Aluminum Matrix Composites Reinforced with Graphene: A Review on Production, Microstructure, and Properties

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Aluminum Matrix Composites Reinforced with Graphene: A Review on Production, Microstructure, and Properties

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ABSTRACT
Graphene, a single-atom-thick sheet of sp² hybridized carbon atoms densely packed within a hexagonal crystal lattice, owes a significant portion of its rapidly expanding usage in various fields of industry and science to its large surface area, lightweight, unique electronic and thermal properties, and extraordinary mechanical properties that it possesses. Using lightweight and high-strength materials leads to a substantial decrease in fuel consumption as well as an increase in payload. Among the numerous candidates to fulfill the aforementioned requirements, Al alloys and specifically, Al matrix composites (AMCs) reinforced with various graphene particles (nano-sheets, nano-platelets, etc.) stand out owing to their inherent lightweight, high specific strength and modulus, superior ductility and excellent thermal conductivity. In this paper, it has been endeavored to provide a comprehensive overview of the various methods of the synthesis and fabrication of graphene-reinforced AMCs with an overall intention of achieving a homogeneous distribution of graphene within the Al matrix. The emphasis of this review has been largely placed upon the detailed examination of the mechanical properties of these composites described in the recent literature published in this field. The strengthening mechanisms of Al/graphene composites, as well as the parameters affecting the strength and ductility achieved by graphene, such as the agglomeration of graphene within the Al matrix and Al/graphene interfacial reactions, have been elucidated. The role of graphene on the electrical and thermal properties of graphene-reinforced AMCs and the directions for future research are addressed. It should be noted, however, that while a diligent analysis of various works published in the field of Al/graphene composites has been carried out in this review, an exact comparison between the varying stages of each work, or their final properties is impractical, mainly due to the differences in the used initial substances, processing, and analyses.

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1. Introduction

Aluminum (Al), with a face-centered cubic (fcc) structure and an atomic radius of 0.143 nm (Figure 1a), is a light metal that has garnered a great deal of interest due to its physical and mechanical properties such as low density, high corrosion resistance, and specific strength. These properties have allowed Al to find widespread applications in aerospace, construction, automobile, marine, and architectural industries. However, due to its rather poor tribological properties, as well as low overall strength, Al is rarely used alone in its pure form, and it is preferred to utilize various Al alloys and composites instead. Common additives to enhance mechanical properties of Al include ceramic reinforcements such as Al₂O₃, SiC, BN, and B₄C. In recent years, a new class of strengthening additives has emerged in the form of novel carbonaceous nanomaterials, namely carbon nanotubes (CNTs), fullerene, and graphene. These reinforcements, when incorporated in a matrix of pure or alloyed Al, not only enhance the physical and mechanical properties of the base metal, they also add a plethora of functional properties such as self-lubricating surfaces.

Graphene, a flat, monolayer of sp² hybridized carbon atoms which are densely packed within a two-dimensional (2D) “beehive” structure, owes a significant portion of its rapidly expanding usage to its high surface area, lightweight, transparency, unique electronic and thermal properties, and extraordinary mechanical properties (Table 1). Even since its discovery, graphene has received widespread attention due to its potential applications in various fields of industry and science including, but not limited to, medicine, electronics, energy generation, and environmental preservation. With a fracture strength and Young’s modulus of 130 GPa and 1 TPa, respectively, graphene is widely considered to be the strongest reinforcement in metal, ceramic, and polymer matrix composites. A particular branch of industry which requires the utilization of lightweight and high-strength materials to be effective is aerospace and automotive industries. Using such materials leads to a substantial decrease in fuel consumption and an increase in payload. Among the numerous candidates to fulfill the aforementioned requirements, AMCs reinforced with graphene in various types, e.g. nanosheets (GNSs), nano-platelets (GNPs), nanoflakes (GNFs), etc. stand out due to their inherent lightweight, high strength, superior ductility, and good electrical and thermal conductivities. Consequently, a great deal of research has been focused on discovering, and thereby improving, the unique properties of Al/graphene composites.

However, prior to the manufacturing of the aforementioned composites and evaluation of their various characteristics and properties, a reliable method of synthesizing graphene of sufficient quality must be considered to meet the current demand. While reviewing different methods of the synthesis of graphene is outside the scope of this review, they have proven to have a significant impact on the final...
properties of the composites regardless. Among the most common methods of graphene production, mechanical exfoliation was possibly the first technique to be developed, which involved peeling layers off of graphite sheets.9 In this technique, also known as “scotch tape” method, the graphite sheets are placed on a substrate and then peeled-off using adhesive tape, until a graphene structure with the desired number of layers is achieved. While this process has proven to be a fast and easy way of graphene production, structural defects tend to emerge in the final product.10 These defects and disorders have a detrimental impact on the properties of graphene, especially on its electrical conductivity. Unfortunately, the defects imposed upon the structure have been found to be irreparable,11 making it the largest drawback for this method of synthesis. Chemical vapor deposition (CVD) method is also a prominent route for the large-scale production of single-layer graphene. A typical CVD growth of graphene involves the deposition of gaseous hydrocarbons as the carbon source at elevated temperatures onto a substrate, and is fully capable of synthesizing high-quality graphene.12 The ability to control the number and the thickness of achieved graphene layers is among the advantages of this technique.

Due to the high cost and difficulty of direct synthesis of graphene, conversion of graphite into graphite oxide via chemical oxidation methods, subsequent exfoliation of graphite oxide to graphene oxide (GO), and further reduction of GO has proven to be a feasible alternative (Figure 1b).13-15 The most popular method of synthesizing graphite oxide is the Hummers method, which involves oxidizing graphite using various oxidants such as H2SO4, HNO3, and KMnO4.16 Similar to Hummers method, Brodie17 and Staudenmaier18 have also successfully synthesized graphite oxide through oxidation of graphite, albeit with different reactants. Due to the presence of various functional groups such as hydroxyl and epoxide on graphite oxide, it possesses a hydrophilic nature and can be easily exfoliated in aqueous solutions to form GO sheets. The resulting GO can later be chemically reduced in presence of multiple reducing agents such as ascorbic acid,19,20 sodium borohydride,21,22 and hydrazine.23,24 The resulting product is usually named as reduced graphene oxide (rGO) or chemically derived graphene, which resembles graphene but contains residual functional groups and structural defects.

Following the successful synthesis of graphene with desired properties, the Al matrix and graphene reinforcement can be hybridized through one of the
numerous techniques discussed further in the following, to fabricate Al/graphene composites with desirable properties to meet the demand of various applications. This review endeavors to investigate the production methods as well as the physical and mechanical properties of the various graphene-reinforced AMCs while elucidating the different mechanisms and phenomena responsible for the changes.

2. Production of Al/graphene composites

2.1. Powder processing
The majority of Al/graphene composites are produced through powder metallurgy (PM) technique. PM is a near-net-shape process that offers an inexpensive route for producing various metallic, nonmetallic, and composite components. PM processing for the production of an Al/graphene composite consists of two main stages: (a) mixing of Al powder particles with graphene particles, and (b) consolidation of the powder mixture into the desired shape or form.

2.1.1. Mixing methods. The first stage in PM processing is the mixing of pure or prealloyed Al powder particles with graphene or GO particles. When GO particles are employed for the composite synthesis, GO is reduced during or after mixing with Al to form rGO. The critical factor in the fabrication of an Al/graphene composite is attaining a uniform distribution of graphene within the Al matrix without forming agglomerates. Since the final properties of the resulting composite will be dependent mainly on this key factor, the initial mixing stage is paramount to ensure homogenous dispersion, and consequently, desired mechanical properties. Ball milling and dispersion in a liquid media via stirring or sonication are common and feasible techniques of mixing the Al and graphene powder particles. Although no individual method has proven to be better than the other, homogeneous dispersion and enhanced properties have since been reported in composites synthesized by one, or both of the aforementioned techniques.

Ball milling. Ball milling, also commonly referred to as “mechanical milling” or “mechanical alloying” when used for metals, is one of the traditional and popular methods of powder mixing. Ball milling can be carried out in different milling devices, ranging from shaker mills to planetary and attritor mills for larger amounts of powder. Various compositions of milling balls and jars can also be used, such as stainless steel, tungsten carbide, and zirconia. The ball milling process is often carried out in the presence of an inert gas such as Ar to prevent oxidation of metallic powders, and various process control agents (PCAs) like stearic acid, methanol, and ethanol are utilized to reduce cold welding between metal powder particles, and thereby inhibits agglomeration.

Despite the fact that the initial energy provided by this method often proves sufficient for mixing purposes, high-energy ball milling process is also utilized for elimination of agglomerates and bonding between graphene and the metal matrix. The energy output of this process can be predominantly changed by adjusting the milling time, rotation speed, and ball-to-powder weight ratio (BPR), where higher energy outputs result in a greater size reduction of the initial powders, at the cost of potential damage to their structure. The forces induced by the impact of the milling balls are applied to the van der Waals interlayer bonds of graphene to relieve agglomerates, while the high-speed rotation of the milling jar promotes uniform and homogeneous dispersion of graphene within the metal matrix.

Ball milling is usually carried out in dry media, but it can be performed in a liquid media as well. Liquid media grants a greater degree of control over some process parameters, with the superior ability to remove graphene agglomerates and create cryogenic temperatures to inhibit grain growth of metal powders with nanocrystalline structure. It has been reported that distribution of graphene within the matrix is more uniform in a liquid media compared to dry milling.

Mechanical exfoliation of bulk graphite into GNP s can also be attained through ball milling. To achieve this purpose, a significant amount of energy is required to fully break down the interlayer van der Waals bonds. Mechanical exfoliation of graphite powders is among the viable methods of synthesizing graphene and considerable efforts have been invested into combining the synthesis and composite mixing stages using high-energy ball milling process. Utilizing ball milling process for exfoliation of graphite requires a liquid media, and various media such as dimethylformamide (DMF), isopropanol, acetone, and deionized water have been utilized.

Another aspect of the ball milling process to be taken into account is the change of metal powder morphology from spherical particles to flake, as a result of repeated impacts by the milling balls. Recently, a novel PM approach termed “flake PM”, which uses flake metal powder, has been employed to produce uniform
dispersion of reinforcements in the metal matrices.\textsuperscript{54} Flaky particles promote a better interfacial bonding with graphene, leading to an improved transfer of stress from the matrix to the reinforcement, therefore enhancing the overall mechanical properties of the MMC.\textsuperscript{26–29,55}

Ball milling technique can also be used for mechanical milling of a metal powder, or mechanical alloying of a mixture of metal powders prior to mixing with graphene. Mechanical milling/alloying process which involves alternating stages of cold welding, fracture and re-welding of powders, often results in an increment of strength through work hardening and grain refinement of the metal powder. In contrast, material transfer involved in the mechanical alloying process results in obtaining a homogeneous alloy with superior strength. This phenomenon has been taken advantage of in synthesis of various MMCs reinforced with graphene.\textsuperscript{26,43,44,56,57}

Table 2 presents an overview of the recent research for hybridizing of Al and graphene utilizing ball milling process as a means of mixing or alloying, and their relevant details, in chronological order. In a study conducted by Pérez-Bustamante et al.,\textsuperscript{28} Al powder was hybridized with different amounts (0.25, 0.5, and 1 wt\%) of GNP by means of a SPEX ball mill for 1, 3, and 5 h. Figure 2a–2d indicate the SEM images for Al and Al-1 wt\% GNP powders after ball milling for 1 and 5 h. Clusters of GNP are not visible even in short milling durations, indicating a proper dispersion of the GNP in the Al matrix by using high-energy ball milling. Moreover, the powder is increased in size and becomes equiaxed with increasing the milling time due to the predominance of cold welding over fracture. Nevertheless, the GNP do not have a significant effect on the morphology of the Al powder particles. Figure 2e represents the Raman spectra of the Al-1 wt\% GNP powders milled for various times. As can be seen, longer milling time leads to the emergence of defects (indicated by the reduced intensity of peaks), consequently increasing the amorphous fraction of GNP within the composite. On the other hand, increasing the milling durations results in better and more uniform dispersion of graphene within the Al matrix. Thus, an optimal balance of dispersion, cold welding and fracture has to be taken into consideration for milling time.

The effect of milling time on the mechanical properties and structural integrity of graphene-reinforced Al6061 composite was investigated in research by Bastwros et al.\textsuperscript{26} Figure 3a–3c shows the morphology of Al6061-1 wt\% graphene powders after ball milling for 0.5, 1, and 1.5 h. It is evident that the size of Al alloy particles is increased while the size of graphene flakes is decreased with increasing the milling time. The fractured graphene flakes are repeatedly embedded into the cold-welded Al particles during ball milling, hence it is very difficult to locate the

<table>
<thead>
<tr>
<th>Year</th>
<th>Composite material</th>
<th>Milling type</th>
<th>BPR</th>
<th>Rotational speed (rpm)</th>
<th>Milling atmosphere</th>
<th>PCA</th>
<th>Milling time (h)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>2011</td>
<td>Al-0.1 wt% graphene</td>
<td>High-energy attritor</td>
<td>–</td>
<td>–</td>
<td>Ar</td>
<td>–</td>
<td>1</td>
<td>31</td>
</tr>
<tr>
<td>2012</td>
<td>Al (surface modified)-0.3 wt% GNS</td>
<td>Attritor</td>
<td>–</td>
<td>325</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>58</td>
</tr>
<tr>
<td>2012</td>
<td>Al2124-3 wt% graphene</td>
<td>Turbula mixer</td>
<td>15:1</td>
<td>400</td>
<td>Ar</td>
<td>–</td>
<td>2</td>
<td>41</td>
</tr>
<tr>
<td>2014</td>
<td>Al6061-1 wt% graphene</td>
<td>SPEX</td>
<td>2.6:1</td>
<td>–</td>
<td>Ambient</td>
<td>–</td>
<td>0.17, 0.5, 1, 1.5</td>
<td>26</td>
</tr>
<tr>
<td>2014</td>
<td>Al-0.25, 0.50, 1.0 wt% GNP</td>
<td>SPEX</td>
<td>5:1</td>
<td>–</td>
<td>Ar</td>
<td>–</td>
<td>1, 3, 5</td>
<td>28</td>
</tr>
<tr>
<td>2014</td>
<td>(Al-3.9 Cu-1.5 Mg)-0.5 wt% GNP</td>
<td>Roller milling tank</td>
<td>10:1</td>
<td>75</td>
<td>–</td>
<td>Ambient</td>
<td>12</td>
<td>59</td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.5, 1, 1.5, 2 wt% GNF</td>
<td>Attritor</td>
<td>40:1</td>
<td>180</td>
<td>–</td>
<td>–</td>
<td>2</td>
<td>33</td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.7 wt% FLG</td>
<td>Planetary ball mill</td>
<td>15:1</td>
<td>200</td>
<td>–</td>
<td>–</td>
<td>1</td>
<td>30</td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.05 wt% graphene</td>
<td>Planetary ball mill</td>
<td>20:1</td>
<td>150</td>
<td>Ambient</td>
<td>–</td>
<td>7.5</td>
<td>60</td>
</tr>
<tr>
<td>2016</td>
<td>Al2024-0.03, 0.05 vol% FLG</td>
<td>High-energy ball mill</td>
<td>15:1</td>
<td>600</td>
<td>Ar</td>
<td>–</td>
<td>6</td>
<td>61</td>
</tr>
<tr>
<td>2016</td>
<td>Al-1 wt% GNP</td>
<td>Planetary ball mill</td>
<td>15:1</td>
<td>500</td>
<td>–</td>
<td>–</td>
<td>6</td>
<td>4</td>
</tr>
<tr>
<td>2017</td>
<td>Al6061-0.7 wt% Ni-decorated graphene</td>
<td>Planetary ball mill</td>
<td>10:1</td>
<td>360</td>
<td>Al</td>
<td>–</td>
<td>1.5</td>
<td>62</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5 wt% rGO</td>
<td>Planetary ball mill</td>
<td>4:1</td>
<td>200</td>
<td>–</td>
<td>Toluene</td>
<td>6</td>
<td>64</td>
</tr>
<tr>
<td>2017</td>
<td>Al2219-0.5, 1, 2 wt% graphene</td>
<td>Planetary ball mill</td>
<td>4:1</td>
<td>200</td>
<td>–</td>
<td>–</td>
<td>2</td>
<td>63</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.25, 0.5, 1 wt% GNS</td>
<td>Planetary ball mill</td>
<td>10:1</td>
<td>250</td>
<td>Ar</td>
<td>–</td>
<td>4</td>
<td>65</td>
</tr>
<tr>
<td>2017</td>
<td>AlMg5-1 vol% GO</td>
<td>Planetary ball mill</td>
<td>10:1</td>
<td>360</td>
<td>Ar</td>
<td>–</td>
<td>20</td>
<td>66</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.5, 0.1, 2.5 wt% graphene</td>
<td>Planetary ball mill</td>
<td>5:1</td>
<td>200</td>
<td>Ambient</td>
<td>–</td>
<td>2</td>
<td>67</td>
</tr>
<tr>
<td>2018</td>
<td>Al-1 wt% GNP</td>
<td>SPEX</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>Wet mixing in Ethanol</td>
<td>0.17</td>
<td>68</td>
</tr>
<tr>
<td>2018</td>
<td>Al-1 wt% CNT</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>2018</td>
<td>Al-(1 wt% CNT + 1 wt% GNP)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>
graphene flakes after 1.5 h milling. As can be seen in the Raman spectra of composite powders (Figure 3d), after 1.5 h of milling, the intensity ratio of D-band to G-band ($I_D/I_G$), representing defects and disorders in the graphene structure, is increased to 1.4 from 1.08. Therefore, it can be deduced that prolonged milling times lead to an increase in defects. However, increasing the milling time to 5 h does not have a significant impact on the aforementioned ratio. This is ascribed to the protection of graphene sheets from further damage through embedment within the Al6061 particles. On the other hand, the intensity ratio of G-band to 2D-band ($I_G/I_{2D}$), indicating the number of graphene layers, is progressively lowered as the milling time is increased. The continuous decrease in this ratio upon ball milling expresses the reduction of the number of graphene layers. This is due to the separation of graphene layers as a result of the physical force of the impacts between the powder and milling balls.

Figure 2. (a–d) Effect of ball milling on the morphology of Al and Al/graphene powders: (a) Al/1 h, (b) Al/5 h, (c) Al-1 wt% GNP/1 h, and (d) Al-1 wt% GNP/5 h (The insets display the optical micrographs from the cross section of the powders). (e) Raman spectra for Al-1 wt% GNPs after high-energy ball milling for 1, 3, and 5 h (Reprinted with permission).
The effect of milling time on the flexural stress-strain curves and fracture surfaces of Al6061-1 wt% graphene powders consolidated by semi-solid sintering is illustrated in Figure 4. Short milling times, such as 10 min, have proven to be ineffective in the full dispersion of graphene within the matrix, resulting in a degradation of mechanical properties, as shown in Figure 4a. By increasing the milling time to 1 and 1.5 h, the flexural strength is significantly improved by 47% and 34%, respectively. On the other hand, the size of ductile dimples of the fracture surface is considerably decreased at longer milling durations (Figure 4b and 4c), revealing the reduction in ductility of the composite.

**Solution-assisted mixing.** Solution-assisted mixing method involves stirring or sonication of a mixture of metal powder and graphene particles in a liquid media. Stirring is commonly accomplished in mechanical agitation systems while sonication is the act of applying sound energy to agitate the particles. The sound waves with frequencies more than 20 kHz resulting from ultrasonication device help mixing the metal/graphene powders within the media, and break the van der Waals interlayer forces present in graphene sheets to inhibit agglomeration.

When it comes to the selection of the mixing media, it is essential that the chosen solution does not readily react with the composite powders, especially graphene, throughout the process. The popular media of choice in this method include various polar and non-polar liquids such as deionized water, ethanol, acetone, and Dimethyformamide (DMF). In comparison to ball milling, solution-assisted mixing technique conveys lower quantities of energy to the powders, therefore posing a considerably lower risk of inducing defects and disorders within the graphene structure. However, solution-assisted mixing methods are less effective in changing the particle size or morphology of metal powders as a result of the absence of shearing forces induced by the impact of milling balls. One of the main advantages of solution-assisted mixing over ball milling is that the contamination issue is almost entirely eliminated in the former method due to the absence of milling balls and jars. Nevertheless, residual traces of the moist media may remain within the substances and potentially interfere with further consolidation procedures, thus, it is almost entirely removed during the subsequent drying process. It is thus inferred that ball milling and solution-assisted mixing methods each possess distinct advantages which complement that of the other. This has lead to a significant amount of research utilizing both techniques to synthesize metal/graphene composite powders with desirable characteristics.

**Figure 3.** SEM images of the milled Al6061-1 wt% graphene powder at different ball milling times: (a) 0.5 h, (b) 1 h, (c) 1.5 h. (d) Raman spectra of the as-received graphene and ball-milled Al6061-2 wt% graphene powders at different milling times (Reprinted with permission).
A chronological overview of the recent research using solution-assisted dispersion method for mixing graphene with Al powder particles is provided in Table 3. In a study conducted by Wang et al., graphite oxide was ultrasonicated in deionized water until exfoliation to GO. During ultrasonication, shear forces and cavitation act on the graphite oxide and induce exfoliation. Concurrently, Al powder particles were ball milled until their morphology was turned into flake shape, before being treated in PVA aqueous solution to introduce a hydrophilic PVA on the surface of the Al flakes. The GO aqueous solution was then added to the PVA-treated Al flakes and mechanically stirred in deionized water. The PVA membrane is able to improve the adsorption of GO nano-sheets on the Al flakes, since the existing hydroxyl groups in PVA had endowed Al with good water wettability, enabling a strong hydrogen-bonding interaction between Al and GO. Figure 5a illustrates SEM micrograph of GO sheets adsorbed on a flake Al particle. The presence of many fine wrinkles on the surface of Al/GO powder indicates that the GO nano-sheets are uniformly dispersed on the Al flakes. The EDS analysis shows the existence of a carbonaceous composition in the composite powder (Figure 5c), while the Raman spectrum of the Al/GO composite powder depicts D and G bands (Figure 5d), both of which are typical of pristine GO. Finally, to obtain Al/GNS composite powders, the Al/GO powder mixture was heated and kept in an Ar atmosphere to decompose the PVA and reduce the GO nano-sheets to GNSs.

**Figure 4.** (a) Flexural stress-strain curves of Al6061-1 wt% graphene composites prepared by ball milling for various durations followed semi-solid sintering (b, c) SEM images of fracture surfaces of the composites prepared at milling times of 10 min and 90 min (Reprinted with permission).26

**Table 3.** Reported Al/graphene powders hybridized using solution-assisted mixing and their processing parameters.

<table>
<thead>
<tr>
<th>Year</th>
<th>Composite material</th>
<th>Mixing method</th>
<th>Media</th>
<th>Mixing time (h)</th>
<th>Speed/Frequency</th>
<th>Temperature (˚C)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>2011</td>
<td>Al-1, 2, 3 wt% GNP</td>
<td>Ultrasonication</td>
<td>Acetone</td>
<td>5</td>
<td>50 kHz</td>
<td>RT</td>
<td>79</td>
</tr>
<tr>
<td>2012</td>
<td>Al-1, 3, 5 wt% GNP</td>
<td>Mechanical agitation</td>
<td>Acetone</td>
<td>1</td>
<td>2000 rpm</td>
<td>RT</td>
<td>80</td>
</tr>
<tr>
<td>2014</td>
<td>Al-0.3 wt% rGO</td>
<td>Mechanical agitation</td>
<td>Ethanol</td>
<td>—</td>
<td>400 rpm</td>
<td>RT</td>
<td>77</td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.1, 0.25, 0.5 wt% rGO</td>
<td>Ultrasonication</td>
<td>Ethanol</td>
<td>0.5</td>
<td>—</td>
<td>RT</td>
<td>82</td>
</tr>
<tr>
<td>2017</td>
<td>Al-1 wt% FLGO</td>
<td>Mechanical agitation</td>
<td>Ethanol/Water</td>
<td>1</td>
<td>—</td>
<td>75</td>
<td>39</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5, 1, 3, 5 wt% GNP</td>
<td>Ultrasonication</td>
<td>Acetone</td>
<td>0.17</td>
<td>—</td>
<td>RT</td>
<td>83</td>
</tr>
<tr>
<td>2017</td>
<td>Al6061-2, 5, 10, 15 vol% graphene</td>
<td>Dispersion in liquid</td>
<td>Isopropyl alcohol</td>
<td>24</td>
<td>—</td>
<td>RT</td>
<td>84</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.2 vol% rGO</td>
<td>Mechanical agitation</td>
<td>PVA</td>
<td>1</td>
<td>—</td>
<td>RT</td>
<td>85</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5, 1 wt% GNP</td>
<td>Ultrasonication</td>
<td>Ethanol</td>
<td>0.75</td>
<td>—</td>
<td>RT</td>
<td>86</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.08, 0.33, 0.66, 0.98, 1.62 wt% GO</td>
<td>Ultrasonication</td>
<td>Water</td>
<td>2</td>
<td>—</td>
<td>RT</td>
<td>87</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.05, 0.1, 0.2 wt% GO</td>
<td>Mechanical agitation</td>
<td>Isopropyl alcohol</td>
<td>1, 3, 6</td>
<td>—</td>
<td>RT</td>
<td>88</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.1, 0.2, 0.3 wt% graphene</td>
<td>Mechanical agitation</td>
<td>Ethanol</td>
<td>0.05</td>
<td>—</td>
<td>RT</td>
<td>89</td>
</tr>
</tbody>
</table>
One of the unique advantages of solution-assisted mixing technique is its ability to form strong interfacial bonds between matrix and reinforcement through adsorption and reduction of GO sheets on the surface of Al particles, which has been elucidated in a number of reports.\textsuperscript{39,77} It is stated that the negatively charged GO sheets within the aqueous solution, due to the presence of functional groups on their surface,\textsuperscript{91} are attracted to the ionized, positively charged $\text{Al}^{3+}$ particles and get attached to their surface.\textsuperscript{77} The electrostatic attraction force between the phases with opposite charge promotes the interaction between Al particles and GO sheets while preventing them from restacking and forming agglomerates. Furthermore, as a result of electron exchange between matrix and reinforcement, GO is partially reduced on the surfaces of Al particles. This phenomenon is known as “electrostatic adsorption”. Li et al.\textsuperscript{77} utilized this method to achieve uniform dispersion of graphene in an Al matrix without the assistance of any chemical agent. An aqueous solution of GO prepared through sonication of graphite oxide was mixed with flaky Al powders dispersed in ethanol by means of mechanical stirring. After filtering, rinsing, and vacuum drying, the obtained Al/GO composite powder was heated under an Ar-H\textsubscript{2} atmosphere to reduce the GO sheets. Figure 6a–6d shows SEM images of the surface of an Al flake after mixing with GO for various durations. As can be observed, GO sheets are adsorbed on the Al surface as the mixing continued. The adsorption process is facilitated by increasing the amount of water in the water/ethanol solution so that by an increase in the volume fraction of water in the adsorption medium from 20% to 80%, the adsorption rate is increased over 10 orders of magnitude (Figure 6e). This was attributed to the positive role of water in accelerating the ionization of Al flakes, as evidenced by the zeta potential measurements (the inset of Figure 6e). Nevertheless, immersing the Al/GO composite powder in bare water has a detrimental effect due to the excessive hydrolysis of Al. Similarly, Asgharzadeh and Sedigh\textsuperscript{39} utilized solution-assisted mixing for dispersing
of few-layer graphene (FLG) and graphene oxide (FLGO) within the Al matrix. First, FLG and FLGO sheets were dispersed in ethanol and water/ethanol (70/30), respectively. Concurrently, gas-atomized (GA) and mechanically milled (MM) Al powder particles were dispersed in ethanol. Afterwards, the FLG and FLGO suspensions were added to the Al suspension and wet mixing was performed at 75°C for 10 min to obtain composite powders. As observed in Figure 7a and 7b, the wrinkled and very thin layers of FLGO sheets are adsorbed and covered the surfaces of Al powder particles. The evaluation of Raman spectra of composite powders (Figure 7e) indicates that ID/IG ratio of FLGO-reinforced composite powders is much higher than that of FLGO sheets. Furthermore, characteristic peaks of FLG are detected in the FTIR spectra of FLGO-reinforced composite powders (Figure 7f). Both of these analyses confirm the partial reduction of FLGO during the wet mixing owing to the electrostatic adsorption mechanism. Nevertheless, FLG sheets are mostly placed beside Al powders (Figure 7c and 7d) and their structure is almost remained unchanged after solution-assisted mixing (Figure 7e and 7f).

2.1.2. Compaction and consolidation

Following the mixing stage, the resulting metal/graphene composite powders need to be compacted into a solid shape before undergoing any secondary or finishing steps or being utilized in the desired application. In general, the compaction procedure involves the application of pressure to the powders, either with or without external heating. However, the application of pressure without heat, also commonly known as cold pressing, leads to weakness and brittleness in the compact. Therefore, an additional sintering stage is required to activate the diffusion mechanisms, strengthen the existing bonds, and improve the strength and ductility. This procedure which is known as “press-and-sinter” method, is the conventional PM process for the production of various parts including MMCs. Since no additional pressure is applied during the sintering stage, this method of consolidation shall be referred to as “pressureless sintering”.

An issue that should be taken into consideration, is the potentially low density of the final product achieved through press-and-sinter route, which can lead to various problems such as degraded mechanical properties, or risk of oxidation of Al and burning of graphene content. To overcome this problem, pressing and sintering stages can be combined into a single operation in which pressure is applied in concurrence with heating to the powders. Indeed, sintering is accelerated by the application of external pressure. This route of consolidation shall be referred to as “sintering with pressure” or “pressure-assisted sintering”. Hot pressing, hot isostatic pressing (HIP), spark plasma sintering (SPS), hot extrusion, hot rolling, and sinter forging are examples of the sintering with pressure method. Regardless of the shaping and consolidation method, this step would determine the density and structure of the bulk material, and thus directly influence the mechanical properties of the composite material.

2.1.2.1. Press-and-sinter. Press-and-sinter route possesses many advantages over other consolidation methods, including low manufacturing cost, high production rates, reduction or otherwise elimination of
machining and capability of the production of complex shapes. In this process, mixed composite powders are fed into a rigid die, compacted into the desired shape at room temperature, ejected from the die, and then sintered at a temperature below the melting point of the metal matrix for an appropriate time in a controlled atmosphere furnace. The applied pressure upon compaction deforms the ductile metal powder.
Table 4. Reported bulk graphene-reinforced AMC parts produced using press-and-sinter and their processing conditions.

<table>
<thead>
<tr>
<th>Year</th>
<th>Composite material</th>
<th>Year</th>
<th>Composite material</th>
<th>Pressing</th>
<th>Sintering</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>2011</td>
<td>Al-3 wt% GNP</td>
<td>Pressure (MPa)</td>
<td>Time (min)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2012</td>
<td>Al-5 wt% GNP</td>
<td>520</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2014</td>
<td>Al-0.25, 0.5, 1 wt% GNP</td>
<td>950</td>
<td>–</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2015</td>
<td>Al-0.1, 0.25, 0.5 wt% rGO</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2017</td>
<td>Al-1 wt% FLGO Al-1 wt% FLG</td>
<td>500</td>
<td>–</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2017</td>
<td>Al-0.5, 1 wt% GNP</td>
<td>500</td>
<td>–</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2018</td>
<td>Al-0.05, 0.1, 0.2 wt% GO</td>
<td>595</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2018</td>
<td>Al-0.1, 0.2, 0.3 wt% graphene</td>
<td>200</td>
<td>–</td>
</tr>
</tbody>
</table>

Particles, resulting in the formation of bonds between them. The sintering step creates metallurgical bonds between the powder particles and forms a dense structure with low presence of voids and porosities, imparting the required properties to the compact. Thus, sintering temperature needs to be sufficiently high as to allow diffusion to occur at an acceptable rate, often at greater than one half of the melting point on Kelvin scale.92

Table 4 presents a number of recent studies using press-and-sinter route for the production of bulk Al/graphene compacts. The relevant details for each step such as applied pressure and pressing time for compaction as well as temperature, time, and atmosphere for sintering are included in the table. According to Table 4, graphene-reinforced AMC powders are commonly compacted in the pressure range of 500–950 MPa, which is higher than the conventional pressures used for Al and its alloys due to the lower compressibility of the composite powders. Furthermore, the sintering temperature depends on the composition of the Al matrix and varies between 400 and 600 ºC, similar to the temperature utilized for the sintering of conventional Al and its alloys. Latief and Sherif80 prepared AMCs reinforced with exfoliated graphene nano-platelets (GnPs) using press-and-sinter route and investigated the impacts of various sintering temperatures and GnP contents on the mechanical properties of the final composite samples. After mixing of Al with 1, 3, and 5 wt% GnP in acetone, the dried powder mixtures were compacted at the pressure of 500 MPa for 5 min and finally, the compacts were sintered at temperatures of 400, 500, and 600 ºC for 5 h. Figure 8a depicts density changes of Al/GnP composites sintered at different temperatures as a function of GnP content. As can be observed, higher sintering temperatures and GnP concentrations result in a lower density. This is attributed to the facilitation of atomic diffusion in higher temperatures, which affects the sinterability of Al as well as the sizeable difference in the melting point of graphene and Al, resulting in a low tendency of bond formation between them. Nevertheless, as shown in Figure 8b and 8c, the compressive strength and hardness of the Al/GnP composites are increased with increasing the sintering temperature and the amount of GnP particles. The improved mechanical properties at high sintering temperatures are related to the higher diffusion rate of atoms which instigates the formation of chemical bonding between the powder particles. On the other hand, the presence of more obstacles opposing the movement of dislocations, and thus the accumulation of dislocations at higher amounts of GnP, have resulted in the higher compressive strength and hardness of composites. The effect of sintering time on the mechanical properties of Al/GNP composites was elucidated by Pérez-Bustamante et al.28 Ball-milled Al/GNP powders were cold compacted under 950 MPa, then sintered at 500 ºC for 0.5, 1, 2, 3, 4, and 5 h under Ar atmosphere. Figure 9 presents the effect of different sintering times in tandem with various milling times and amount of graphene content on the microhardness of the composite samples. It can be deduced that the highest microhardness is achieved after 5 h of milling. A greater amount of graphene content is also observed to yield higher microhardness values. The optimal sintering time is also found to be 2 h, which leads to the highest amount of microhardness across all samples when used along with 5 h of milling. The composite sample produced through the aforementioned optimal conditions exhibits a 138% increase in microhardness over the pure Al sample. This is ascribed to the hardening of Al/GNP composite in prolonged milling durations, resulting in a higher rate of diffusion and shorter sintering time.93

Asgarzadeh and Sedigh39 prepared graphene-reinforced AMCs by press-and-sinter route and investigated the role of the type of reinforcement (FLG or FLGO) on the densification, microstructure, and mechanical properties of the composites. The wet-mixed composite powders were compacted under the pressure of 500 MPa and then were sintered at 600 ºC for 1 h under N2 atmosphere. The reduction of functional groups in graphene-reinforced AMCs, especially those containing FLGO was detected during sintering. The density, compressive strength, and hardness of composites reinforced with 1 wt% of FLG or FLGO are depicted in Figure 10.
addition of FLG or FLGO has a negative influence on the density so that both green and sintered densities of the composite compacts are slightly less than those of their unreinforced samples (Figure 10a). As observed in Figure 10b, mechanical properties of FLGO-reinforced composites are superior to those of the monolithic Al samples and the composites reinforced with FLG sheets. This was attributed to the uniform distribution of
graphene within the Al matrix and the formation of the appropriate interface between the matrix and reinforcement.

2.1.2.2. Pressure-assisted sintering. Application of pressure along with heating, or sintering with pressure, is a concurrent method of shaping and consolidation of powders which are normally resistant to sintering, such as composites. This method results in faster densification of the powders owing to the presence of external pressure. On the other hand, the temperature required for sintering is decreased as the available pressure is increased. With the application of pressure, the existing contact points between the particles amplify the incoming stress to exceed the yield stress of the material, leading to the significant densification of the powders through plastic flow. The initial densification continues until the local pressure falls below the yield stress at the processing temperature, and then the slower diffusion events take over. Therefore, the densification rate through pressure-assisted sintering declines over time. The initial densification is increased in proportion with applied pressure due to plastic flow, whereas the temperature plays an essential role during the later stages of densification owing to its considerable role in diffusion mechanisms.

Pressure-assisted sintering techniques employ combinations of temperature and pressure to eliminate pores. In general, the high-temperature processes rely on lower amounts of pressure, while the high-pressure processes depend upon lower temperatures. The state of stress depends on the means for the application of pressure, ranging from isostatic pressing with no shear to methods with significant amounts of induced shear. The hydrostatic pressure determines the shrinkage of pores and controls the rate of densification whereas shear leads to rearrangement of particles through sliding, collapsing of large pores, and disrupting particle surface films.

Table 5 represents the relevant information about recent literature employing pressure-assisted sintering for production of Al/graphene composites. Unfortunately, the effects of consolidation parameters on the densification, microstructure, and properties of graphene-reinforced AMCs have been scarcely reported. The most common methods of sintering with pressure are briefly described in the following.

Hot pressing. Hot pressing is the consolidation of a powder in a rigid die using simultaneous heating with loading from unidirectionally aligned punches. Although the pressure is applied along the vertical axis, there is a radial pressure against the die wall. The differential stress between the axial and radial directions generates shear which is effective in particle bonding. Initial densification includes particle rearrangement and plastic flow. As densification progresses, creep by grain boundary diffusion and volume diffusion become the dominant factors. According to Table 5, the typical operating conditions for graphene-reinforced AMCs depend on the composite material and component size, yet most likely range from 480 to 630 °C for temperature, and 30–600 MPa for pressure. The most common tooling is graphite, but other materials like hot work tool steels have also been used. To avoid the oxidation of Al powders and die damage, the die is enclosed within a protective atmosphere such as Ar gas or vacuum chamber. The die is usually lubricated by a thin layer of boron nitride to prevent a potential reaction between Al and the die at elevated temperatures.

Li et al. fabricated Al/GNS composite with a density of more than 99% of theoretical density by means of high-energy ball milling and vacuum hot
Table 5. Reported bulk Al/graphene parts produced using pressure-assisted sintering and their processing parameters.

<table>
<thead>
<tr>
<th>Year</th>
<th>Composite material</th>
<th>Stage 1</th>
<th>Stage 2</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>2011</td>
<td>Al-0.1 wt% graphene</td>
<td>HIP (375°C, 20 min)</td>
<td>Hot extrusion (65 ksi, 550°C, 12.5 mm s⁻¹, 4:1)</td>
<td>31</td>
</tr>
<tr>
<td>2012</td>
<td>Al-0.3 wt% GNS</td>
<td>Sintering (580°C, 2 h, Ambient)</td>
<td>Hot extrusion (440°C, 20:1)</td>
<td>58</td>
</tr>
<tr>
<td>2012</td>
<td>Al2124-3 wt% graphene</td>
<td>Cold pressing (525 MPa)</td>
<td>Hot extrusion (525 MPa, 305°C, 2:1)</td>
<td>41</td>
</tr>
<tr>
<td>2014</td>
<td>Al6061-1 wt% graphene</td>
<td>Cold pressing (50 MPa)</td>
<td>Hot Pressing (100 MPa, 630°C, 10 min)</td>
<td>26</td>
</tr>
<tr>
<td>2014</td>
<td>Al-0.3 wt% rGO</td>
<td>Cold pressing (500 MPa)</td>
<td>Hot pressing (600 MPa, 530°C, 1 h, Vacuum)</td>
<td>77</td>
</tr>
<tr>
<td>2014</td>
<td>(Al-3.9 Cu-1.5 Mg)-0.5 wt% GNF</td>
<td>HIP (110 MPa, 480°C, 2 h)</td>
<td>Hot extrusion (300 kN, 450°C, 2.2 mm s⁻¹, 11:1)</td>
<td>59</td>
</tr>
<tr>
<td>2014</td>
<td>Al-0.3 wt% GNP</td>
<td>Press-and-sinter (170 MPa, 600°C, 6 h)</td>
<td>Hot extrusion (470°C, 1 m min⁻¹)</td>
<td>81</td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.5, 1, 1.5, 2 wt% GNF</td>
<td>Hot extrusion (300°C, 17.6:1)</td>
<td>Annealing (300°C, 2 h)</td>
<td>33</td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.7 wt% FLG</td>
<td>Hot rolling (500°C, 12% reduction per pass)</td>
<td>Hot extrusion (440°C, 24 h, Vacuum)</td>
<td>61</td>
</tr>
<tr>
<td>2015</td>
<td>Al2024-0.5 vol% FLG</td>
<td>Hot pressing (140 MPa, 500°C, 1 h)</td>
<td>Hot pressing (500 MPa, 525°C)</td>
<td>4</td>
</tr>
<tr>
<td>2015</td>
<td>Al-1 wt% GNP</td>
<td>Cold pressing (200 MPa)</td>
<td>Sintering (400°C, 24 h, Vacuum)</td>
<td>–</td>
</tr>
<tr>
<td>2015</td>
<td>Al6061-0.7 wt% Ni-decorated graphene</td>
<td>Hot pressing (45 MPa, 580°C)</td>
<td>–</td>
<td>62</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5 wt% rGO</td>
<td>Hot pressing (240 MPa, 480°C)</td>
<td>Sintering (510°C, 30 min, N₂)</td>
<td>63</td>
</tr>
<tr>
<td>2017</td>
<td>Al2219-0.5, 1, 2 wt% graphene</td>
<td>SPS (40 or 30 MPa)</td>
<td>–</td>
<td>64</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5, 1, 3, 5 wt% GNP</td>
<td>SPS (50 MPa, 550°C, 40 min)</td>
<td>–</td>
<td>82</td>
</tr>
<tr>
<td>2017</td>
<td>Al6061-2, 5, 10, 15 vol% graphene</td>
<td>SPS (500°C, 2 min, Ar)</td>
<td>–</td>
<td>84</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.2 vol% rGO</td>
<td>Hot rolling (480°C, 12% reduction per pass)</td>
<td>Hot pressing (30 MPa, 610°C, 4 h, Vacuum)</td>
<td>85</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.25, 0.5, 1 wt% GNS</td>
<td>Hot pressing (30 MPa, 610°C, 4 h, Vacuum)</td>
<td>–</td>
<td>65</td>
</tr>
<tr>
<td>2017</td>
<td>AlMg5-1 vol% GO</td>
<td>Hot pressing (570 MPa, 550°C, 4 s)</td>
<td>–</td>
<td>66</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.05, 0.2, 0.4, 1 vol% FLG</td>
<td>SPS (50 MPa, 600°C, 20 min)</td>
<td>Hot extrusion (500 kN, 500°C, 20:1)</td>
<td>87</td>
</tr>
<tr>
<td>2018</td>
<td>Al-1 wt% GNP Al-1 wt% CNT + 1 wt% GNP</td>
<td>SPS (10–30 MPa, 420°C, vacuum)</td>
<td>–</td>
<td>68</td>
</tr>
</tbody>
</table>

Figure 11. TEM images of Al/GNS composites prepared by hot pressing: (a) Al-0.25 wt% GNS; (b) Al-0.5 wt% GNS; (c) Al-1 wt% GNS, and (d) SAD pattern (Reprinted with permission).65
pressing. The powder mixtures containing 0.25, 0.5, and 1 wt% GNP s were pressed into a graphite die and then sintered at 610 °C for 4 h under a pressure of 30 MPa. The relative density of Al/GNS composites is slightly lower than that of pure Al, which is attributed to the absorption of gas elements, such as O, N, and CO, resulting in an increase of air gap. TEM images of the Al/GNS composites (Figure 11) show a good interfacial bonding, but the Al carbide (Al4C3) phase with granular and short rod-like morphology is detected at the interface. With an increase of GNSs content in the AMC sample, the amount of Al4C3 is increased. The mechanism of Al4C3 formation and its effect on the mechanical properties of graphene-reinforced AMCs are discussed in Section 5.2.

In another study conducted by Kwon et al., fully densified Al-Mg alloy matrix composite reinforced with 1 vol% GO was prepared through hot pressing. The ball-milled composite powder mixture was preheated for 1.5 h at 550 °C in the air within a steel mold, then quickly transferred to a uniaxial press to apply the pressure of 570 MPa for 4 s. It has shown that the incorporation of only 1 vol% of GO results in a noteworthy increment of the mechanical behavior of the Al alloy matrix.

**Spark plasma sintering (SPS).** SPS is similar to hot pressing with the exception that heating occurs via the passage of current through the die and powder with rapid heating and uniaxial compression. Densification happens promptly once temperature and pressure are

![Figure 12. Optical micrographs of cross section of SPSed Al/GNP composites with reinforcement content of (wt%): (a) 0, (b) 0.5, (c) 1, (d) 3, and (e) 5. (Reprinted with permission)](Image)
attained, but temperature distribution in the powder compact is not uniform.\textsuperscript{95}

Bisht et al.\textsuperscript{83} produced Al/GNP composites via SPS subsequent to solution-assisted mixing. Various contents of GNPs (0.5, 1, 3, and 5 wt\%) and Al powder particles were dispersed in acetone using ultrasonication, and then the dried powder mixtures were sintered in SPS furnace under Ar atmosphere with maximum pressure of 50 MPa and temperature of 550°C, along with a holding time of 40 min. Although high relative densities are achieved for AMCs reinforced with 0.5 and 1 wt\% GNP (>99.8\%), the relative density is decreased at ≥1 wt\% GNP content due to the agglomeration of GNPs. The microstructure of the SPSed compacts (Figure 12) indicates that the incorporation of GNPs aids in uniform grain size distribution due to the pinning action of GNPs at the grain boundaries, restricting grain growth during sintering. However, at ≥3 wt\% GNP content, the agglomeration of GNPs can be clearly observed as black regions along the grain boundaries and thus the average grain size is increased. Moreover, XRD and TEM results do not show the formation of carbide layer at the interface of Al with GNPs in a noticeable amount.

Hot extrusion. Hot extrusion is the process of reducing the cross-section of a heated component by forcing it to flow through a die orifice using high pressure.\textsuperscript{92} Long shapes with constant cross-section are main products of extrusion. The material encounters compressive and shear stresses during hot extrusion. Due to the application of high compressive stress at elevated temperatures, almost all pores and cracks are eliminated during the hot extrusion process.

Hot extrusion is one of the main routes for the production of fully dense Al/graphene composites with superior properties. This method of consolidation is usually performed after pre-compaction of composite powders by various procedures, including cold pressing, press-and-sinter, HIP, SPS, etc. Hot extrusion is typically performed at temperatures above the recrystallization temperature, to facilitate the area reduction and enhance strength. According to Table 5, Al/graphene powder compacts are usually pre-heated in the temperature range of 300–550°C for various durations. The imposed strain during the hot extrusion is typically high, usually in the range of 2:1 to 20:1 in terms of the extrusion ratio (the starting cross-sectional area divided by the cross-sectional area of the final extrudate).

Ghazaly et al.\textsuperscript{41} employed hot extrusion technique for consolidation of Al2124 alloy reinforced with various amounts of graphene content (0.5, 3, and 5 wt\%). The ball-milled Al and graphene powder mixtures were cold compacted under 525 MPa, and then hot extruded at 305 °C with an applied pressure of 525 MPa and an extrusion ratio of 2:1. Figure 13 indicates the density changes as a function of the amount of graphene content. It is clear that the relative density of the extrudates is progressively lowered at higher graphene concentrations. This is ascribed to Al particles being coated with graphene layers during the milling process, which hinders the diffusion ability of Al particles, therefore resulting in deteriorated density.

In a study performed by Yan et al.,\textsuperscript{59} Al-Cu-Mg alloy matrix composites reinforced with 0.15 and 0.5 wt\% GNFs were produced through HIP followed by hot extrusion. First, GNFs were mixed with Al alloy powders using solution-assisted mixing in ethanol, and then the mixture solution was ball milled. After sealing and evacuating of the dried Al/GNF powder mixture in a cylinder-shaped Al package, it then underwent HIP treatment at 480°C for 2 h under the pressure of 110 MPa. Finally, the HIPed billet was pre-heated at 450°C for 1 h, and then hot extruded with an extrusion ratio of ~11:1 at an extrusion pressure force of ~300 kN. The microstructural analyses showed that GNFs were well dispersed within the matrix and no chemical reactions were observed at the interfaces between the GNFs and Al alloy matrix.

Zhou et al.\textsuperscript{87} produced fully densified Al/FLG composites via a combination of SPS and hot extrusion methods. First, GO colloid was added to the Al aqueous suspension and mixed. The Al/GO powder mixtures were sintered using SPS at 600°C for 20 min under a pressure of 50 MPa. The SPSed compacts were hot extruded at 500°C with an extrusion ratio of 20:1 and an applied force of 500 kN. Figure 14 shows the Raman spectra and microstructure of Al/FLG composite samples. According to the Raman spectroscopy results
Figure 14, (a) Raman spectra of GO and Al/FLG composite after SPS and SPS followed by hot extrusion; (b, c) FESEM and HRTEM images for the transverse cross-section of SPS-sintered Al-0.4 vol% FLG composite [insets in (c) show the SAD pattern of Al₂O₃ layer taken from yellow spot and the profile plot along red line]; (d, e) TEM and HRTEM images of the hot-extruded Al-0.4 vol% FLG composite [insets in (e) show the EDS analysis of Al₂O₃ layer taken from red spot, the SAD pattern of Al matrix taken from yellow spot and the profile plot along white line] (Reprinted with permission).87

(Figure 14a), the \( I_D/I_G \) ratio of GO is slightly increased from 0.97 to 1.09 after SPS, indicating the recovery of GO and thermal reduction to FLG during SPS densification. Furthermore, a 3D-network of FLG platelets, in the grain boundaries sandwiched between the thin amorphous Al₂O₃ layers, is created (Figure 14b and 14c). Subsequent plastic flow of the Al matrix by hot extrusion causes the destruction of this structure and the alignment of individual FLG platelets along the extrusion direction, as evident from Figure 14d. It is also deduced from the HRTEM image (Figure 14e) that a direct-contact interface between the FLG and the Al matrix without the formation of Al₄C₃ impurities is created.

2.2. Casting
Casting is another method of producing various types of MMCs including those reinforced with graphene. In general, the casting production of MMCs involves producing the matrix material in molten form, followed by the introduction of reinforcement particles into the melt and dispersing them within the molten metal. Afterwards, the melt containing reinforcement particles is solidified under controlled conditions to obtain the final composite component with desirable properties. Although various casting techniques are developed to produce MMCs, stir casting process is the most widely utilized among the others. In this method, the reinforcement particles are mixed with a molten metal matrix by means of mechanical stirring. During stir casting for the production of MMCs, stirring helps in two ways: (a) transferring particles into the melt, and (b) maintaining the particles in a state of suspension.96 The stirrer used for agitation of the melt is generally made of a material which can withstand the high temperature of the molten metal, like graphite.
In comparison with the powder processing method as the other prominent and widespread method of producing MMCs, stir casting has several advantages as well as drawbacks. These advantages are mainly consisting of the continuous matrix material, high rate of production, low cost, and simplicity of instrumentation which make stir casting an ideal route for economical production, capable of fabricating very large-sized components. On the other hand, stir casting process is considerably harder to control than powder metallurgy, and the poor distribution of nanoparticles within the matrix along with weak interfacial bonding between the phases can lead to undesirable properties of the final product.

Several factors need to be taken into consideration for the preparation of MMCs using stir casting method, such as difficult dispersion of reinforcement in the matrix melt leading to the inhomogeneous distribution of particles, presence of porosities, the possibility of chemical reactions occurring between the reinforcement and molten metal matrix, and wettability between these phases. Under ideal circumstances, the optimal composite product of stir casting would possess a uniform distribution of reinforcing particles in the matrix, minimum amount of porosities, good bonding or wettability between the substances while ascertaining that no chemical reactions occur between the substances.

As was mentioned above, the uneven distribution of particles in the matrix melt is one of the major problems of stir casting method, which is mainly related to the density mismatch between the two phases. The melt temperature, pouring rate, the shape and speed of the agitator, gating systems, method of the introduction of particles into the matrix melt, solidification rate and type, amount and nature of the particles have an impact on the distribution of particles in the matrix. For example, one of the best approaches used to create and maintain a good distribution of the reinforcement particles in the liquid metal is the vortex method, in which the melt is vigorously stirred to form a vortex at its surface, and the reinforcement particles are then introduced from the side of this vortex. The development of the vortex during stirring is helpful for transferring the particles into the matrix melt as the pressure difference between the inner and the outer surface of the melt sucks the particles into the liquid. However, air bubbles and all the other impurities on the surface of the melt are also sucked into the liquid, resulting in considerable number of porosity and inclusions in the final cast product. Secondary processing techniques, such as extrusion, can modify the distribution of the particles, but complete declustering cannot be achieved even at high levels of deformation. It should be noted that the uniform dispersion of smaller particles like nanoparticles within the metal matrix is more difficult due to their inherently greater surface area and tendency for agglomeration.

There are multiple other factors which have the potential of impacting the final mechanical properties of the casting product, such as wettability between the melt and reinforcement, as well as the presence of porosities within the final component. While these issues can have potentially detrimental effects on properties of the composite, they lie outside the scope of this paper and as such, will not be discussed here.

Table 6 presents a chronological overview of the studies on the fabrication of Al/graphene composites using casting methods. Bhanu Prakash et al. produced Al7075/graphene composite using stir casting. Al7075 alloy was first melted between the
temperatures 700–750°C in a graphite mold and then 0.5, 1, 1.5, 2 wt% graphene was added to the melt. Afterwards, the melt was agitation with the speed of 600–650 rpm using mild steel blades. It was observed that graphene was uniformly dispersed at the boundaries of Al alloy dendrites. Praveen Kumar et al. attempted to incorporate graphene and SiC particles into the Al alloy matrix using ultrasonic cavitation-assisted stir casting process. A356 alloy was melted under N2 atmosphere and then pre-heated reinforcement particles were added to the melt. After stir casting for 5 min at 630°C, the melt was ultrasonicated for 10 min. Finally, the slurry was poured into pre-heated metallic mold. The application of ultrasonic vibration on the composite during melting refined the grain structure of the Al alloy matrix and improved the distribution of reinforcement nanoparticles.

Alipour et al. employed a combination of ball milling and ultrasonic-assisted stir casting in order to create a uniform distribution of GNPs within the Al7068 alloy matrix. The Al powder and the GNP dispersion were ball-milled to produce Al/GNP composite powder. Concurrently, the Al7068 ingot was melted at ~750°C. Pre-heated Al/GNP powder was added to the Al alloy melt and then stirred for 10–15 min with the help of a mechanical stirrer before being sonicated for 1 min with 2000 W ultrasonic waves. After preparing the composite melts containing various amounts of GNPs (0.1–1 wt%), the slurries were poured into permanent molds. The microstructure of the Al7068 matrix composites reinforced with GNPs is shown in Figure 15. As can be observed, the addition of GNPs to the Al alloy significantly refines the coarse columnar primary α-Al grains to fine equiaxed ones. The grain refinement is related to the presence of GNPs which act as nucleation sites during the solidification of α-Al grains. Nevertheless, as the amount of GNP is exceeded 0.5 wt%, the Al grain size is not changed further.

In another study conducted by Li et al., Al/graphene composites were produced using a combination of ball milling, cold pressing, continuous casting, and rolling. First, pure Al and GNP powder particles were ball-milled to produce Al-2 wt% GNP composite
After cold compaction of the powder mixture, it was added to Al melt with a mass proportion of 1:9, and finally Al-0.2 wt% GNP composite samples were obtained through continuous casting and the subsequent rolling of the components. GNPs are homogeneously distributed in the as-cast specimen and then transformed into fibers orientated to the rolling direction after deformation to form a lamellar structure, as shown in Figure 16a and 16b. The interface between GNPs and Al is well-bonded in general and no trace of Al4C3 is detected at the interface (Figure 16c), but a few micro-cracks are observed in the microstructure.

### 2.3. Severe plastic deformation (SPD)

SPD technique is a method of producing ultrafine-grained (UFG) materials by subjecting them to significantly large strains below the recrystallization temperature and without subsequent heat treatment.\(^{110-113}\) Materials with ultrafine grain structures have attracted a great deal of scientific interest and attention due to their superior mechanical and physical properties.\(^{114-117}\) However, traditional processing techniques such as drawing, forging, extrusion and rolling, along with their corresponding equipment, face limitations in exerting sufficient levels of strain to cause a homogenous ultrafine grain structure.\(^{111,112,118-120}\) In comparison with common methods of material processing using plastic deformation, SPD methods are capable of creating a uniform nanostructured material with the proper mechanical properties. Moreover, while cracks and fractures are common in samples exposed to sizeable magnitudes of plastic deformation, it should be noted that proper die design and mode of load application in SPD can allay this problem. On the other hand, the more recent methods of producing nanocrystalline materials, such as high-energy ball milling and inert gas condensation, have proven incapable of large-scale production of UFG materials with a uniform structure.\(^{111,112,118,119,121,122}\)

Furthermore, the presence of impurities and porosities within the structure, which commonly occurs during consolidation by plastic deformation techniques, is almost entirely eliminated in SPD processing methods.

Although SPD processing techniques have been frequently used for achieving grain refinement in numerous metallic materials, the processing of MMCs via SPD has garnered interest only over the last decade. Applying SPD has two main effects on MMC materials including a significant grain refinement of the metallic matrix and an improvement in the distribution of reinforcement particles within the matrix, which results in enhancing the mechanical properties of MMCs.

Table 7 presents a number of recent studies utilizing SPD processing methods for the production of AMCs reinforced with graphene. The relevant process details for each individual SPD technique are also included in the table. Common methods of SPD processing for the production of Al/graphene composites include friction-stir processing (FSP) and high-pressure torsion (HPT), which are briefly described in the following.

#### 2.3.1. Friction stir processing (FSP)

FSP is among the SPD processes mostly used for modification of microstructure in near-surface layers of metallic...
components. In this process, a rotating tool is plunged into a work-piece and then moved in the direction of interest. The tool serves two primary functions of heating and deformation of work-piece material. The heat is primarily generated by the friction of the rotating shoulder with the work-piece, while the rotating pin stirs the heated material. The heated material softens and flows around the rotating pin and then fills the cavity at the rear end of the tool. The material that flows around the tool is subjected to intense plastic deformation and thermal exposure, which leads to significant refinement of microstructure in the processed zone. It should be noted that unlike most SPD techniques which modify bulk properties, SPD through FSP involves only surface modification while the bulk material structure and properties are retained.128–131

FSP is the most widely used method among SPD techniques for the production of Al/graphene composites with improved mechanical properties. Maurya et al.125 studied the effect of carbonaceous reinforcement phases, namely graphite, graphene, and CNT, on mechanical properties of Al6061 using FSP processing. Prior to FSP treatment, the reinforcement was applied into a groove created on the surface of Al6061 sheet in the form of a homogeneous paste of silicone oil and the corresponding carbonaceous particles. FSP was performed at a rotational speed of 1100 rpm and a traverse speed of 0.2 mm min⁻¹. To inhibit grain growth, the samples were quenched in ice water after completion of the FSP treatment. Raman spectra of carbonaceous powders and FSPed samples are shown in Figure 17. The appearance of D-band, G-band, and 2D-band after FSP processing confirms the retention of carbonaceous reinforcement in the stir zone. However, the intensity of G-band is decreased after FSP, indicating the damage accumulation in carbonaceous particles due to the intense plastic deformation and shear stresses involved in the FSP. On the other hand, the I_D/I_2D ratio of composites is observed to decrease when compared with that of carbonaceous powders, indicating exfoliation of graphene sheets as a result of the increased stresses between the graphene layers during the FSP processing.

The dispersion and strengthening effect of graphene on an Al alloy matrix produced using PM and FSP were investigated by Zhang et al.127 First, the GNPAs were mixed with Al2009 alloy powder particles through solution-assisted mixing route. Following the cold compaction, consolidation of composite powders was carried out using hot pressing followed by hot forging. Finally, the plates were subjected to FSP for 1–4 passes at a tool rotational speed of 2000 rpm and a travel speed of 100 mm min⁻¹. Figure 18a presents the Raman spectra of Al2009/GNP powder and composite fabricated under different conditions. It is evidenced that the FSP treatment increases the I_D/I_G ratio, and this value is boosted as the number of FSP passes increases. This proves that the FSP treatment has introduced defects and disorders within the composite structure. Microstructural examinations indicated that the GNPs distribution within the Al alloy matrix was considerably improved as the number of FSP passes increased due to the intense stirring incurred by the FSP. The effect of FSP processing on the tensile stress-strain curve of Al2009-1 wt% GNP composite is depicted in Figure 18b. As can be observed, tensile properties of FSPed composite are significantly improved in comparison to the forged sample due to the reduced number and size of voids and improved
distribution of GNPs. The two-pass FSP composite exhibits the highest strength and ductility, but further increasing the number of FSP passes results in declined tensile properties. This is attributed to the noticeable breaking up of GNPs and thus reducing their lateral size after a large number of FSP passes, as shown in Figure 18c and 18d.

Dixit et al. manufactured multi-layer graphene-embedded AMCs by direct exfoliation of graphite into graphene using FSP technique. An Al-40 at% graphite powder mixture was filled in a groove on Al plate and then FSP was carried out for eight passes. The exfoliation of graphite, and thus the presence of graphene layers with a thickness of $\frac{1}{24} \text{nm}$ in the Al matrix, was confirmed by Raman spectroscopy, TEM, and AFM techniques.

### 2.3.2. High-pressure torsion (HPT)

In this process, a disk-shaped sample is held between two anvils and strained in torsion under significant applied pressure. A lower holder is rotated, and the surface friction forces deform the material through shear. Due to the specific geometric shape of the sample, the main volume of the material is strained in conditions of quasi-hydrostatic compression under the applied pressure. Thus, the deformed sample remains relatively intact in spite of the sizeable strain. One of the main characteristics of HPT is that an exceptional level of grain refinement, often to the nanometer level and thus an extremely high strength, can be attained.

Nevertheless, a major drawback of HPT is that the imposed strain varies across the sample, therefore the microstructure is tremendously inhomogeneous. By adjusting the processing parameters like the number of turns and the applied pressure, the inhomogeneity of microstructure can be lowered.

Zhao et al. investigated the role of graphene and CNT on the microstructure and mechanical properties of AMCs produced using HPT technique. Initially, Al reinforced with 0.25 and 0.5 wt% graphene or CNT composite powders were produced by powder mixing method including ultrasonic-assisted mixing in ethanol followed by ball milling. The resulting powders were later cold-pressed and consolidated using HPT
technique at room temperature for 10 revolutions under 3 GPa with a rotational speed of 2 rpm. Figure 19 illustrates the TEM images and grain size distribution curves for pure Al, Al-0.5 wt% CNT, Al-0.25 wt% graphene, and Al-0.5 wt% graphene. As can be observed, a UFG microstructure is obtained for...
each sample, but the degree of grain refinement is more pronounced in the composite samples. Furthermore, the grain size is decreased with increasing the number of graphene platelets in the Al/graphene composite. This is ascribed to the pinning effect of graphene and CNT on the dislocations which hinders the motion of grain boundaries. As shown in Figure 19f and 19h, graphene is more effective than CNT when it comes to grain refinement. This is attributed to the role of size and morphology of reinforcement on grain refinement, meaning that reinforcement particles with smaller size and higher specific surface area, i.e. graphene, resulting in a finer microstructure.28

In another study carried out by Czeppe et al.,60 HPT was used in tandem with mechanical milling for the production of Al/graphene composite. First, Al-0.05 wt% graphene composite powder was prepared by ball milling, and the powder mixture was then consolidated using HPT under pressure of 4 GPa in 5 and 10 revolutions. The experimental results show that HPT process influences the structure of graphene by increasing its defects density and reducing its dimensions. Furthermore, a higher number of rotations results in a greater magnitude of strain being imposed on the microstructure, subsequently leading to more grain refinement in the Al matrix.

2.4. Additive manufacturing.
Powder-based additive manufacturing (AM) techniques have emerged as a novel method of producing specialized composites, namely MMCs. The process involves the joining of materials to make objects from a three-dimensional (3D) model data, usually layer-by-layer, as opposed to subtractive manufacturing methods. This process is also referred to as freeform fabrication, layer manufacturing, and additive layer manufacturing.135

Selective laser melting (SLM) is one of the additive manufacturing processes that has been recently brought into the spotlight for fabricating MMCs. It is a procedure during which metallic or composite components are made from powders. In this method, high-intensity laser is used to melt and subsequently fuse selective regions of powder, layer-by-layer, in accordance with predetermined computer-aided design (CAD) data.136 The building chamber in SLM process is often filled with inert gas, such as nitrogen or argon, in order to protect the heated parts against oxidation. Studies show that SLM is fully capable of melting the powders and producing dense net-shape components without the need for secondary processing steps. In comparison to selective laser sintering (SLS) process which binds powders via solid-state sintering resulting in high porosity and low strength, SLM is a superior AM procedure.136

Recently, SLM has been utilized by Hu et al. to fabricate 3D-printed graphene-reinforced AMCs.67 Initially, Al powders and graphene sheets were mixed together using a planetary ball mill to produce composite powders including 0.5, 1, and 2.5 wt% graphene. The resulting powders were then placed upon a substrate made of Al6061, where a pulsed laser beam with a scanning area of 5 mm × 5 mm was used to melt the powders under the Ar atmosphere. This step was repeated until a 3D-printed structure was achieved. Following the laser processing on composite powders, it was discovered that the number of defects present on the graphene sheets has increased, as indicated by the increased ID/IG ratio after SLM in comparison to the original graphene Raman spectrum. This phenomenon has been ascribed to the thermal accumulation during SLM processing. Moreover, Al4C3 phase was witnessed to have formed within the matrix throughout the process. It should be noted that there are contradictory reports on this issue, with previous studies indicating a decrease in the amount of defects on graphene sheets.137 Overall, the hardness (micro and nano-) of 3D-printed composite samples was found to be enhanced in proportion with graphene content, where the sample reinforced with 2.5 wt% graphene exhibited a microhardness value of 66.6 HV, in comparison to pure Al with 38 HV, resulting in roughly 75% improvement of hardness.

3. Mechanical properties of Al/graphene composites
Due to the excellent mechanical properties of graphene, it can serve as an outstanding candidate for usage as a strengthening component in the composites. Indeed, enhancement of mechanical properties, such as strength, hardness, and Young’s modulus, can be regarded as the most important purpose of hybridizing light metals like Al with graphene. The significant improvement in mechanical properties of AMCs by the addition of graphene into the Al matrix has been stated frequently.28,31,41,58,79–81 Increment in mechanical behavior is normally considered as a result of the proper dispersion of graphene within the matrix and strong graphene/Al interface, which leads to grain refinement and efficient load transfer. Nevertheless, the degradation of mechanical properties owing to graphene agglomeration, detrimental
chemical reactions, or highly defective structure of graphene has been affirmed in some cases.²⁶,¹³⁸

The majority of the studies undertaken with the purpose of investigating the effects of graphene additives on mechanical properties of AMCs have utilized relatively small amounts of graphene as reinforcement phase of choice.⁴⁴,⁴⁸,⁵⁷,⁵⁹,⁶⁵–⁹⁹,¹³⁹–¹⁴⁵ The reason for low graphene content is mainly the difficult dispersion of reinforcement particles and risk of formation of agglomerates within the Al matrix, which would severely hamper the mechanical properties of Al/graphene composites. Consequently, a general trend is usually observed in which the mechanical properties of the composite begin to deteriorate after a threshold in graphene concentration, where the formation of agglomerations within the matrix, and the subsequent focus of stress on them, results in the nucleation of cracks and pores within the structure. In spite of various modern techniques successfully overcoming this drawback at lower concentrations of graphene, no remarkable progress has been made as of yet when it comes to large quantities of graphene additives. On the other hand, various factors such as the graphene specifications, the amount of graphene content along with the method of production play an essential role in the resulting mechanical properties of the final product. For instance, it is stated that thinner GnP particles from 1 to 3 wt% into the matrix, while larger GNPs had a greater tendency of forming agglomerates, thus proving detrimental to mechanical properties.³⁴

Table 8 summarizes the mechanical properties of Al/graphene composites which have been reported in the literature. In the following sections, the impact of graphene on mechanical properties of AMC materials is investigated, while different mechanisms responsible for improvement or degradation of these properties are elucidated.

3.1. Effect of graphene on hardness and strength of AMCs

3.1.1. Hardness
The hardness of Al/graphene composites is usually measured by indentation methods. Figure 20a shows the variations of Vickers hardness of Al/GnP composites as a function of the GnP content. It is clear that the addition of GnP particles from 1 to 3 wt% into pure Al increases the hardness of the composites. The continuous increment of hardness with increasing the amount of graphene content has also been reported for AMCs reinforced with 0.25–1 wt% GNSs⁶⁵ and 1–2 wt% graphene sheets.¹⁰⁴ The improved hardness of Al/graphene composites is ascribed to the presence of graphene having extraordinary mechanical properties, and providing high restraining force for deformation during indentation.⁶⁵

Nevertheless, a different trend of hardness variations has been identified for AMCs reinforced with 1 to 5 wt% GNPs. Figure 20b depicts the effect of GNPs content on the average hardness of composites measured by nanoindentation test. As can be observed, reinforcing the Al with GNPs up to 1 wt% increases the hardness of the composite. This improvement is attributed to the homogeneous dispersion of GNPs, which shares the load transferred from the Al matrix and provides the matrix with high resistance against deformation. However, GNPs content of >1 wt% reduces the hardness of composites due to the increase of porosity and agglomeration of GNPs.⁸³

3.1.2. Strength
The strength of Al/graphene composites can be evaluated by conventional tests including tension, compression, and bending. However, uniaxial tensile testing is the most commonly used test. The representative experimental results of tensile test for two Al/graphene composites are presented in Figure 21. As can be seen in Figure 21a, both the yield stress and ultimate tensile strength (UTS) are significantly enhanced by the introduction of FLG into the Al matrix. For instance, with the addition of only 0.7 vol% FLGs, the yield stress of the composite increases by ~71.8% over the monolithic Al. Considering the minute quantity of graphene has been added into the matrix, the strength enhancement induced by graphene is considerable.

Several studies have attempted to compare graphene with other reinforcements in terms of the level of increase in the matrix strength as a result of reinforcement addition or strengthening efficiency.¹⁴⁷,¹⁴⁸ The strengthening efficiency of a reinforcement incorporated in a matrix can be evaluated by the following relationship¹³⁸:

\[
\eta = \frac{\sigma_c - \sigma_m}{V_f \sigma_m}
\]  \hspace{1cm} (1)

where \(\sigma_c\) and \(\sigma_m\) are the yield stress of the composite and the matrix, respectively and \(V_f\) is the volume fraction of the reinforcement. Zhao et al.¹²⁴ investigated the effect of graphene and CNT addition on the tensile behavior of AMCs. Figure 22 shows the tensile stress-strain curves for Al/FLG and Al/CNT composites. As can be observed, reinforcing with FLG and CNT results in a considerable strength improvement.
<table>
<thead>
<tr>
<th>Year</th>
<th>Composite material</th>
<th>Hardness</th>
<th>Tensile yield stress (MPa)</th>
<th>Ultimate tensile strength (MPa)</th>
<th>Compressive strength (MPa)</th>
<th>Flexural Strength (MPa)</th>
<th>Elongation (%)</th>
<th>Failure strain (%)</th>
<th>Young’s modulus (GPa)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>2000</td>
<td>Al7050-0.33, 0.55, 0.77 wt% graphene</td>
<td>Decreased from 85.9 to 57.1 BHN (−33%)</td>
<td>–</td>
<td>Increased from 90 to 145 (+61%)</td>
<td>–</td>
<td>Increased from 150 to 210 (+40%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>101</td>
</tr>
<tr>
<td>2011</td>
<td>Al-0.1 wt% graphene</td>
<td>Increased from 83 to 99 VHN (+19%)</td>
<td>–</td>
<td>Decreased from 330 to 270 (−18%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>Decreased from 5.5 to 2.5 (−45%)</td>
<td>–</td>
<td>31</td>
</tr>
<tr>
<td>2011</td>
<td>Al-1, 2, 3 wt% GNP</td>
<td>Increased from 45 to 75 VHN (+66%)</td>
<td>–</td>
<td>–</td>
<td>Increased from 145 to 175 (−21%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>79</td>
<td></td>
</tr>
<tr>
<td>2012</td>
<td>Al-0.3 wt% GNS</td>
<td>–</td>
<td>–</td>
<td>Increased from 154 to 249 (+62%)</td>
<td>–</td>
<td>Decreased from 25 to 13 (−48%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>58</td>
</tr>
<tr>
<td>2012</td>
<td>Al-1, 3, 5 wt% GNP</td>
<td>Increased from 50 to 70 VHN (+40%)</td>
<td>–</td>
<td>–</td>
<td>Increased from 145 to 180 (+24%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>80</td>
<td></td>
</tr>
<tr>
<td>2012</td>
<td>Al124-2, 4, 6 wt% graphene</td>
<td>Increased from 180 to 265 VHN (+47.5%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>41</td>
<td></td>
</tr>
<tr>
<td>2014</td>
<td>Al6061-1 wt% graphene</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>Decreased from 13.4 to 9.53 (−40.6%)</td>
<td>–</td>
<td>–</td>
<td>30</td>
<td></td>
</tr>
<tr>
<td>2014</td>
<td>Al-0.25, 0.5, 1 wt% GNP</td>
<td>Increased from 39 to 93 VHN (+138%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>28</td>
<td></td>
</tr>
<tr>
<td>2014</td>
<td>Al-0.3 wt% rGO</td>
<td>Increased from 1.34 to 1.59 GPa (+18%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>77</td>
<td></td>
</tr>
<tr>
<td>2014</td>
<td>Al3.9 Cu-1.5 Mg)-0.15, 0.5 wt% GNF</td>
<td>–</td>
<td>Increased from 214 to 319 (+49%)</td>
<td>Increased from 373 to 467 (+25%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>59</td>
<td></td>
</tr>
<tr>
<td>2014</td>
<td>Al-0.3 wt% GNP</td>
<td>Increased from 76 to 85 VHN (+11.8%)</td>
<td>Increased from 170 to 195 (14.7%)</td>
<td>Increased from 493 to 457 (−7.8%)</td>
<td>–</td>
<td>Decreased from 13.4 to 9.53 (−40.6%)</td>
<td>–</td>
<td>–</td>
<td>81</td>
<td></td>
</tr>
<tr>
<td>2014</td>
<td>Al5052 (H32)-GO</td>
<td>–</td>
<td>Decreased from 218 to 192 (−13%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>123</td>
<td></td>
</tr>
<tr>
<td>2014</td>
<td>Al-0.25, 0.5 wt% graphene</td>
<td>Increased from 49 to 122 VHN (+149%)</td>
<td>–</td>
<td>Increased from 157 to 197 (−25%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>146</td>
<td></td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.5, 1, 1.5, 2 wt% GNF</td>
<td>–</td>
<td>Increased from 125 to 190 (52%)</td>
<td>Increased from 145 to 245 (+69%)</td>
<td>–</td>
<td>Decreased from 50 to 20 (−60%)</td>
<td>–</td>
<td>–</td>
<td>33</td>
<td></td>
</tr>
<tr>
<td>2015</td>
<td>Al7050-0.3, 0.5, 0.7 wt% graphene</td>
<td>Increased from 54 to 60 HRC (+11%)</td>
<td>–</td>
<td>Increased from 90 to 160 (+78%)</td>
<td>–</td>
<td>Increased from 30 to 40 (+33%)</td>
<td>–</td>
<td>–</td>
<td>102</td>
<td></td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.3, 0.5, 0.7 vol% FLG</td>
<td>–</td>
<td>Increased from 262 to 440 (+72%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>Decreased from 13 to 3 (−77%)</td>
<td>–</td>
<td>30</td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.1, 0.25, 0.5 wt% rGO</td>
<td>Increased from 17 to 32 VHN (+88%)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>82</td>
<td></td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.05 wt% graphene</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
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</tr>
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</table>

(Continued)
<table>
<thead>
<tr>
<th>Year</th>
<th>Composite material</th>
<th>Mechanical property</th>
<th>Ref.</th>
</tr>
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<tbody>
<tr>
<td>2016</td>
<td>Al6061-Graphene</td>
<td>Increased from 0.9 to 1.5 GPa (+66%)</td>
<td>125</td>
</tr>
<tr>
<td>2016</td>
<td>Al-1 wt% GNP</td>
<td>Decreased from 111 to 98 VHN (-13%)</td>
<td>4</td>
</tr>
<tr>
<td>2016</td>
<td>Al356-5 wt% SiC + 1, 3, 7 wt%</td>
<td>Increased from 66 to 77 Rockwell (+16%)</td>
<td>103</td>
</tr>
<tr>
<td>2016</td>
<td>Al-1, 2 wt% graphene</td>
<td>Increased from 41.4 to 47.6 (+15%)</td>
<td>104</td>
</tr>
<tr>
<td>2016</td>
<td>Al6063-2, 4, 6 wt% graphene</td>
<td>Increased from 58 to 101 VHN (+74%)</td>
<td>106</td>
</tr>
<tr>
<td>2016</td>
<td>Al6061-0.5, 0.7, 1 wt% Ni nanoparticle-decorated graphene</td>
<td>Increased from 50.5 to 67.2 VHN (+33%)</td>
<td>62</td>
</tr>
<tr>
<td>2016</td>
<td>Al-0.5 wt% graphene</td>
<td>Increased from 92 to 180 (+95.6%)</td>
<td>105</td>
</tr>
<tr>
<td>2017</td>
<td>Al6061-2 vol% graphene</td>
<td>Almost constant</td>
<td>84</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.2 vol% rGO</td>
<td>Increased from 31 to 98 VHN (+216%)</td>
<td>85</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5, 1 wt% GNP</td>
<td>Increased from 44 to 57 VHN (+29%)</td>
<td>86</td>
</tr>
<tr>
<td>2017</td>
<td>Al-1 wt% FLG or FLGO</td>
<td>Increased from 28 to 58 VHN (+107%)</td>
<td>39</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.3, 0.6, 0.9, 1.2 wt% graphene</td>
<td>Increased from 65 to 98 VHN (+51%)</td>
<td>63</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5, 1, 2 wt% graphene</td>
<td>Increased from 75 to 88 BHN (+10%)</td>
<td>64</td>
</tr>
<tr>
<td>2017</td>
<td>Al-40 at% graphene</td>
<td>Increased from 33 to 48 VHN (+45%)</td>
<td>126</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5, 1, 3, 5 wt% GNP</td>
<td>Increased from 68 to 83 GPa (+22%)</td>
<td>83</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5, 0.25, 0.5, 1 wt% GNS</td>
<td>Increased from 81 to 112 (+38.3%)</td>
<td>65</td>
</tr>
<tr>
<td>2017</td>
<td>AlMg5-1 vol% GO</td>
<td>Increased from 260 to 556 (+138%)</td>
<td>66</td>
</tr>
<tr>
<td>Year</td>
<td>Alloy Composition</td>
<td>FLG, GNP, GO %</td>
<td>VHN</td>
</tr>
<tr>
<td>------</td>
<td>------------------</td>
<td>----------------</td>
<td>-----</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.05, 0.2, 0.4, 1 vol% FLG</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0, 0.5, 1, 2.5 wt% graphene</td>
<td>Increased from 38 to 66.6 VHN (±75.3%)&lt;sup&gt;*&lt;/sup&gt;</td>
<td>–</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.1, 0.2, 0.3 wt% graphene</td>
<td>Increased from 42 to 52 VHN (±24%)&lt;sup&gt;*&lt;/sup&gt;</td>
<td>–</td>
</tr>
<tr>
<td>2018</td>
<td>Al7075-0.5, 1, 1.5, 2 wt% graphene</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.2 wt% GNP</td>
<td>Increased from 37.4 to 43.6 (±16%)</td>
<td>–</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.05, 0.1, 0.2 wt% GO</td>
<td>Increased from 33 to 37 VHN (±12%)&lt;sup&gt;*&lt;/sup&gt;</td>
<td>–</td>
</tr>
<tr>
<td>2018</td>
<td>Al-1 wt% GNP</td>
<td>120 VHN</td>
<td>–</td>
</tr>
</tbody>
</table>

<sup>*</sup> Optimal sample.
of the monolithic Al, but higher strengthening is achieved with the introduction of graphene in the same amount of reinforcement. In another research conducted by Shin et al., FLG was found to be ~3.5 orders of magnitude more effective than CNT in the strengthening of Al. The higher strengthening efficiency of graphene is the result of its larger specific surface area compared with CNT due to the geometric difference. Moreover, several studies have shown that the strengthening efficiency of graphene in AMCs significantly exceeds that of any other reinforcement, affirming the huge potential of graphene
as the ideal reinforcement in AMCs. Thus, graphene has a greater impact on the load transfer from the matrix, coarsening of matrix grains during thermal processing, and restraining the dislocation motion and crack propagation during tensile testing.59,61

One of the key points regarding the strengthening efficiency of graphene-reinforced AMCs is the amount of graphene content added to the matrix. Generally, Al/graphene composites have been investigated for various graphene contents ranging from 0.05 to 7 wt%. It can be deduced from Figure 21a that the strength is boosted as the graphene volume fraction in the Al matrix increases. Similar observations have been reported for Al-(0.25 and 0.5) wt% GNF59 Al-(0.25 and 0.5) wt% graphene124 and Al-(1 and 2) wt% graphene104 composites as well. However, in some other researches, it has been observed that the strength of composite begins to deteriorate after a threshold in the graphene amount. Figure 21b shows the tensile stress-strain curve of Al/GNP composites with various amounts of reinforcement particles. It is clear that GNPs increase the strength of the composite up to 1 wt% GNP addition. Nonetheless, a further increase in the concentration of GNPs continuously degrades the strength (Figure 21c). The highest improvement of ~85% in the yield stress and ~55% in the UTS is achieved for Al-1 wt% GNP composite which is the outcome of good dispersion of GNPs and strong interfacial bonding between Al and GNPs. The reduction of strength at higher concentrations of graphene is ascribed to the agglomeration of GNPs, which generates irregularities, leading to stress concentration and generates porosity, making interfacial GNP/Al bonding weak.83 Similar results have been reported by Li et al.149 and Li and Xiong,65 in which the optimal amount of graphene content for achieving the highest strength was determined to be 1 wt% GNF and 0.25 wt% GNS, respectively.

Li et al.33 attempted to calculate the optimum amount of graphene content in order to achieve the highest strength in AMCs. Their proposed model is schematically shown in Figure 23. The assumptions adopted before calculation are: (a) Al matrix grains are equiaxed; (b) graphene flakes are uniformly dispersed along the grain boundaries of the Al matrix; (c) each Al grain is coated evenly by graphene flakes; (d) graphene flakes are multi-layers without overlapping. Based on these assumptions, the surface area of Al grains should be equal to that of graphene flakes within the Al/graphene composite, which can be written as the following equation:

\[
m_c A_f w_f = \frac{m_c (1-w_f)}{n} = \frac{\rho_m}{\frac{4}{3} \pi r^3} 4 \pi r^2
\]

where \(m_c\) is the mass of the composite, \(w_f\) the weight fraction of graphene, \(A\) the specific surface area of the single-layer graphene, \(n\) the number of graphene layers, \(\rho_m\) the density of Al, and \(r\) the radius of Al grains. With the simplification of Equation (2), the optimum weight fraction of graphene can be calculated as:

\[
w_f = \frac{1}{1 + \frac{A \rho_m}{3n}}
\]

The calculated content of graphene flakes (~4 wt%) was found to be higher than the appropriate experimental value (1 wt%) obtained by Li et al. mainly due to the inhomogeneous dispersion of graphene along the Al grain boundaries, agglomeration of graphene.

Figure 22. Tensile stress-strain curves for pure Al, Al/FLG, and Al/CNT samples prepared by HPT (Reprinted with permission).124

Figure 23. Schematic model for calculation of the optimum amount of graphene content in Al/graphene composite (Reprinted with permission).33
and non-uniform size and morphology of the Al grains. Nonetheless, Equation (3) can provide insight into the selection of reinforcement content for preparing Al/graphene composites.

Another issue which should be considered is that the efficiency of graphene in reinforcing the Al matrix can be significantly affected by the loading type. The tensile and compressive stress-strain curves of Al-0.3 wt% GNP together with pure Al are shown in Figure 24. It can be observed that the tensile yield stress and UTS of the Al/GNP composite are higher than those of the unreinforced Al. Nevertheless, the ultimate compressive strength of the Al/GNP composite is lower than that of the pure Al while their compressive yield stress is almost identical. Lower compressive strength of Al/GNP composite can be attributed to the flake buckling, bending, and even breakup of 2D flexible graphene sheets upon compressive loading which considerably degrades the strengthening effect of GNPs in the Al matrix. It has also demonstrated that the addition of 1 vol% of GO significantly improves the tensile and bending strengths of an AlMg5 alloy. However, the degree of improvement is not identical so that the UTS and bending strength of the composite are increased almost twofold and fourfold over those of the pure bulk AlMg5 alloy, respectively.

3.2. Strengthening mechanisms

Usually, there are two strengthening mechanisms that are associated with MMCs: (a) indirect strengthening resulted from the influence of reinforcement on the matrix microstructure or deformation mode and (b) direct strengthening caused by the load transfer from the metal matrix to the reinforcement. The strengthening effect in Al/graphene composites is the direct result of the incredibly high strength of graphene itself and largely depends upon its uniform distribution within the matrix and good bonding with the Al. The enhanced strength generated in graphene-reinforced AMCs can be attributed to (a) refined grains, (b) high density of dislocations, (c) Orowan looping, and (d) load bearing from the Al matrix. In the following, an overview of different strengthening mechanisms active in graphene-reinforced AMCs is concisely presented.

3.2.1. Grain refinement

Grain refinement is a major factor of enhanced strength in different metals and composites. Most famously represented by the well-established Hall-Petch relationship, where strength is increased as the grain size is decreased. The strength improvement through grain refinement ($\Delta \sigma_d$) can be expressed by:

$$\Delta \sigma_d = \frac{k_y}{\sqrt{D}}$$

where $k_y$ is the locking parameter depending on the material of choice and $D$ is the average grain size of the matrix.

The graphene particles either located on grain boundaries or dispersed within the grains pin and hinder the movement of dislocations during plastic deformation, effectively suppressing grain growth during thermal processing and resulting in a finer grain structure. Smaller grains lead to the presence of more grain boundaries within the structure, thus increasing the strength of the composite material. Zhao et al. have reported a significant boost to
UTS from 157 to 197 MPa as a result of grain refining effect from 498 to 112 nm after addition of 0.5 wt% GNP to the Al matrix (see Figure 19).

3.2.2. Dislocation strengthening

The increase in density of dislocations as a result of the difference in the coefficient of thermal expansion (CTE), also referred to as CTE mismatch, of matrix and reinforcement can also induce strengthening to the final composite product. The strength gains through this mechanism, $\Delta \sigma_{\text{CTE}}$, can be formulated as:

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

where $\Delta T$ is the temperature change, $\Delta C$ is the CTE difference between matrix and reinforcement, $f_v$ is the volume fraction of reinforcement phase, and $d_p$ is the average size of reinforcement particles.

According to Table 1, Al and graphene have a significant CTE mismatch which would result in the prismatic punching of dislocations at the interface. Since dislocation density depends on the surface area of reinforcement particles, graphene with a high surface area can effectively enlarge the number of created dislocations. These newly generated dislocations impose a stress field in the opposite direction of the movement of original dislocations, impeding and hindering their further movement within the structure, resulting in the enhanced strength of the composite.

According to Equation (5), when the composites are processed at elevated temperatures (high $\Delta T$), the CTE mismatch is expected to play a significant role in the strengthening. Thus, during the room temperature processing of Al/graphene composites (for example via HPT), the temperature elevation is not high enough to take the CTE mismatch strengthening into account.

3.2.3. Orowan looping

Orowan looping mechanism is caused by the restricted movements of dislocations due to the presence of fine precipitates. As a result of the collision of dislocations with these precipitates, dislocations within the grain tend to bend, and eventually form semi-circular shapes. Afterwards, the distorted dislocations form rings around precipitate particles, referred to as “Orowan loops”, further suppress the movement of dislocations, resulting in a significant degree of work hardening. The strength gain achieved through Orowan mechanism is formulated in Equation (6):

$$\Delta \sigma_p = \frac{0.4 M G b \ln \left(\frac{d_p}{3} \sqrt{\frac{F_0}{\tau_m}} \sqrt{1 - \frac{\nu}{3}}\right)}{\pi d_p \sqrt{\frac{2}{3} \left(\frac{F_0}{\tau_m} - 1\right)} \sqrt{1 - \nu}} \quad (6)$$

in which $M$ is the Taylor factor, $d_p$ is the average distance between reinforcement particles and $\nu$ is Poisson’s ratio.

Graphene sheets can be considered as non-shearable precipitates which hinder the movement of dislocations. Orowan looping is probably the most common mechanism of enhancing strength and hardness in graphene-reinforced AMCs, especially in composites produced by high amounts of shear stresses. For this purpose, the graphene particles need to be dispersed in a uniform manner within the microstructure, where they are able to effectively hinder the movement of dislocations. It should be noted that graphene content above a certain threshold will result in the formation of agglomerates and deterioration of mechanical properties, therefore the amount of graphene additives need to be optimized.

3.2.4. Load transfer

For composites reinforced with short fibers or whiskers, an applied force is transferred from the matrix to the reinforcement by means of shear stresses developed along with the reinforcement/matrix interface. In this case, the shear lag model is usually used to estimate the strength of composites assuming (a) the perfect contact between the matrix and reinforcements and (b) the alignment of fiber reinforcements to a single direction. Kelly-Tyson’s formula is expressed as:

$$\sigma_c = \sigma_f V_f \left(1 - \frac{l_c}{2l}\right) + \sigma_m (1 - V_f) \quad (7)$$

$$\sigma_c = \sigma_f V_f \left(\frac{l_c}{2l}\right) + \sigma_m (1 - V_f) \quad (8)$$

where $\sigma_c$ and $\sigma_m$ are the strength of the composite and the matrix, respectively; $\sigma_f$ and $l$ are the strength and average length of the fiber, respectively; $l_c$ is the critical fiber length, indicating the minimum length in which the fiber is loaded up to its fracture strength. The critical fiber length can be calculated as:

$$l_c = \frac{d \sigma_f}{2 \tau_m} \quad (9)$$

where $d$ is the fiber diameter and $\tau_m$ is the shear strength of the matrix ($\sim 0.5 \sigma_m$). Considering the 2D nature and high specific surface area of graphene sheets, Shin et al. modified the shear lag model and derived the following equation for $l_c$:

$$l_c = \frac{d \sigma_f}{2 \tau_m} \quad (9)$$
where \( A \) and \( S \) are the cross-sectional area and interfacial area of graphene sheets, respectively. Assuming that the length, width, and thickness of a graphene sheet is \( l \), \( w \) and \( t \), respectively, \( A \) and \( S \) are \( wt \) and \( 2(w + t)l \), respectively. If the reinforcement length exceeds the critical length \( (l \geq l_c) \), the reinforcement carries stress equal to its tensile strength throughout a certain length \( (l - l_c) \). Considering that the thickness of graphene sheets is usually much smaller than their width, Equation (10) can be simplified as,

\[
l_c = \frac{Al_{ef}}{2\tau_m}
\]

The stress transfer from the matrix to the graphene sheets can be evidenced by the fracture surface morphology of Al/graphene composites, as shown in Figure 25. It can be observed that dimples are elongated in the direction of tensile loading and some graphene sheets are pulled out at the edges of the dimples.

As was stated before, an efficient transfer of stress from the composite matrix to the reinforcement depends solely on the strength of interface existing between these two phases. Furthermore, the load transfer efficiency relies on the surface area of the reinforcement, i.e. the reinforcement with a higher surface area will carry a larger amount of load. 2D structure, crumpled surface and large surface area of graphene are beneficial to enhancing the interfacial contact with the Al matrix and thus load transfer efficiency. In order to further improve the interfacial bonding between the Al matrix and graphene, a few methods have been suggested including changing the morphology of Al matrix powders from spherical to flake and increasing their specific surface area via ball milling, using graphene sheets with larger lateral size, or decoration of graphene sheets with metallic nanoparticles.

### 3.3. Theoretical strength of Al/graphene composites

Based on the strength of matrix Al and the fracture strength of perfect single-layer graphene (according to Table 1), the improvement in strength can theoretically be expected from the rule of mixtures. However, the actual increment in strength is usually much lower than the theoretical strength. This can be attributed to five main reasons. The first is that most of the graphene sheets used in AMCs are few-layer graphene and their fracture strength is much less than that of the single-layer graphene sheets. Second, the out-of-plane strength (i.e. weak physical bonding between atoms of adjacent layers) of graphene sheets is much less than the in-plane strength (i.e. strong chemical bonding between adjacent atoms in the same layer), thus those sheets not aligned along the loading direction cannot reach their full potential. The third is the existence of pores and cavities which were left after composite fabrication (especially PM and casting routes), therefore these pores are responsible for crack initiation during loading. The fourth possible reason can be related to the interfacial reaction and bonding of the Al and graphene composite. Finally, the distribution of graphene sheets within the matrix is not uniform and the graphene agglomerations significantly promote the premature fracture.

### 3.4. Ductility

The introduction of reinforcement particles into metal matrices improves the hardness and strength of metals, albeit at the cost of ductility. The tradeoff dilemma between ductility and strength has been extensively identified in AMCs reinforced with ceramic particles. The prominent advantage of Al/graphene composites over the conventional AMCs, is their simultaneous high strength and superior ductility. Figure 26a presents the tensile stress-strain curves of Al-3.9Cu–1.5Mg (wt%) alloy matrix composites.
reinforced with 0, 0.15, and 0.5 wt% GNFs. It can be observed that not only GNFs show an excellent reinforcing effect on the mechanical strength of the Al alloy, but also the elongation to failure of Al/GNF composites is not decreased with the addition of reinforcement. Interestingly, the elongation to failure of the Al alloy reinforced with 0.15 wt% GNFs is exceeding that of the unreinforced Al alloy (Figure 26b). The retention and even improvement of the ductility of the Al matrix in Al/graphene composites are mainly attributed to the multiply wrinkled structure of graphene and a good combination of graphene sheets with the Al matrix.59 During plastic deformation, graphene sheets with a multiply and highly wrinkled structure are straightened and flattened, which in turn leads to the improvement or maintaining ductility.

Similar behavior has been reported for Al-0.5 wt% GNF composites.33 As shown in Figure 26c and 26d, both the elongation and area contraction of pure Al are maintained in the composite with 0.5 wt% GNFs. Nevertheless, by increasing the graphene content from 0.5 to 1 wt%, the tensile elongation of the sample is significantly decreased, from 19.9% of Al-0.5 wt% GNF to 8.3% in Al/GNF with 1 wt% GNFs. TEM images of the composite samples reveal graphene agglomerations with approximately 50 nm thickness situated along the grain boundaries of Al. The graphene agglomeration has resulted in a weak interfacial bonding between the matrix and the reinforcement, leading to degraded ductility. Nevertheless, unlike ductility, the strength of the composite material shows no deterioration because of increased graphene content from 0.5 to 1 wt% (Figure 26c). These findings are in agreement with other reports in the literature.30,62,65,83,87 Furthermore, increasing graphene content beyond 1 wt% leads to a simultaneous decline in both strength and ductility of the sample, as shown in Figure 26d. This phenomenon is ascribed to the difficult dispersion of graphene content within the matrix at higher quantities, rendering their homogenous dispersion almost impossible.158

Figure 27a shows a crack on the fracture surface of the bulk Al6061/graphene composite which is formed.
due to the poor interface between the large graphene cluster and the Al alloy matrix. Under the loading, these cracks propagate and grow, resulting in the inferior mechanical behavior of the composite sample.\textsuperscript{26} Therefore, poor distribution of graphene results in an increased extent of agglomeration, as well as degraded strength and ductility. Conversely, homogeneously dispersed graphene can act as bridges resisting against crack propagation in the matrix,\textsuperscript{126} as shown in Figure 27b.

4. Physical properties of Al/graphene composites

In addition to its remarkable mechanical properties, graphene also possesses outstanding physical properties, such as high thermal conductivity,\textsuperscript{159} negligible thermal expansion\textsuperscript{160–165} and excellent electrical conductivity.\textsuperscript{166} Consequently, graphene-reinforced AMCs tend to exhibit desirable thermal and electrical conductivity, in addition to their enhanced mechanical properties as a result of hybridizing Al with graphene. Unlike the majority of the research already undertaken in this field which analyze and investigate the changes incurred to mechanical properties of Al/graphene, only a limited number of scientific papers have set out to study the effect of graphene addition on the physical properties of AMCs. The most common traits to be investigated for this purpose are thermal conductivity, coefficient of thermal expansion (CTE) and electrical conductivity. Table 9 presents an overview of the physical property changes reported in the literature. Throughout the following sections, these changes will be investigated further while the underlying mechanisms responsible for this particular behavior will be brought into light.

4.1. Thermal properties

The dissipation of thermal energy produced by electronic devices and systems is of great importance in modern electronics which can robustly influence their...
performance, reliability, and lifetime. On the other hand, appropriate thermal properties are essential for the structural parts operating at elevated temperatures in order to prevent the heat accumulation and thereby the descent in their structural integrity. It should be noted that most polymers and ceramics are insulators or have negligible thermal conductivity. Ultrahigh thermal conductivity makes graphene the most promising filler for thermal conductive polymer and ceramic matrix composites. On the other hand, metals like Al possess high thermal conductivity and CTE. Although the addition of ceramic particles usually reduces the CTE, but these reinforcements concurrently reduce the thermal conductivity of MMCs.

Graphene, with a combination of an exceptionally high intrinsic thermal conductivity (over 5000 Wm\(^{-1}\) K\(^{-1}\)) and a low CTE (\(-6 \times 10^{-6}\) K\(^{-1}\)), is an excellent candidate to be utilized as the filler in MMCs. Thus, hybridizing of Al with graphene can enhance the thermal properties of AMC parts.

Figure 28 displays the thermal conductivity and specific heat capacity of Al/rGO composites at 20°C (Reprinted with permission).89

![Figure 28. Thermal conductivity and specific heat capacity of Al/rGO composites at 20°C (Reprinted with permission).](image)

Table 9. Reported physical properties of graphene-reinforced AMCs and their change in comparison to the unreinforced matrix.

<table>
<thead>
<tr>
<th>Year</th>
<th>Composite material</th>
<th>Thermal properties</th>
<th>Electrical properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>2014</td>
<td>Al-rGO</td>
<td>Increased from 150 to 172 (±14.6%) at 250°C</td>
<td>–</td>
</tr>
<tr>
<td>2015</td>
<td>Al-0.1, 0.25, 0.5 wt% rGO</td>
<td>Increased from 48.5 to 58.5 (±20.6%) at 100°C</td>
<td>–</td>
</tr>
<tr>
<td>2016</td>
<td>AI024-0.3, 0.5 vol% FLG</td>
<td>Decreased from 22.9 \times 10^{-6} to 19.9 \times 10^{-6} (−13.1%) at 25°C</td>
<td>–</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5 wt% graphene</td>
<td>–</td>
<td>Increased from 33.8 to 36.8 (±8.9%) at 200°C</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.2 wt% rGO</td>
<td>–</td>
<td>Decreased from 35 to 30 (−14.3%) at 20°C</td>
</tr>
<tr>
<td>2017</td>
<td>Al2219-0.1, 0.5, 2 wt% graphene</td>
<td>–</td>
<td>Decreased from 29 to 21 (−27.6%)</td>
</tr>
<tr>
<td>2017</td>
<td>Al-0.5, 1 wt% GNP</td>
<td>Increased from 188 to 192 (+2.1%) at 150°C</td>
<td>–</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.2 wt% GNP</td>
<td>–</td>
<td>Decreased from 36.3 to 35.8 (−1.1%)</td>
</tr>
<tr>
<td>2018</td>
<td>Al-0.1, 0.2, 0.3 wt% rGO</td>
<td>Increased from 143 to 165 (+15%) at 20°C</td>
<td>–</td>
</tr>
</tbody>
</table>

*Optimal sample. **Thermal contact conductivity.
techniques. As shown in Figure 29, the thermal conductivity of all samples is decreased by increasing the temperature. Moreover, a slight increase in the thermal conductivity is witnessed for the Al-0.5 wt% GNP composite, while the composite sample reinforced with 1 wt% of GNPs suffers a noticeable drop in the thermal conductivity. By comparing the results achieved by the two production methods, it is revealed that press-and-sinter method yields higher thermal conductivity than hot rolling, albeit slightly. The reason for the lower thermal conductivity of the hot rolled samples compared to the ones fabricated through press-and-sinter is stated to be the higher level of porosity present within the final composite component. Moreover, owing to the likeliness of formation of graphene agglomerates at higher graphene content, the Al-1 wt% GNP sample fabricated via either route exhibits even lower thermal conductivity.

In a research conducted by Shin et al., the impact of graphene or CNT addition on the CTE of AMCs was studied. Figure 30 represents the CTE values for Al2024 hybridized with MWCNTs and FLG. As can be observed, the CTE value exhibits an increasing trend with the temperature, however, in the case of AMCs reinforced with graphene and MWCNTs, this value is lower than that of the unreinforced alloy. Furthermore, CTE is lowered with increasing the MWCNTs or FLG content, which is ascribed to the reinforcement constraint on the expansion behavior of the matrix. It is stated that the length-to-diameter (l/d) ratio of reinforcement can also impact the CTE of the composite so that particles with higher l/d ratios are more effective in decreasing the CTE. Consequently, the composite sample reinforced with MWCNTs which has a significantly greater l/d ratio and a larger volume fraction has proven more effective in hampering the CTE of the final composite in comparison to the sample reinforced with FLG, while

Figure 29. Thermal conductivity of the Al/GNP composites manufactured using (a, b) press-and-sinter and (c, d) hot rolling (Reprinted with permission).

Figure 30. Dependence of CTE values of Al2024/MWCNT and Al2024/FLG composites with temperature (Reprinted with permission).
both samples exhibit significantly lower CTE in comparison to commercial and heat-treated Al2024 alloys.

4.2. Electrical properties

The discovery of graphene with extraordinary electrical properties such as high electronic conductivity and electron mobility has released an unbelievable revolution in the electronic device industry.\(^{172}\) The addition of graphene into ceramic and polymer matrices with the intention of enhancing their intrinsic poor electrical conductivity without compromising other properties has been deeply explored.\(^{15}\) Nevertheless, there are a number of studies in the literature which have investigated the electrical properties of graphene-reinforced MMCs mainly due to the good electrical conductivity of metals. Al/graphene composites have been proven to be promising alternatives for applications which require high strength in tandem with high conductivity, such as Al wires and cables for power transmission.\(^{108}\) The reported electrical conductivity values for bulk Al/graphene composites are given in Table 9. In a study conducted by Kim et al.,\(^{85}\) the different aspects of matrix-reinforcement interface are investigated, and their impacts upon the electrical properties of rGO-reinforced AMCs produced by a PM route are elucidated. Figure 31 demonstrates the electrical conductivity of hot-rolled samples from non-milled and ball-milled Al powders, as well as Al-0.2 vol% rGO composite powders. It can be seen that the non-milled Al sample exhibits 61% International Annealed Copper Standard (IACS) conductivity which is very close to that of the pure Al (62%).\(^{173}\) However, it is observed that the ball-milled Al sample has suffered a rather significant drop (by 11%) in electrical conductivity compared to the non-milled sample, down to 51% IACS. It should be noted that the resistivity of an Al matrix can be partitioned into the contribution of four scattering mechanisms including phonon scattering, dislocation scattering, interface scattering and impurity scattering.\(^{174}\) Thus, the reduction in electrical conductivity is related to the possible increase of grain boundaries and dislocations in the wake of the milling process, which acts as electron scattering sites, effectively hindering conductivity. Furthermore, the electrical conductivity remains almost unchanged for the rGO-reinforced AMCs in comparison to their monolithic counterpart, i.e. the ball-milled sample, despite the different reduction atmospheres. In contrast to graphene, rGO is known to possess a considerably lower electrical conductivity than Al, resulting in rGO acting as an impurity and yet another potential electron scattering site when it comes to electron transfer. A similar reduction in the electrical conductivity of AMCs was also observed by Kumar Pillari et al.\(^{64}\) with the addition of 0.5–2 wt% graphene into Al2219, and Li et al.\(^{108}\) with the addition of 0.2 wt% GNP into the pure Al. Consequently, owing to the limited conductivity of the filler in addition to its minute amount, rGO is considered to have a negligible impact upon the electrical conductivity of the final composite sample. However, the impressive enhancement of mechanical properties as a result of the addition of graphene to the Al matrix should also be taken into account, despite its minor effect on the electrical conductivity.

5. Challenges and obstacles related to Al/graphene composites

As was mentioned in the previous sections, graphene has proven to be remarkably potent in improving the mechanical and physical properties of AMCs in comparison to the base metal or its common alloys. However, progress and development of Al/graphene composites have been hindered due to various challenges and difficulties underlying the nature of the involved phases. Among these challenges, inhomogeneous dispersion of graphene within the Al matrix and weak Al/graphene interfacial bonding, formation of carbides, structural damage during the fabrication process, and decreased ductility of the fabricated composite stand out. In the following sections, the aforementioned obstacles shall be individually discussed, and potential solutions or method of overcoming these hurdles will be elucidated.

5.1. Inhomogeneous dispersion and weak interfacial bonding

One of the major obstacles in preparation of AMCs reinforced with graphene is ensuring proper and
homogeneous dispersion of graphene sheets within the matrix. The sizeable density mismatch of the Al matrix and graphene reinforcement often results in poor dispersion of graphene in the Al matrix, which not only lowers graphene’s reinforcing efficiency drastically, but also leads to stacking of graphene sheets and subsequent slipping under applied pressure.\textsuperscript{138} The strong van der Waals interaction between graphene sheets also results in the agglomeration and formation of a separate graphene phase, which often proves detrimental for the properties of the composite. Furthermore, due to the poor wettability of graphene sheets by Al matrix, the bonds formed between the phases are weak which hinder the mechanical properties of the composite even further.

A great deal of effort has been placed on ensuring the uniform dispersion of graphene within the metallic matrix and enhancing metal/graphene interfacial bonding.\textsuperscript{77,79,176} Various techniques such as high-energy ball milling (for mechanical exfoliation of graphene, or better dispersion of graphene within the matrix),\textsuperscript{177} CVD (layered deposition of graphene and metal),\textsuperscript{178,179} flake powder metallurgy (using flake metal powders),\textsuperscript{58,59,77,180} and decorating of graphene with metallic nanoparticles\textsuperscript{62,137,157} (for improving the wettability and interfacial bonding) have proven successful in countering the inherent challenges of producing graphene-reinforced MMCs, namely the agglomeration and poor dispersion of graphene. For instance, studies reveal that the incorporation of graphene using novel PM methods such as flake powder metallurgy and molecular-level mixing significantly improve the resulting mechanical properties via enhancing the distribution of graphene sheets within the matrix,\textsuperscript{58,181,182} whereas traditional PM processing has resulted in much lower properties overall.\textsuperscript{31} Another possible solution for week bonding and dispersion of graphene in the matrix that has recently been noticed is the incorporation of metallic nanoparticles (MNPs) on the graphene sheets. Research has been carried out on composites reinforced with graphene which in turn were decorated with various MNPs such as Ni, Ag, Au, Pd, and Pt.\textsuperscript{138,183,184} The MNPs act as effective spacer agents which prevent restacking and aggregating of graphene sheets, improving the state of dispersion in the composite structure. Moreover, the MNPs contribute to strengthening the matrix-reinforcement interface via the formation of chain molecules and facilitating the formation of a solid solution within the composite.\textsuperscript{137} This results in a stronger interface and bonding between the phases, in addition to the improved dispersion of graphene within the matrix.

### 5.2. Formation of carbides

The formation of Al carbide, Al\textsubscript{4}C\textsubscript{3}, is known to occur due to the reaction between the Al matrix and carbonaceous nanomaterials including graphene during the composite production or the subsequent processing at high temperatures by the following reaction:

\[
4\text{Al}(s) + 3\text{C}(s) \rightarrow \text{Al}_{4}\text{C}_3
\]

This reaction is thermodynamically favorable with below zero Gibbs free energy. Al\textsubscript{4}C\textsubscript{3} phase usually grows on the high surface free energy prismatic planes of carbon and its formation is promoted at defects in the graphitic planes.\textsuperscript{185} The formation of Al\textsubscript{4}C\textsubscript{3} at the graphene/Al interface has been frequently reported.\textsuperscript{28,31,60,65,68,127} An example of the Al\textsubscript{4}C\textsubscript{3} phase created during the fabrication of Al/GNS composite via PM route is elucidated in Figure 11.

Since the interfacial reaction for the formation of Al\textsubscript{4}C\textsubscript{3} is extremely under the influence of reaction kinetics and Al diffusion,\textsuperscript{68} it can be anticipated that this phase is mostly formed when Al/graphene composites are processed at high temperatures for a prolonged time (e.g. via press-and-sinter, pressure-assisted sintering, and casting methods). Thus, employing low temperature and short-time consolidation processes like SPS can be beneficial in removing or at least hindering the formation of a thick layer of Al\textsubscript{4}C\textsubscript{3}.\textsuperscript{83} Nevertheless, the formation of Al\textsubscript{4}C\textsubscript{3} phase during the low-temperature processes like HPT has been reported as well.\textsuperscript{60}

It is known that the interfacial Al\textsubscript{4}C\textsubscript{3} has detrimental effects on the final mechanical properties of AMCs reinforced with carbonaceous materials due to its brittle nature. Bartolucci et al.\textsuperscript{31} reported that AMC reinforced with 0.1 wt% graphene platelets produced by a PM route consisting of ball milling, HIP and hot extrusion, exhibited decreased strength, hardness, and ductility due to the formation of Al\textsubscript{4}C\textsubscript{3}. The promotion of Al\textsubscript{4}C\textsubscript{3} creation was related to the defective nature of graphene platelets produced by thermal exfoliation/reduction of graphite oxide. Therefore, it may be difficult to produce Al/graphene composites with good mechanical properties unless careful attention is given to the processing temperatures and times in order to avoid the formation of Al\textsubscript{4}C\textsubscript{3} phase. Nonetheless, the formation of a thin carbide layer has been reported to be helpful in improving mechanical behavior, by improving the Al/C interfacial bonding.\textsuperscript{186} Consequently, the amount of produced...
interfacial Al₄C₃ phase effectively changes the final mechanical properties of composites.

5.3. Structural damage during fabrication process

In addition to the points made above, there is also the issue of structural damage incurred upon the composites throughout the fabrication processes which can negatively affect the properties of the final product. These damages are created almost in all the methods of Al/graphene production techniques, albeit not to the same extent. The most common instances of these damages occur mostly during the ball milling process and can be categorized into three distinct categories. First, is the contamination of composite powders. This problem is witnessed due to the abrasion of milling balls following repeated impacts between the balls (commonly steel milling balls) and the powder, which results in their gradual abrasion and inclusion of contaminations within the AMC powders. These inclusions also tend to cause further complications at the compaction stage. In order to solve this issue, ceramic milling apparatus, such as tungsten carbide, can be utilized during the ball milling process which greatly diminishes the presence of unwanted compounds and phases within the composite powder. Second, is the creation of defects in the structure of graphene. The repeated deformation, micro-welding, and fracture mechanisms occurring during the ball milling process of AMC powders can result in the accumulation of some damage to the surfaces and edges of the graphene sheets. High-energy ball milling or prolonged milling time, directly increases the number of impacts between the milling balls and the powder, leading to increased defects within the structure of graphene, therefore degrading the eventual mechanical properties. Third, is the deterioration of various mechanical properties, namely ductility. The repeated impacts between the powder and milling balls serve to break agglomerations within the composite powder, however, in cases where the incoming energy to the composite powders is too high (either due to long milling duration or significantly high rotation speed), residual stresses starts to build within the powders, giving rise to amorphous structures with low ductility. Although this phenomenon simultaneously leads to elevated hardness, it might not be a desirable evolution based on the application of the final composite product due to decreased ductility. Consequently, balancing the milling parameters in order to achieve optimized mechanical properties remains an important challenge for the fabrication of AMC's via PM route.

5.4. Decreased ductility

The impact of graphene addition on the mechanical properties of AMCs has been discussed in great detail in Section 3. Furthermore, it was stated in Section 3.4 that AMCs containing a maximum graphene content of approximately 0.5 wt% exhibited highly enhanced strength and ductility simultaneously. From the perspective of ductility, the enhance ductility was mainly attributed to the multi-layer and wrinkled structure of graphene sheets that tend to straighten and flatten during plastic deformation. However, increasing the graphene content from 0.5 to 1 wt% results in a significant loss of the ductility of AMCs, whereas the strength of composite materials usually continues to increase in proportion along with graphene content. This is due to the formation of agglomerations throughout the composite structure, leading to the deterioration of ductility. This phenomenon essentially means that the mechanical properties of the final composite product, namely strength and ductility, will need to be balanced according to its desired application via adjusting the graphene content within the matrix. While elevated levels of composite strength can be achieved by incorporating higher amounts of graphene in the composite, it will result in noticeable ductility loss. However, the decrease in ductility might not be of any major concerns in applications where strength is the primary requirement for performance.

It should be noted that above a threshold in the graphene content (~1 wt%), both strength and ductility begin to deteriorate due to the formation of extensive agglomeration of graphene in the Al matrix. Thus, fine-tuning the exact amount of graphene content in AMCs in order to optimize the mechanical properties according to future application is yet another challenge that needs to be considered prior to the manufacturing process.

6. Potential applications

Despite their outstanding properties, graphene-reinforced AMCs still remain at a research stage. Although no commercial applications or products based on these composites have been made available as of yet, there are several fields which might benefit from potential applications of these composites. In this section, a brief description of these applications, as well as the desired
properties which enable the usage on Al/graphene composites, will be presented.

Due to their high strength and wear resistance, along with their reasonably low density, graphene-reinforced AMCs can be used in the automotive industry for making piston rings, brake shoes and various gears. Furthermore, owing to their low coefficient of thermal expansion and high thermal conductivity, these composites can be utilized in piston combustion face for lowering engine emissions at elevated temperatures. In aerospace industries, these composites can be used in brakes and landing gears thanks to their outstanding wear resistance, good thermal conductivity, high strength, and low density. Additionally, mainly because of their good electrical conductivity, they can be utilized in making of antennas. The outstanding elastic modulus of Al/graphene-composites combined with their enhanced strength makes them ideal candidates to be used for manufacturing lightweight bicycles and tennis rackets when it comes to the sports industry. In the end, their large surface area, high current density, and enhanced hydrogen adsorption-desorption rates enable their potential use for energy storage purposes, where these composites can be employed in anodes and coatings, as well as hydrogen storage materials.

7. Summary and future outlook

Desirable properties of Al, in addition to the outstanding characteristics of graphene, have attracted a great deal of attention towards the development of AMCs reinforced with graphene. Along with the enormous success in the experiments carried out in recent years, there has been ever-increasing interest in the graphene-reinforced composites as viable alternatives for the mainstream materials. Although graphene is usually referred to as the successor of CNTs, it is in fact the fundamental structural unit which makes up the CNTs, and usually proves to be more effective than CNTs when it comes to mechanical behavior.

Synthesis of graphene is among the most crucial challenges in this regard. Several methods such as exfoliation or CVD have come into recognition as feasible methods of achieving graphene, which can later be used in various applications and industries. Oxidation of graphite, exfoliation, and further reduction of GO into rGO has been utilized in numerous studies to great effect as well. Regardless of the method of synthesis, the aim is to include as few flaws and defects within the structure of graphene as possible, since these flaws often contain great detrimental effects on the final physical and mechanical properties.

The incorporation of graphene into the Al matrix significantly improves the strength and hardness of the composite material like other reinforcements in MMCs. However, the outstanding advantage of graphene over the other reinforcements is the retention and possibly even the improvement of ductility of the Al matrix. In addition to the aforementioned improvements in mechanical properties, the addition of graphene also results in a significant boost to physical properties of the final composite product, such as enhanced electrical and thermal conductivity, as well as reduced coefficient of thermal expansion. Yet as was greatly discussed in the previous sections, the final properties of AMCs will largely depend upon the distribution of graphene sheets within the matrix, where more homogeneous distributions will result in a better overall outcome. The powder metallurgy is the main technique for production of Al/graphene composites due to its higher capability for the distribution of graphene within the Al matrix. Several techniques have been suggested to promote the uniform dispersion of reinforcement within the matrix, and to prevent the formation of agglomerates which greatly deteriorate mechanical and physical properties. Ball milling, electrostatic adsorption, and decoration of graphene sheets with metallic nanoparticles are among these proposed methods. It should be noted that the production of Al/graphene composites with appropriate mechanical properties is only possible when careful attention is given to the processing parameters in order to achieve strong bonding between graphene and Al and avoid the detrimental interfacial reactions.

Another important factor is to optimize the amount of reinforcement phase within the matrix. It was observed that past a certain threshold in graphene content, the properties start to rapidly degrade, namely ductility. Determining this threshold and attempting to strike a near-perfect balance between strength gains while combatting the ductility loss will be among the greatest challenges of this particular field of science.

In addition to the points mentioned above, several gaps in knowledge still remain ahead. These areas of potential interest for future research mainly include the following:

- While the majority of investigations use different graphene-based materials, there is often little to no control over their characteristics. Thickness, lateral size, crystallinity, impurity content of graphene, etc. still need to be elucidated further to increase
the mechanical and physical gains obtained from graphene in AMCs.

- Although theoretical predictions based on a single-layer graphene phase distributed homogeneously in perfect alignment indicate exceptional physical and mechanical properties, the results achieved in practice have thus been far from ideal. This noticeable drop in properties is often due to practical considerations such as the formation of agglomerations, misalignment of graphene particles within the matrix and folding of graphene sheets, as well as the usual defects present in both the matrix and reinforcement phases. Consequently, there still remains a great deal of untapped potential for improvement of the various properties of AMCs that can be achieved after the aforementioned issues have been addressed.

- The synergistic effect between graphene and other reinforcement phases such as CNTs or ceramic particles like Al₂O₃ are in need of further exploration to fully determine their bonding and distribution within the Al matrix and thus the obtained mechanical and physical properties.

- Some derivatives of graphene, namely rGO, while leading to a remarkable boost to mechanical properties, possess certain undesirable physical properties (e.g. low electrical conductivity) than Al itself. Striking a balance between mechanical and physical properties of a composite based on the desired application needs to be investigated and experimented further.

- Process parameters of the existing synthesis methods can still be improved and optimized. Furthermore, computational studies such as density functional theory (DFT) and molecular dynamics (MD) at a nanometer scale can be useful means of studying fundamental interactions at phase interfaces, the impact of graphene defects on final properties of the composite, as well as comparison between various properties of single layer versus multi-layer graphene sheets.

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**References**


57. L. Zhang, J. Liang, Y. Huang, Y. Ma, Y. Wang, and Y. Chen, Size-controlled synthesis of graphene oxide sheets on a large scale using chemical exfoliation, Carbon 47, 3365 (2009).


121. A. Gholiniia, F. Humphreys, and P. Prangnell, Production of ultra-fine grain microstructures in Al–Mg alloys by conventional rolling, Acta Mater. 50, 4461 (2002).
133. H. Asgharzadeh, S.-H. Joo, and H. S. Kim, Consolidation of carbon nanotube reinforced


