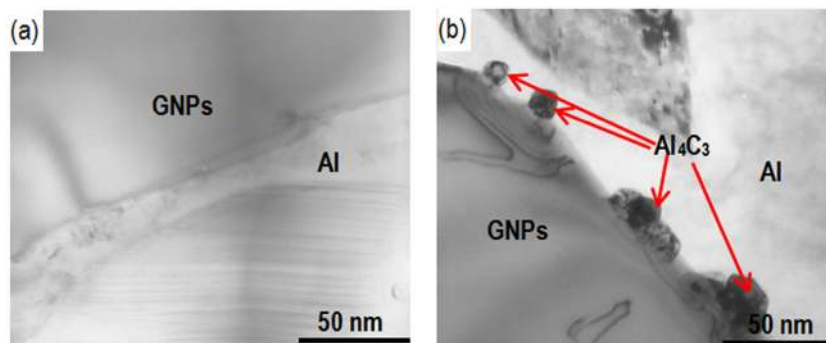


Figure 4: TEM images of interface in GNP/Al composites: (a) 0.3 vol% of GNP/Al and (b) 1.2 vol% of GNP/Al. Reproduced from ref. [54].

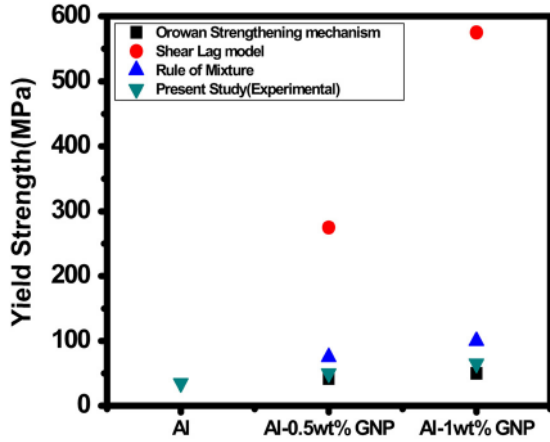


Figure 5: Comparison of yield strength (MPa) calculated from various strengthening mechanism models and the experimental data. Reproduced from ref. [32].

result strongly indicates that a smaller size and uniformly distributed GNP posed a hindrance to the dislocation loops that significantly enhance the composite strength.

3.2 Load transfer from Al to graphene

Load transfer can be explained by the shear lag model [54]. The shear lag model defines the load transfer from the matrix to the reinforcement and described a strengthening mechanism with a high aspect ratio. Interfacial shear stress and strength are two major effects that come from the load transfer and depend highly on interfacial bonding between matrix and composites [63]. The shear lag model, which calculates the UTS $\Delta\sigma_{load}$ as in equations (2) and (3), was introduced by Kelly and Tyson [64,65].

$$\Delta\sigma_{load} = \sigma_{re}V_{re}\left(\frac{1}{2l_c}\right) - \sigma_mV_{re} \quad \text{for } l \leq l_c, \quad (2)$$

$$\Delta\sigma_{load} = \sigma_{re}V_{re}\left(1 - \frac{1}{2l_c}\right) - \sigma_mV_{re} \quad \text{for } l > l_c, \quad (3)$$

where σ_{re} , σ_m are tensile strengths for reinforcement and matrix, respectively, V_{re} is the volume fraction for reinforcement, and l_c is the critical length.

The critical length (l_c) was also introduced in this model as according to equation (4). During tensile tests, loads may be transferred from the metal matrix to GNPs via interfacial shear stress (τ_m) along the tensile direction. Given that the length of GNP exceeds the critical length ($l \geq l_c$) determined by equation (3), the graphene will fail through the fracture. Meanwhile, when the length

of GNP is shorter than the critical length ($l < l_c$), the GNP will be pulled out through interfacial de-bonding.

$$l_c = \frac{d\sigma_{re}}{2\tau_m}, \quad (4)$$

where σ_{re} is the tensile strength for reinforcement, τ_m is the matrix shear strength, and d is the diameter of reinforcement.

The criteria for the composite must be either short fibres or whiskers that have an intimate contact between reinforcement and matrix and the alignment must be in a single direction. Therefore, graphene is assumed to align along the tensile direction for this model.

Li et al. [66] applied the PM route and heat treatment on 2.0 wt% of GNP on matrix 1060 Al. The effects of heat treatment on the interface reaction, interface relation, and enhancement of GNP/Al composite performance were evaluated. From their previous work, they found that load transfer could be attributed to the enhanced mechanical properties and relied heavily on the strong interface bonding. With the increasing annealing temperature between 630 and 650°C, the composite fracture mode has progressed from GNP pull out to GNP fracture, which implies improved interfacial bonding strength. Thus, the strength significantly increased. The shear-lag model-projected UTS agreed well with the experimental results, indicating that load transfer played a significant role in the composite strength. Thus, because of the increased interfacial bonding between GNP and Al matrix, the strengthening performance of graphene was significantly improved with increasing annealing temperature.

3.3 Dislocation strengthening via CTE mismatch

The mismatch of CTE has a significant effect on the strengthening mechanism due to density dislocation. This mechanism is commonly known as thermally activated dislocation. Proper graphene distribution in the Al matrix inhibits dislocation motion in the Al matrix, resulting in high dislocation density at the Al-graphene interface. Because the dislocation density depends on the surface area of the particle reinforcement, the smaller the particle size, the higher the surface area. Dislocation density will be higher because of the small particle size of graphene reinforcement. Meanwhile, the smaller particle size will cause a high surface area that results in the increment in strength because of the effectiveness in increasing the number of dislocations created [66].

Graphene has a lower CTE than Al, which are $1 \times 10^{-6} \text{ K}^{-1}$ and $23.6 \times 10^{-6} \text{ K}^{-1}$, respectively. The large CTE mismatch between graphene and Al induces prismatic punching of dislocation in the composite, which strengthened the composite. This mechanism can be formulated as in equation (5)

$$\Delta\sigma_{\text{CTE}} = \alpha G b \sqrt{\frac{12\Delta T \Delta C f_v}{b d_p}}, \quad (5)$$

where α is a constant value (1.25), G and b are the shear modulus and burger vectors of the Al matrix, respectively, ΔT is the temperature difference between Al matrix and graphene, ΔC is the CTE difference between Al matrix and graphene, f_v is the volume fraction of graphene, and d_p is mean particle size of graphene.

Prior studies have compared and selected the major strengthening mechanism based on the match between the theoretical and experimental values. A recent study by Fadavi Boostani *et al.* [63] evaluated the effect of SiC nanoparticles encapsulating graphene sheets and A357 alloy through the thixoforming method. Two different shapes for graphene sheets were characterized to prevent agglomeration such as onion-like graphene sheets (OLGS) encapsulating SiC particles and disk-shaped graphene sheets (DSGS). Orowan looping, shear lag, and dislocation density mechanism were used to evaluate the most strengthening contribution factor. The result in Figure 6 shows that the thermally activated dislocation contributed the most for strengthening the composite due to negative thermal expansion coefficient of graphene sheets. The yield strength and tensile strength were enhanced by up to 45 and 84%, respectively. They explained that the reason for this is the pinning capacity of nano-sized rod-like Al_4C_3 was triggered, which led to a strong interface bonding for SiC nanoparticles with the matrix.

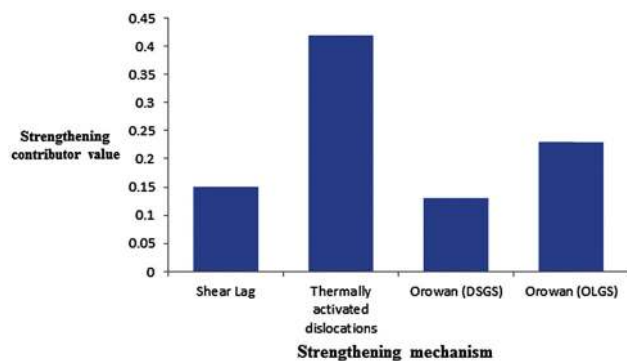


Figure 6: Comparison of strengthening mechanism factor on enhancing the tensile yield strength of shear lag, thermally activated dislocations, Orowan looping disc-shaped graphene sheets (DSGS), and Onion-like graphene sheets (OLGS). Reproduced from ref. [63].

Rashad *et al.* [30] reported a minimum increment in UTS from 252 to 280 MPa as a result of the dislocation density after the addition of 0.3 wt% of GNP to the Al matrix. The composites did not perform up to the expectation at higher GNP content because of the agglomeration of GNP. The scanning electron microscopy (SEM) images of pure Al and 0.3 wt% of GNP presence are illustrated in Figure 7. Cavities and black holes were discovered, resulting in a modest increase in tensile strength and the introduction of fracture. The presence of GNP particles caused obstacles to the movement of Al. The movement was hindered by GNP particles that cause piling up in the dislocation during their motion, which strengthened the Al-GNP composite.

Another reported study from Tang *et al.* compared the strengthening mechanism between the graphene sheet and encapsulating graphene in SiC particulate on the Al matrix [68]. They found that thermal mismatch is the most contributed mechanism to attain high strength up to 140 MPa, followed by Orowan strengthening (40 MPa), and fine grain was the last (15 MPa) as shown in Figure 8. They employed finite element analysis (FEA) to investigate the strengthening effect in enhancing the tensile strength of encapsulating graphene in the Al-SiC composite. By changing the theoretical strengthening model, FEM simulation is well matched with the experimental data and contributes to understanding the most dominant strengthening mechanism for strength.

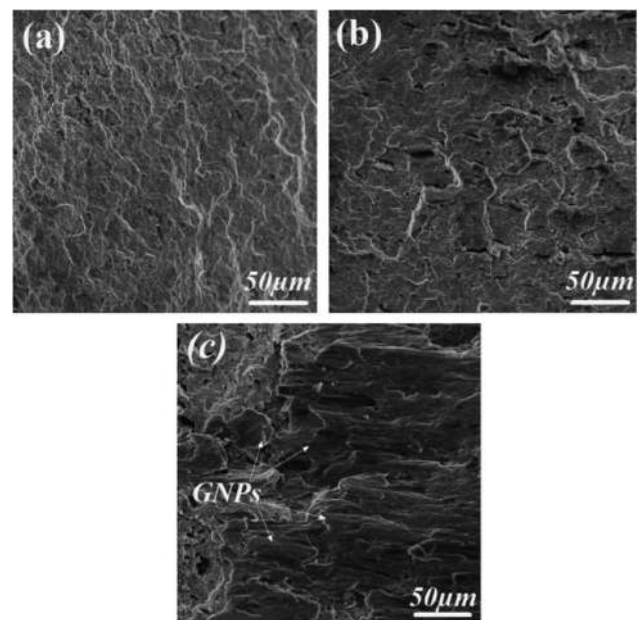


Figure 7: SEM images of fracture surface: (a) pure Al, (b) 0.3 wt% of GNP/Al, and (c) 0.3 wt% of GNP/Al which is perpendicular to the extrusion direction. Reproduced from ref. [30].

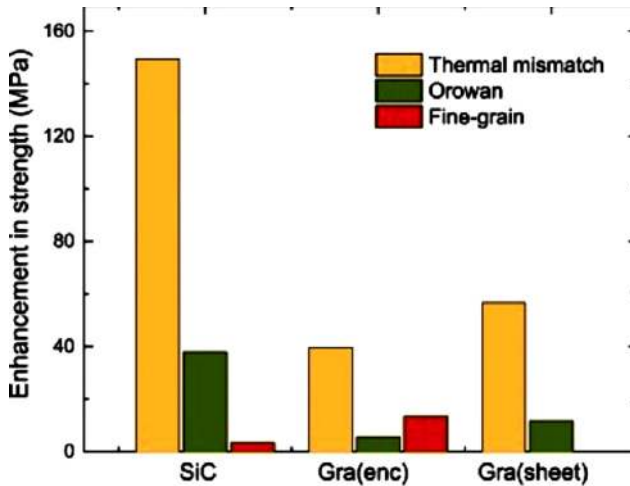


Figure 8: Strengthening mechanism in enhancing the tensile strength with the addition of SiC, encapsulating graphene, and sheet graphene. Reproduced from ref. [66].

The thermal mismatch mechanism may not be ideal for studying the strengthening effects of graphene due to its two-dimensional flexible structure. Recently, Li et al. [66] reported that the thermal mismatch mechanism is the most dominant strengthening mechanism in fabricating GNP/Al composite via PM, which is further heat-treated at various temperatures (570–650°C). Meanwhile, the load transfer has a moderate effect and grain refinement did not have an effect as illustrated in Figure 9(a). However, the ultimate tensile values from the shear lag (load transfer) show consistency in theoretical value as compared to the experimental one observed in Figure 9(b). Tensile strength increased by 23% because they varied the annealed temperature from 570 to 650°C. The consistency occurs

because they consider the strengthening effect of graphene itself from the shear lag mechanism, while the grain size and dislocation density as a result of the addition of graphene were not taken into consideration. They neglected the Orowan looping mechanism because of the low contribution strengthening effect on metal composites reinforced with carbonaceous nano-materials of large aspect ratios. The improvement in strength is well corroborated with the values predicted by the load transfer mechanism.

3.4 Grain refinement

The grain refinement mechanism is commonly represented by Hall–Petch mechanism. This mechanism is also one of the major factor in strength enhancement in AMC [68]. The grain size of Al is refined in graphene-Al composite because the GNP particles act as an obstacle to the growth of Al grain during fabrication processes [63,69]. Therefore, a high density of grain boundary can affect dislocation movement and propagation to adjacent grain, which results in strength enhancement. The grain refinement $\Delta\sigma_{GR}$ was famously represented by Hall–Petch relationship that can be described as equation (6).

$$\Delta\sigma_{GR} = \frac{k_y}{D}, \quad (6)$$

where k_y represents the material constant for Al ($k = 0.068 \text{ MPa}\cdot\text{m}^{-0.5}$) and D is the average grain size of the matrix.

Another model reported by Tang et al. [68] shows that the contribution of grain refinement can be further

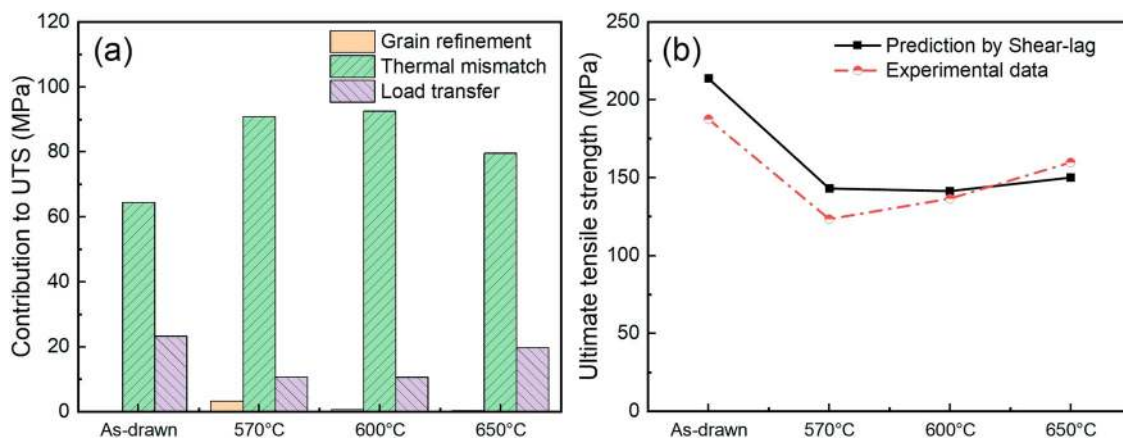


Figure 9: Evaluation of calculated strengthening mechanism of UTS in fabricating 2 wt% of GNP/Al composite. (a) Comparison of each strengthening mechanism and (b) calculated UTS predicted by shear lag mechanism (load transfer) against experimental data. Reproduced from ref. [66].

evaluated by the following relationship referred as equation (7).

$$\Delta\sigma_{GR} = k_y \left(d_c^{-\frac{1}{2}} - d_m^{-\frac{1}{2}} \right), \quad (7)$$

where k_y is a material constant and d_c and d_m are the average grain size in composites and matrix, respectively. Based on result finding from published literatures [20,70–72], it clearly shows that the Al grain was refined by the GNP particles. Xiong *et al.* [54] reported that the grain size of Al matrix is refined in the composite due to the presence of GNP particles that hinders the Al grain growth. Table 2 shows the comparison of volume fraction of GNP and the average grain size of Al matrix in pure Al and GNP/Al composite. The results show that the grain size of composite with the addition of 1.2 vol% of graphene has reduced by 49% as compared to the grain size of pure Al.

Some studies found a great match with the increment in strength in terms of theoretical and experimental data which indicates a good mechanism, while others found a large difference. As reported by Leng *et al.* [55], GNP presence caused limitations to the growth of Al grain during the hot extrusion process, which reduced the average grain size. Thus, the grain refinement mechanism contributed to enhancing the strength when the grain size is micron. The grain size was reduced from 11.2 to 5.3 μm because of the addition of GNP-reinforced Al7075 alloy. The results matched with the increment in yield strength and tensile strength of GNP/Al7075 composite by 15 and 10% (about 578 and 673 MPa) by 0.2 wt% of GNP addition [55]. Thus, it can be summarised that the finer the grain, the more the grain boundaries, and the stronger the resistance to dislocation motion, which therefore increased the yield strength and tensile strength.

Meanwhile, Zheng *et al.* [73] produced GNP/Al5083 composite by ball milling and hot extrusion process and found a decrease in grain size from 500 nm (Al5083) to 100 nm (1.0 wt% of GNP/Al5083 composite). Unlike other studies, their result showed a large difference in value between the experimental and calculated strengthening effect that is, 113 and 224 MPa, respectively, using grain refinement Hall–Petch formula. They explained the reason for this situation as follows: first, the input values for the

calculation were measured using TEM observations. Because of the site of the TEM observation, the results may not be correct, especially when it comes to grain sizes. Second, the modified shear lag model and the Orowan-Ashby model may not be ideal for GNP and Al_4C_3 phase. In terms of the modified shear lag model, the grain sizes fabricated by these methods were not efficiently aligned to the GNP in the same direction as per assumption according to the shear lag model. Meanwhile, for the Orowan mechanism, the predicted rod diameter ratio was barely adequate to match the assumption for the Orowan mechanism with the rod-shaped second phase.

4 Factors limiting the mechanical properties of graphene–Al composite

Several difficulties in the manufacture of graphene-reinforced Al composite matrix have limited their development in their production area. Three main factors contributing to the challenges and limiting the performance of graphene-reinforced Al composites are the fabrication processes, the variations in graphene structure, and the amount of graphene content.

The enhancement of mechanical properties is characterised by tensile strength, hardness, and fracture strength. They exhibit good mechanical properties because of the good dispersion of graphene within the Al matrix, high interfacial bonding, and their strengthening mechanism. However, the issue of cluster, poor wettability, porosity, and structural damage lead to degradation of the mechanical properties [40,47,73,74]. Therefore, it has been suggested that the fabrication processes, the difference in graphene structure, and the amount of graphene reinforcement play a significant role in determining the mechanical properties.

4.1 Fabrication processes

In previous studies, non-homogenous distribution of graphene was observed in many cases, particularly with high graphene content (>2 wt%) [2,35]. Several methods have been developed to fabricate the graphene-Al composite to achieve uniform distribution and avoid agglomeration. PM [25,60,75–77], casting technique [61,75,77], ultrasonication [37,78–80], and spark plasma sintering

Table 2: Grain size of Al matrix in pure Al and GNP/Al composite. Reproduced from ref. [54]

Volume fraction of GNP (vol%)	0	0.3	0.6	0.9	1.2
Average grain size (μm)	2.26	1.87	1.56	1.36	1.14

[32,49,54,73,81] have been commonly used as their production path. Most of these methods combine with post-processing technique to remove the void from the interface and aim to break the Van der Waals interaction between graphene sheets.

The most common fabrication method, PM, has emerged as the most successful technique in having a uniform dispersion and ease of fabrication. However, it has caused structural damage to the composite, which negatively affected the mechanical properties. Of three basic steps in PM, such as mixing, compacting, and sintering, the mixing part causes direct damage to the graphene structure due to the ball milling time or high rotation speed.

Another alternative method has been investigated by Rashad et al. [30,67] who used the semi-powder method in which they replaced ball milling with ultrasonication. The 0.3 wt% of GNP was ultrasonicated in acetone for 1 h before being mixed with aluminium powder slurry in acetone. Then, the mixtures were mechanically agitated, filtered, and vacuum dried for 12 h at 70°C to form a composite powder. Finally, to obtain the composite billet, the powder was sintered in a furnace at 600°C followed by hot extrusion. The experiment results show that the composite exhibited higher hardness and tensile strength by 14 and 11% as compared to pure Al.

Meanwhile, for the casting technique, several advantages and disadvantages can be observed in attempting this route as compared to the PM technique. The advantages are low cost, high rate of production, and simple instrumentation used which makes this technique an

ideal route. However, the drawbacks are difficulty in controlling the components, poor distribution of nanoparticles, and weak interfacial bonding between the phases [30]. These disadvantages arise because the melt temperature, pouring rate, shape, and speed of the agitator affect the particle distribution. Wettability and porosity are also the main challenges considered in affecting the mechanical properties. A study conducted by Li et al. [48] used stir casting and rolling technique as their fabrication route. First, they ball-milled 0.2 wt% of GNP/Al at a speed of 300 rpm before being cold-pressed. Then, with a ratio of 1:9, the powder was added to Al melt, and finally the composite underwent continuous casting and subsequent rolling. Their results show the homogenous distribution of GNP with a lamellar structure of GNP fibres. However, some microcracks were observed between the interfaces. This lamellar structure contributes to the increment in UTS by 38% as compared to pure Al, while the microcracks cause a decrease in ductility [64].

4.2 Graphene content

The amount of graphene will determine the optimum mechanical properties of the Al-graphene composite rather than the type of graphene structure. The incorporation of graphene in a small amount will strengthen the composite, whereas the addition of more graphene weakens them. The maximum strengthening efficiency is determined by graphene dispersion. When graphene is ineffectively dispersed,

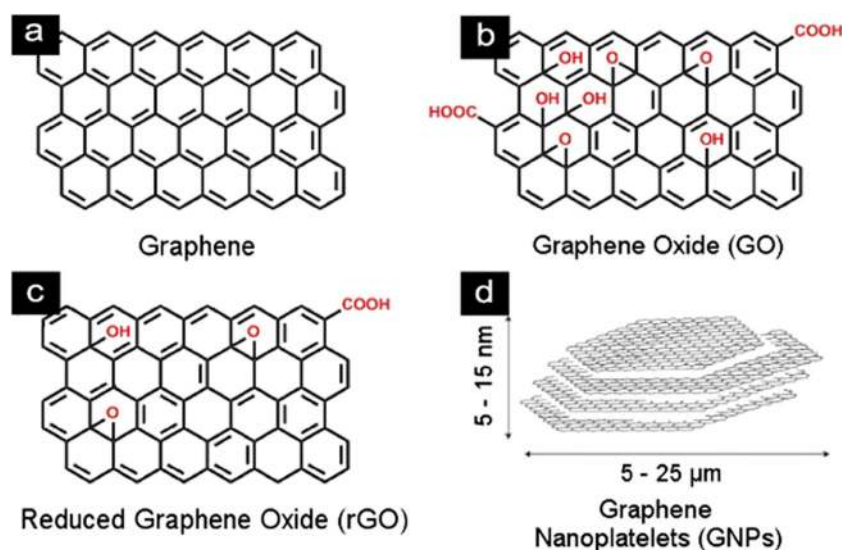


Figure 10: Schematic diagram of (a) graphene, (b) graphene oxide, (c) reduced graphene oxide, and (d) graphene nanoplatelets. Reproduced from ref. [88].

it agglomerates. The agglomeration of graphene not only reduces its strength, but also creates defects in Al/graphene composite, causing the composites to fail prematurely. Rashad *et al.* [30] and Wang *et al.* [61] mentioned that the optimised amount of graphene is 0.3 wt%, while other studies [61,66,81] reported that the optimum values of graphene are between 0.7–1.0 wt%.

Most of the published literature also mentioned that only a small percentage of graphene as low as 0.3 wt% was required in the matrix to improve the tensile strength and the hardness. Venkatesan and Xavier [78] evaluated three-graphene percentage content between 0.33, 0.55, and 0.77 wt% via liquid processing route and found that tensile and hardness properties increased relative to the amount of graphene. However, the properties started to decrease beyond 0.33 wt% due to agglomeration [30,83]. Meanwhile, Wang *et al.* [61] compared 0.3 wt% of graphene-Al composite and pure Al and observed that the tensile strength increased by 62% as compared to pure Al. Hu *et al.* [84] compared 0.5, 1.0, and 2.5 wt% of graphene content via ball milling combined with selective laser melting fabrication method and found that when graphene reached 2.5 wt%, the agglomeration of graphene was observed and the mechanical properties were reduced.

Hence, initially, the strength of Al increased with an increase in graphene content. However, the material properties started to deteriorate once they reached a critical level of graphene content because of the agglomeration issue [85,86].

4.3 Variation in graphene structure

The mechanical properties enhancement is also determined by the graphene structure. Various structures of graphene, such as rGO, GNP, and GO increase the mechanical properties up to a certain percentage, as shown in Figure 10 [82,87]. Other studies found their tensile strength to be in-between 240 and 280 MPa, while others found their tensile strength to be up to 110 MPa. Thus, changes in mechanical properties can be deduced from a variety of graphene structures.

A single layer of graphene is called graphene, while a multi-layer of graphene is called GNPs. Another structure used as much as graphene is called GO. The similarity between GO and graphene is the presence of a hexagonal C structure, while their difference is the presence of hydroxyl (OH), alkoxy (COC), carbonyl (CO), carboxylic acid (COOH), and other oxygen-based functional groups. In addition to the unique properties of graphene, GO is

Table 3: Mechanical properties change with respective type of graphene structure and fabrication processes

Matrix/Reinforcement	Optimum value	Production method	Maximum strength/hardness (MPa)	Changes in Mechanical properties	Reference
Al/GO (0.1, 0.3, and 0.5) wt%	0.3 wt% of GO	PM	110	30% increase in UTS	[89]
Al/GO (0.15, 0.3, and 0.6) wt%	0.3 wt% of GO	Electrostatic interaction & PM	167	73.9% increase in UTS	[90]
Al/BNP (0.3) wt%	0.3 wt% of GNP	Vacuum hot pressing	249	62% increase in UTS	[61]
Al2O3/BNP (0.25, 0.5, 1, and 2) wt%	2 wt% of GNP	PM	325	50% increase in UTS	[91]
Al/BNP (0.5, 1.0, 1.5, and 2.0) wt%	0.5 wt % of GNP	Cryomilling & Hot extrusion	170	18% increase in UTS	[59]
Al5083/BNP (0.5 and 1.0) wt%	1.0 wt% of GNP	PM & Hot extrusion	470	50% increase in UTS	[92]
Al/rGO (0.3, 0.5, 0.7, and 1.0) wt%	1.0 wt% of rGO	PM & SPS	320	49% increase in UTS	[93]

PM = powder metallurgy; SPS = spark plasma sintering.

easier to synthesize [82]. Nevertheless, there are only a few studies related to GO-reinforced AMC due to alumina formation during fabrication process. Meanwhile, rGO is prepared by the chemical reduction of GO.

Recently, rGO and GO have been studied in various fields due to their ability to achieve balance in strength and ductility of the composite. Zhao et al. [16] evaluated three kinds of lateral size effects (0.23, 1.1, and 15.4 μm) reinforced in the form of rGO to Al matrix on the composite. They assumed that the increment in lateral size of rGO caused a significant increase in the strength. However, their results show that larger lateral size caused a slight decrease in the strength (from 314 to 299 MPa) and a significant increase in the ductility of the composite. They explained that while the small lateral size of rGO is uniformly distributed in the Al matrix, large lateral sizes of rGO concentrate on Al flakes, which is more effective in strengthening the Al. Thus, the large lateral size of reinforcement achieved an excellent balance between strength and ductility. Overall, several authors have investigated the changes in mechanical properties of graphene nanosheet, GO, and rGO reinforced to Al matrix as summarised in Table 3.

5 Conclusion and future scopes

The insights gained from this review highlight the significant role of understanding the strengthening mechanism in graphene AMC and the contributing factors to maximise its enhanced mechanical properties. It is revealed that in the fabrication process, the addition of graphene content to the Al matrix enhances the properties up to a certain limit, otherwise, their properties are affected negatively.

The enhancement of mechanical properties is contributed mainly by the effective load transfer expressed in strengthening mechanism attributed as the Orowan looping of nanoparticles, load transfer from Al matrix to graphene, dislocation strengthening by the CTE mismatch, and grain refinement.

A better final property will depend on the homogeneous distribution and strong interfacial bonding between graphene and Al matrix. Various fabrication methods have been discussed, and PM has been selected as the main technology in producing graphene-reinforced Al composite because of its higher capability of good dispersion of graphene in the matrix. The amount of graphene will also determine the final composite performance and the variation in graphene structure.

The chemical bonding between the graphene and Al need to be explored further along with the optimum amount of Al_4C_3 formation, which are critical elements in attaining effective strengthening of the reinforced composite. Although positive results have been achieved from the Al_4C_3 , some ambiguities in influencing factors still need to be addressed in graphene-reinforced MMC.

Another point to note is the existence of a certain threshold in which the amount of graphene content determines the optimum mechanical properties. Above the threshold, the properties will degrade rapidly because of the agglomeration formation of graphene in the Al matrix. Therefore, a potential point of interest for future research is in determining the threshold and seeking the perfect balance between strength gain and ductility of the composite before the manufacturing process.

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References

- [1] Srivivas, P. D. and M. S. Charoo. Role of reinforcements on the mechanical and tribological behavior of aluminum metal matrix composites – a review. *Materials Today: Proceedings*, Vol. 5, 2018, pp. 20041–20053.
- [2] Zhao, Z., P. Bai, W. Du, B. Liu, D. Pan, R. Das, et al. An overview of graphene and its derivatives reinforced metal matrix

- composites: preparation, properties and applications. *Carbon*, Vol. 15, 2020, pp. 969–979.
- [3] Hashim, H., M. S. Salleh, M. Z. Omar, A. B. Sulong, and A. A. Rahman. Influence of short heat treatment on the microstructures and mechanical properties of Thixoformed aluminum alloy composite. *Jurnal Tribologi*, Vol. 28, 2021, pp. 96–104.
- [4] Alhawari, K. S., M. Z. Omar, M. J. Ghazali, M. S. Salleh, and M. N. Abdulrazaq. Effect of thixoforming on the wear properties of Al-Si-Cu aluminum alloy. *Jurnal Teknologi*, Vol. 79, 2017, pp. 83–87.
- [5] Arif, M. A., M. Z. Omar, and Z. Sajuri. Formation of spheroidal microstructure in semi-solid state of Al-4.8Si-2.8Cu-0.5Mg aluminium alloy. *Jurnal Kejuruteraan*, Vol. 30, 2018, pp. 275–280.
- [6] Anis, A. L., M. K. Talari, N. Kishore Babu, M. H. Ismail, G. D. Janaki Ram, and I. A. Mohd Arif. Grain refinement of Ti-15V-3Cr-3Sn-3Al metastable β titanium alloy welds using boron-modified fillers. *Journal of Alloys and Compounds*, Vol. 749, 2018, pp. 320–328.
- [7] Vijaya Ramnath, B., C. Elanchezian, M. Jaivignesh, S. Rajesh, C. Parswajinan, and A. Siddique Ahmed Ghias. Evaluation of mechanical properties of aluminium alloy-alumina-boron carbide metal matrix composites. *Materials and Design*, Vol. 58, 2014, pp. 332–338.
- [8] Hanizam, H., M. S. Salleh, M. Z. Omar, A. B. Sulong, and M. A. M. Arif. Effects of hybrid processing on microstructural and mechanical properties of thixoformed aluminum matrix composite. *Journal of Alloys and Compounds*, Vol. 836, 2020, id. 155378.
- [9] Novoselov, K. S., A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, et al. Discover of graphene: electric field effect in atomically thin carbon films. *Science (New York, N.Y.)*, Vol. 306, 2004, pp. 666–669.
- [10] Talangkun, S., C. Potisawang, and P. Saenpong. Thixoforming and rheo-die-casting of A356/SiC composite. *Metals*, Vol. 10, 2020, id. 251.
- [11] Krishna, S., and M. Karthik. Evaluation of hardness strength of aluminium alloy (AA6061) reinforced with silicon carbide. *International Journal on Recent Technologies in Mechanical and Electrical Engineering (IJRMEE)*, Vol. 1, No. 4, 2014, pp. 14–18.
- [12] Naik, L., Nagaraj, K., and Chikmeti, R. A review and analysis of mechanical properties of aluminium silicon carbide – metal matrix composite. *International Journal of Advances in Scientific Research and Engineering*, Vol. 2, 2016, pp. 24–30.
- [13] Mehta, V. R. and M. P. Sutaria. Investigation on the effect of stirring process parameters on the dispersion of SiC particles inside melting crucible. *Metals and Materials International*, Vol. 27, 2020, pp. 2989–3002.
- [14] Arifin, A., G. Gunawan, I. Yani, M. Yanis, and R. Pradifta. Optimization of stir casting method of aluminum matrix composite (AMC) for the hardness properties by using Taguchi method. *Jurnal Kejuruteraan*, Vol. 29, 2017, pp. 35–39.
- [15] Ju, B., W. Yang, P. Shao, M. Hussain, Q. Zhang, Z. Xiu, et al. Effect of interfacial microstructure on the mechanical properties of GNPs/Al composites. *Carbon*, Vol. 162, 2020, pp. 346–355.
- [16] Zhao, M., D. B. Xiong, Z. Tan, G. Fan, Q. Guo, C. Guo, et al. Lateral size effect of graphene on mechanical properties of aluminum matrix nanolaminated composites. *Scripta Materialia*, Vol. 139, 2017, pp. 44–48.
- [17] Yoganand, C. R. and P. Ramaswamy. Dispersion of graphene in aluminum powder to aid synthesis of homogenous nanocomposites. *Materials Today: Proceedings*, Vol. 5, 2018, pp. 27928–27935.
- [18] Cataldi, P., A. Athanassiou, and I. S. Bayer. Graphene nanoplatelets-based advanced materials and recent progress in sustainable applications. *Applied Sciences (Switzerland)*, Vol. 8, 2018, id. 1438.
- [19] Omar, M., A. Rashid, N. Muhamad, A. B. Sulong, and N. N. Kadiman. Effect of mechanical properties with addition of graphene nano-platelets in Cu/GNPs composite-green part. *Jurnal Kejuruteraan*, Vol. 32, 2020, pp. 729–734.
- [20] Ezatpour, H. R., M. T. Parizi, and G. R. Ebrahimi. The effects and improvements of GNPs + CNTs on the mechanical properties and microstructure of AZ80 matrix composite. *International Journal of Lightweight Materials and Manufacture*, Vol. 4, 2021, pp. 323–338.
- [21] Xu, Z. Y., C. J. Li, Z. Wang, D. Fang, P. Gao, J. M. Tao, et al. Balancing the strength and ductility of graphene oxide-carbon nanotube hybrid reinforced aluminum matrix composites with bimodal grain distribution. *Materials Science and Engineering A*, Vol. 796, 2020, id. 140067.
- [22] Hashim, H., M. S. Salleh, and M. Z. Omar. Homogenous dispersion and interfacial bonding of carbon nanotube reinforced with aluminum matrix composite: a review. *Reviews on Advanced Materials Science*, Vol. 58, 2019, pp. 295–303.
- [23] Hanizam, H., M. S. Salleh, M. Z. Omar, A. B. Sulong, S. H. Yahaya, and N. Siswanto. Effect of magnesium surfactant on wettability of carbon nanotube in A356 alloy composite. *Journal of Advanced Manufacturing Technology*, Vol. 13, 2019, pp. 33–44.
- [24] Iijima, S. Helical microtubules of graphitic carbon. *Nature*, Vol. 354, 1991, pp. 56–58.
- [25] Kumar, D. and S. Abhishek. Effect of graphene reinforcement on the mechanical properties of Aluminium 6061 nanocomposite prepared by stir casting method, *International Journal for Scientific Research & Development*, Vol. 6, 2018, pp. 45–48.
- [26] Su, J. and J. Teng. Recent progress in graphene-reinforced aluminum matrix composites. *Frontiers of Materials Science*, Vol. 15, 2021, pp. 79–97.
- [27] Hidalgo-Manrique, P., S. Yan, F. Lin, Q. Hong, I. A. Kinloch, X. Chen, et al. Microstructure and mechanical behaviour of aluminium matrix composites reinforced with graphene oxide and carbon nanotubes. *Journal of Materials Science*, Vol. 52, 2017, pp. 13466–13477.
- [28] Peigney, A., C. Laurent, E. Flahaut, R. R. Bacsá, and A. Rousset. Specific surface area of carbon nanotubes and bundles of carbon nanotubes. *Carbon*, Vol. 39, 2001, pp. 507–514.
- [29] Hu, Z., G. Tong, D. Lin, C. Chen, H. Guo, J. Xu, et al. Graphene-reinforced metal matrix nanocomposites – A review. *Materials Science and Technology (United Kingdom)*, Vol. 32, 2016, pp. 930–953.
- [30] Rashad, M., F. Pan, A. Tang, and M. Asif. Effect of Graphene Nanoplatelets addition on mechanical properties of pure aluminum using a semi-powder method. *Progress in Natural Science: Materials International*, Vol. 24, 2014, pp. 101–108.

- [31] Nieto, A., A. Bisht, D. Lahiri, C. Zhang, and A. Agarwal. Graphene reinforced metal and ceramic matrix composites: a review. *International Materials Reviews*, Vol. 62, 2017, pp. 241–302.
- [32] Bisht, A., M. Srivastava, R. M. Kumar, I. Lahiri, and D. Lahiri. Strengthening mechanism in graphene nanoplatelets reinforced aluminum composite fabricated through spark plasma sintering. *Materials Science and Engineering A*, Vol. 695, 2017, pp. 20–28.
- [33] Prashantha Kumar, H. G. and M. Anthony Xavier. Graphene reinforced metal matrix composite (GRMMC): a review. *Procedia Engineering*, Vol. 97, 2014, pp. 1033–1040.
- [34] Li, Z., G. Fan, Q. Guo, Z. Li, Y. Su, and D. Zhang. Synergistic strengthening effect of graphene-carbon nanotube hybrid structure in aluminum matrix composites. *Carbon*, Vol. 95, 2015, pp. 419–427.
- [35] Liang, A., X. Jiang, X. Hong, Y. Jiang, Z. Shao, and D. Zhu. Recent developments concerning the dispersion methods and mechanisms of graphene. *Coatings*, Vol. 8, 2018, id. 33.
- [36] Baig, Z., O. Mamat, and M. Mustapha. Recent progress on the dispersion and the strengthening effect of carbon nanotubes and graphene-reinforced metal nanocomposites: a review. *Critical Reviews in Solid State and Materials Sciences*, Vol. 43, 2018, pp. 1–46.
- [37] Chandrashekar, A. Mechanical and tribological properties of AA-7075 and graphene reinforced metal, Vol. 3, 2018, pp. 230–235.
- [38] Naseer, A., F. Ahmad, M. Aslam, B. H. Guan, W. S. W. Harun, N. Muhamad, et al. A review of processing techniques for graphene-reinforced metal matrix composites. *Materials and Manufacturing Processes*, Vol. 34, 2019, pp. 957–985.
- [39] Jiang, Y., Z. Tan, G. Fan, Z. Zhang, D. B. Xiong, Q. Guo, et al. Nucleation and growth mechanisms of interfacial carbide in graphene nanosheet/Al composites. *Carbon*, Vol. 161, 2020, pp. 17–24.
- [40] Saboori, A., M. Pavese, C. Badini, and P. Fino. Microstructure and thermal conductivity of Al-graphene composites fabricated by powder metallurgy and hot rolling techniques. *Acta Metallurgica Sinica (English Letters)*, Vol. 30, 2017, pp. 675–687.
- [41] Luis Vanderlei, T., T. Luis Fernando, and E. J. Zoqui. Microstructural characterization of the A356 alloy in different processing routes used in the thixoforming process. *Revista Materia*, Vol. 22, 2017.
- [42] Arif, M. A. M., M. Z. Omar, Z. Sajuri, and M. S. Salleh. Effects of Cu and Mg on thixoformability and mechanical properties of aluminium alloy 2014. *Transactions of Nonferrous Metals Society of China*, Vol. 30, 2020, pp. 275–287.
- [43] Bastwros, M., G. Y. Kim, K. Zhang, and S. Wang. Fabrication of graphene reinforced aluminum composite by semisolid processing. *ASME International Mechanical Engineering Congress and Exposition, Proceedings (IMECE)*, 2013.
- [44] Niteesh Kumar, S. J., R. Keshavamurthy, M. R. Haseebuddin, and P. G. Koppad. Mechanical properties of aluminium-graphene composite synthesized by powder metallurgy and hot extrusion. *Transactions of the Indian Institute of Metals*, Vol. 70, 2017, pp. 605–613.
- [45] Fadavi Boostani, A., S. Tahamtan, Z. Y. Jiang, D. Wei, S. Yazdani, R. Azari Khosroshahi, et al. Enhanced tensile properties of aluminium matrix composites reinforced with graphene encapsulated SiC nanoparticles. *Composites Part A: Applied Science and Manufacturing*, Vol. 68, 2015, pp. 155–163.
- [46] Baig, Z., O. Mamat, M. Mustapha, S. Ali, and M. Yasir. Low energy solution ball milling of graphene nanoplatelets (GNPs) reinforced aluminium nanocomposites and its mechanical properties. *IOP Conference Series: Materials Science and Engineering*, Vol. 380, 2018, id. 12009.
- [47] Baig, Z., O. Mamat, M. Mustapha, A. Mumtaz, M. Sarfraz, and S. Haider. An efficient approach to address issues of graphene nanoplatelets (GNPs) incorporation in aluminium powders and their compaction behaviour. *Metals*, Vol. 8, 2018, id. 90.
- [48] Li, G. and B. Xiong. Effects of graphene content on microstructures and tensile property of graphene-nanosheets/aluminum composites. *Journal of Alloys and Compounds*, Vol. 697, 2017, pp. 31–36.
- [49] Khoshghadam-Pireyousefan, M., R. Rahmanifard, L. Orovciik, P. Švec, and V. Klemm. Application of a novel method for fabrication of graphene reinforced aluminum matrix nanocomposites: synthesis, microstructure, and mechanical properties. *Materials Science and Engineering A*, Vol. 772, 2020, id. 138820.
- [50] Bastwros, M., G. Y. Kim, C. Zhu, K. Zhang, and S. Wang. Effect of mechanical alloying on AL6061-graphene composite fabricated by SEMI-solid powder processing. *ASME 2013 International Manufacturing Science and Engineering Conference Collocated with the 41st North American Manufacturing Research Conference, MSEC 2013*, 2013.
- [51] Pérez-Bustamante, R., D. Bolaños-Morales, J. Bonilla-Martínez, I. Estrada-Guel, and R. Martínez-Sánchez. Microstructural and hardness behavior of graphene-nanoplatelets/aluminum composites synthesized by mechanical alloying. *Journal of Alloys and Compounds*, Vol. 615, 2014, pp. S578–S582.
- [52] Bastwros, M., G. Y. Kim, C. Zhu, K. Zhang, S. Wang, X. Tang, et al. Effect of ball milling on graphene reinforced Al6061 composite fabricated by semi-solid sintering. *Composites Part B: Engineering*, Vol. 60, 2014, pp. 111–118.
- [53] Shao, P., W. Yang, Q. Zhang, Q. Meng, X. Tan, Z. Xiu, et al., “Microstructure and tensile properties of 5083 Al matrix composites reinforced with graphene oxide and graphene nanoplates prepared by pressure infiltration method,” *Composites Part A: Applied Science and Manufacturing*, Vol. 109, No. 2, 2018, pp. 151–162.
- [54] Xiong, B., K. Liu, W. Xiong, X. Wu, and J. Sun. Strengthening effect induced by interfacial reaction in graphene nanoplatelets reinforced aluminum matrix composites. *Journal of Alloys and Compounds*, Vol. 845, 2020, id. 156282.
- [55] Leng, J., Y. Dong, B. Ren, R. Wang, and X. Teng. Effects of graphene nanoplates on the mechanical behavior and strengthening mechanism of 7075Al alloy. *Materials*, Vol. 13, 2020, pp. 1–13.
- [56] Mandal, A., J. K. Tiwari, N. Sathish, C. P. Paul, S. K. Mishra, A. N. V. Ch, et al. Microstructure and microhardness study of aluminium graphene composite made by laser additive manufacturing. *Applied Innovative Research (AIR)*, Vol. 1, No. 1, 2019, pp. 66–74.
- [57] Bartolucci, S. F., J. Paras, M. A. Rafiee, J. Rafiee, S. Lee, D. Kapoor, et al. Graphene-aluminum nanocomposites. *Materials Science and Engineering A*, Vol. 528, 2011, pp. 7933–7937.
- [58] Etter, T., P. Schulz, M. Weber, J. Metz, M. Wimpler, J. F. Löffler, et al. Aluminium carbide formation in interpenetrating

- graphite/aluminium composites. *Materials Science and Engineering A*, Vol. 448, 2007, pp. 1–6.
- [59] Li, J. L., Y. C. Xiong, X. D. Wang, S. J. Yan, C. Yang, W. W. He, et al. Microstructure and tensile properties of bulk nanostructured aluminum/graphene composites prepared via cryomilling. *Materials Science and Engineering A*, Vol. 626, 2015, pp. 400–405.
- [60] Shin, S. E., H. J. Choi, J. H. Shin, and D. H. Bae. Strengthening behavior of few-layered graphene/aluminum composites. *Carbon*, Vol. 82, 2015, pp. 143–151.
- [61] Wang, J., Z. Li, G. Fan, H. Pan, Z. Chen, and D. Zhang. Reinforcement with graphene nanosheets in aluminum matrix composites. *Scripta Materialia*, Vol. 66, 2012, pp. 594–597.
- [62] Sharma, A., V. M. Sharma, A. Gugaliya, P. Rai, S. K. Pal, and J. Paul. Friction stir lap welding of AA6061 aluminium alloy with a graphene interlayer. *Materials and Manufacturing Processes*, Vol. 35, 2020, pp. 258–269.
- [63] Fadavi Boostani, A., S. Yazdani, R. Taherzadeh Mousavian, S. Tahamtan, R. Azari Khosroshahi, D. Wei, et al. Strengthening mechanisms of graphene sheets in aluminium matrix nanocomposites. *Materials and Design*, Vol. 88, 2015, pp. 983–989.
- [64] Seyed Pourmand, N. and H. Asgharzadeh. Aluminum matrix composites reinforced with graphene: a review on production, microstructure, and properties. *Critical Reviews in Solid State and Materials Sciences*, Vol. 45, 2020, pp. 289–337.
- [65] Kelly, A. and W. R. Tyson. “Tensile properties of fibre-reinforced metals: Copper/tungsten and copper/molybdenum,” *Journal of the Mechanics and Physics of Solids*, Vol. 13, No. 6, 1965, pp. 329–338.
- [66] Li, J., X. Zhang, and L. Geng. Effect of heat treatment on interfacial bonding and strengthening efficiency of graphene in GNP/Al composites. *Composites Part A: Applied Science and Manufacturing*, Vol. 121, 2019, pp. 487–498.
- [67] Rashad, M., F. Pan, A. Tang, M. Asif, J. She, J. Gou, et al. Development of magnesium-graphene nanoplatelets composite. *Journal of Composite Materials*, Vol. 49, 2015, pp. 285–293.
- [68] Tang, X. C., L. Y. Meng, J. M. Zhan, W. R. Jian, W. H. Li, X. H. Yao, et al. Strengthening effects of encapsulating graphene in SiC particle-reinforced Al-matrix composites. *Computational Materials Science*, Vol. 153, 2018, pp. 275–281.
- [69] Gupta, R., G. P. Chaudhari, and B. S. S. Daniel. Strengthening mechanisms in ultrasonically processed aluminium matrix composite with in situ Al₃Ti by salt addition. *Composites Part B: Engineering*, Vol. 140, 2018, pp. 27–34.
- [70] Ahmad, A. H., S. Naher, S. N. Aqida, and D. Brabazon. Routes to spheroidal starting material for semisolid metal processing. *Comprehensive Materials Processing*, Vol. 5, 2014, pp. 135–148.
- [71] Chak, V. and H. Chattopadhyay. Fabrication and heat treatment of graphene nanoplatelets reinforced aluminium nanocomposites. *Materials Science and Engineering A*, Vol. 791, 2020, id. 139657.
- [72] Arif, I. A. M., M. K. Talari, A. L. Anis, M. H. Ismail, and N. Kishore Babu. Grain refinement, microstructural and hardness investigation of C added Ti-15-3 alloys prepared by Argon Arc melting. *Transactions of the Indian Institute of Metals*, Vol. 70, 2017, pp. 861–865.
- [73] Zheng, Z., X. X. Zhang, J. C. Li, and L. Geng. High-content graphene nanoplatelet reinforced aluminum composites produced by ball milling and hot extrusion. *Science China Technological Sciences*, Vol. 63, 2020, pp. 1426–1435.
- [74] Boostani, A. F., S. Yazdani, R. A. Khosroshahi, Z. Y. Jiang, and D. Wei. A novel graphene-stimulated semi-solid processing to fabricate advanced aluminium matrix nanocomposites. *Materials Science and Engineering A*, Vol. 736, 2018, pp. 316–322.
- [75] Şenel, M. C., M. Gürbüz, and E. Koç. Mechanical and tribological behaviours of aluminium matrix composites reinforced by graphene nanoplatelets. *Materials Science and Technology (United Kingdom)*, Vol. 34, 2018, pp. 1980–1989.
- [76] Boostani, A. F., R. T. Mousavian, S. Tahamtan, S. Yazdani, R. A. Khosroshahi, D. Wei, et al. Graphene sheets encapsulating SiC nanoparticles: a roadmap towards enhancing tensile ductility of metal matrix composites. *Materials Science and Engineering A*, Vol. 648, 2015, pp. 92–103.
- [77] Yan, S. J., S. L. Dai, X. Y. Zhang, C. Yang, Q. H. Hong, J. Z. Chen, et al. Investigating aluminum alloy reinforced by graphene nanoflakes. *Materials Science and Engineering A*, Vol. 612, 2014, pp. 440–444.
- [78] Venkatesan, S. and M. A. Xavior. Characterization on aluminum alloy 7050 metal matrix composite reinforced with graphene nanoparticles. *Procedia Manufacturing*, Vol. 30, 2019, pp. 120–127.
- [79] Lokesh, K. S., I. C. Chethan, K. R. Naveen Kumar, V. Kannan, and J. R. Naveen Kumar. Determination of compressive strength of graphene reinforced with aluminium-7075 metal matrix composites. *International Journal of Mechanical Engineering and Technology*, Vol. 9, 2018, pp. 327–335.
- [80] Shil, A., S. Roy, P. S. Balaji, J. Kumar Katiyar, S. Pramanik, and A. Kumar Sharma. Experimental analysis of mechanical properties of stir casted aluminium-graphene nanocomposites. *IOP Conference Series: Materials Science and Engineering*, Vol. 653, 2019, id. 12021.
- [81] Tian, W., S. Ming, B. Mei Li, X. Wang, J. Chen, M. Liu, et al. Graphene-reinforced aluminum matrix composites prepared by spark plasma sintering. *International Journal of Minerals Metallurgy and Materials*, Vol. 23, 2016, pp. 723–729.
- [82] Güler, Ö. and N. Bağcı. A short review on mechanical properties of graphene reinforced metal matrix composites. *Journal of Materials Research and Technology*, Vol. 9, 2020, pp. 6808–6833.
- [83] Venkatesan, S. and M. A. Xavior. Mechanical behaviour of Aluminium metal matrix composite reinforced with graphene particulate by stir casting method. *Journal of Chemical and Pharmaceutical Sciences*, Vol. 10, 2017, id. 55.
- [84] Hu, Z., F. Chen, J. Xu, Q. Nian, D. Lin, C. Chen, et al. 3D printing graphene-aluminum nanocomposites. *Journal of Alloys and Compounds*, Vol. 746, 2018, pp. 269–276.
- [85] Asgharzadeh, H. and M. Sedigh. Synthesis and mechanical properties of Al matrix composites reinforced with few-layer graphene and graphene oxide. *Journal of Alloys and Compounds*, Vol. 728, 2017, pp. 47–62.
- [86] Zhang, J., Z. Chen, J. Zhao, and Z. Jiang. Microstructure and mechanical properties of aluminium-graphene composite powders produced by mechanical milling. *Mechanics of Advanced Materials and Modern Processes*, Vol. 4, 2018, id. 4.

- [87] Li, M., H. Gao, J. Liang, S. Gu, W. You, D. Shu, et al. Microstructure evolution and properties of graphene nanoplatelets reinforced aluminum matrix composites. *Materials Characterization*, Vol. 140, 2018, pp. 172–178.
- [88] Chintalapudi, K. and R. M. R. Pannem. An intense review on the performance of graphene oxide and reduced graphene oxide in an admixed cement system. *Construction and Building Materials*, Vol. 259, 2020, id. 120598.
- [89] Gao, X., H. Yue, E. Guo, H. Zhang, X. Lin, L. Yao, et al. Preparation and tensile properties of homogeneously dispersed graphene reinforced aluminum matrix composites. *Materials and Design*, Vol. 94, 2016, pp. 54–60.
- [90] Wu, Y., K. Zhan, Z. Yang, W. Sun, B. Zhao, Y. Yan, et al. Graphene oxide/Al composites with enhanced mechanical properties fabricated by simple electrostatic interaction and powder metallurgy. *Journal of Alloys and Compounds*, Vol. 775, 2019, pp. 233–240.
- [91] AbuShanab, W. S., E. B. Moustafa, E. Ghandourah, and M. A. Taha. Effect of graphene nanoparticles on the physical and mechanical properties of the Al2024-graphene nanocomposites fabricated by powder metallurgy. *Results in Physics*, Vol. 19, 2020, id. 103343.
- [92] Zhang, H., C. Xu, W. Xiao, K. Ameyama, and C. Ma. Enhanced mechanical properties of Al5083 alloy with graphene nanoplates prepared by ball milling and hot extrusion. *Materials Science and Engineering A*, Vol. 658, 2016, pp. 8–15.
- [93] Hirt, G. and R. Kopp. *Thixoforming: semi-solid metal processing*, John Wiley & Sons, Chicester, 2009.