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A Review On Recent Advances Of Aluminium/Graphene Nano Composite

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ABSTRACT

Graphene is a very suitable filler material for the production of composite material due to its outstanding physical, chemical, electrical, and mechanical properties. It is Sp^2 hybridized with single atom thick and having hexagonal crystal lattice. The most attractive property of graphene includes a large specific surface area and lightweight. Among the various metals, aluminium has large areas of application. It has been observed that graphene reinforced aluminium composite with lightweight with excellent properties has brought the focus of scientists. The attractive force between aluminum and carbon is more than the Vander wall force between carbon and carbon layer. This property helps to form a stable composite of carbon with aluminum. The high aspect ratio and fluidity property of graphene make it suitable filler materials in aluminum matrices to form a coating layer over aluminum. In this chapter systematic effect has been taken to explore different synthesis routes that have been used for successfully fabricating Al/graphene composite by controlling the dispersion of graphene in the matrix. Especially it is observed that the powder metallurgy technique is a relatively more efficient technique for developing good quality composite of Al/graphene by enhancing properties of composites. Different macroscopic, microscopic, spectroscopic, and other analytical instruments like four-probe electrometers and nanoindentation are used to investigate the properties of the developed materials.

1. Introduction

Graphene is a monoatomic layer of graphite. It is an allotrope of the carbon family. When SP^3 hybridized graphite is converted to SP^2 hybridized C-C bonded with a pie bond electron cloud, it forms graphene. It is a two dimensional material of one atom thickness. It has drawn the attention in the field of research, academic & industries due to its unique properties such as high mechanical strength, current density, high electron mobility, ballistic transport, chemical inertness, high thermal conductivity, electrical conductivity, biosensor, energy field, display field superhydrophobicity. Some typical properties of graphene are listed in Table 1 [1-31]. Graphene is categorized into different types based on its structure and number of layers. Based on association and different layers they are classified into different categories.

Different types of Graphene's as per the layers and structure:

As per the layers

- Single-layer Graphene
- Very few layers Graphene (VFLG): 1-3 graphite layers
- Few layer Graphene (FLG): 2-5 graphite layers
- Multilayer Graphene (MLG): 5-10 graphite layers
- Graphene Nanoplatelets: 11 or more graphite layers

As per association and structure

- Functionalized Graphene
- Graphene Quantum Dots
- Graphene Oxides (Go)
- Reduced Graphene Oxide (rGo)
- Graphene in a Suspension
- Graphene in a Powder Form
- Graphene in a Paste.
- Graphene in a Master Batch Formulation
- Other

Day by day across the globe the demand for graphene is very highly impacted and presented in Fig. 1 and Fig. 2 by review the literature [32-36]. Directly or indirectly many scientists have been working in the area of graphene. The trend of annual publication rate is found increasing in graphene-based research. This prominently confirms the importance of graphene in different industrial application areas.

Table-1: *Typical Properties of graphene*

Properties	Values
Carrier mobility	200000 $cm^2V^{-1}S^{-1}$
Young's modulus	~ 1-1.5 TPA
Optical transmission	97.3%
The theoretical specific surface area of single-layer graphene	2630 m^2/g
Electrical conductivity	10^8 S/m
Thermal conductivity	4000 watt.$m^{-1}K^{-1}$
Current density	~10^9 Amp/cm^2
Sheet resistance for 5 layers	120 ohm/sq
Transmittance for 5 layers at 530 nm	87.7%
Transmittance for 10 layers at 530 nm	76%
Electron mobility	x100 of Si

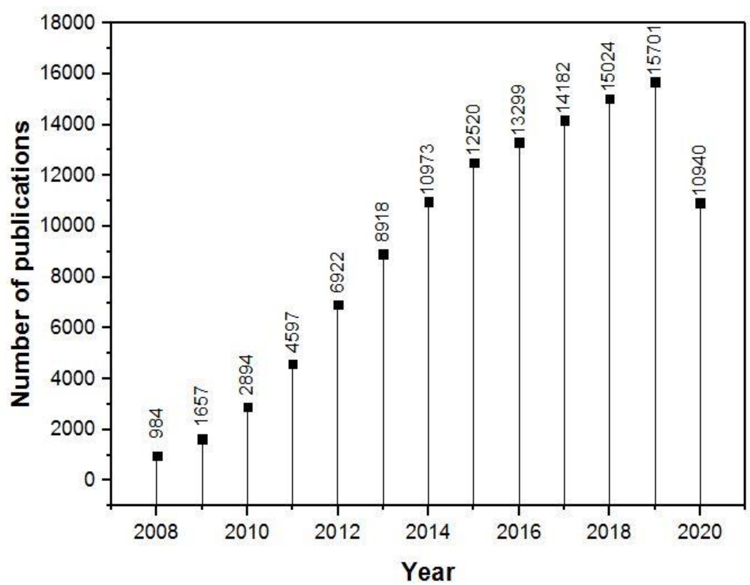

Fig. 1: *Annual number of publications in graphene*

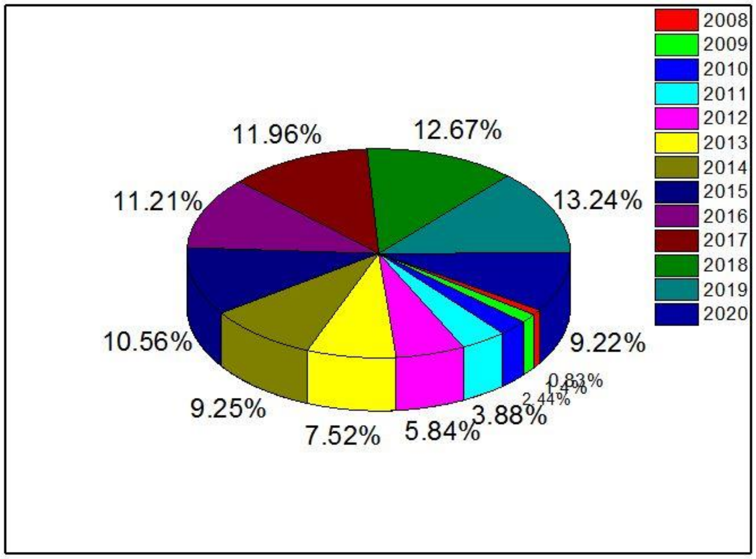

***Fig. 2**: Update publication % rate of graphene*

Aluminum (Al) has a crystal structure of FCC (face-centered cubic). It has an atomic radius of nearly 0.143 nm. It is metallic in nature. This light metal is owned with outstanding physical and mechanical properties such as low density, high corrosion resistance, and good ductility. These outstanding properties of Al make it a super candidate material used in various critical areas in aerospace, construction, automobile, marine, and architectural industries. Because of its poor tribological properties and having low strength, it is rarely used in its original pure form. Production industries have started focusing on the development of alternate kinds of material with making alloy or composite with aluminum. In this way, the engineering material composite of Al comes into the first. Composite consists of two or more phases. One of these phases is called reinforcement and the other one is called matrix. Fibers, particulates, or whiskers belong to reinforcements, and metals, plastics, or ceramics belong to the matrix. Aluminum has been drawn special attention as a metal matrix especially for the last two decades in this regard [37]. Normally various required additives are added to it to improve its properties. Mostly Al is used in alloy or composite form. Various ceramic additives such as Al_2O_3, SiC, BN, and B_4C are usually added to improve its mechanical properties [38]. But in recent times an emerging type of materials such as carbon nanotubes (CNTs), fullerene, and graphene is in focus for improving the properties of aluminium.

Recently, focus on graphene-decorated metal matrix composites has been given by researchers. Different advanced techniques have been developed in the last few years for developing novel aluminium-graphene composite including powder metallurgy, melting and solidification, electrochemical deposition, and some novel deposition techniques. Different typical methods used for composite synthesis have different effects over dispersion and homogeneity of graphene in aluminium matrix. The great challenge involved in the fabrication of Al/graphene-composite is how to achieve a uniform distribution of graphene in the Al matrix. Though graphene contributes excellent electrical, thermal, and mechanical properties, all these contributions of graphene can be achieved in case of formation and presence of single-layer graphene in the composite. Different outstanding properties, process parameters, and influencing factors on Al/graphene composite are also focused in this chapter. Among the different synthesis techniques, powder metallurgy has been found as an efficient one for exploring most of the outstanding properties of Al/graphene composite.

2. Synthesis of Al/graphene composite by powder metallurgy route

Powder metallurgy (PM) synthesis route is a very important technique for producing majority Al/graphene composites. Various metallic, nonmetallic, and composite components can be synthesized by this technique. PM technique can be applied for preparing composite of Al with graphene by following two main steps: (i) Aluminium and graphene composition mixing by grinding or mechanical alloying (ii) mixed composition consolidated by following different techniques such as sintering, spark plasma sintering, cold isostatic pressing and hot isostatic pressing.

2.1 Mechanical ball milling

The mixing process has a very important role in achieving the homogeneous mixture without forming agglomerates of the graphene and aluminium. Ball milling and dispersion of composition mixture through stirring or sonication are generally used by researchers for developing homogenous mixing between Al and graphene. Among other techniques, ball milling has a highly used technique for developing composites with good quality. It is commonly called “mechanical milling” or “mechanical alloying”. It is a very popular technique for metals. Ball milling is generally carried out by different devices including “shaker mills for small quantities of powder, high-energy, planetary and attritor mills for larger amounts of powder with good quality and properties” [39-40]. Composition of the mixture, their percentage, milling time, milling media, and quality of balls and jars are mainly are the basic parameters to decide the quality of composite. Balls and Jars are made up of stainless steel, tungsten carbide, and zirconia, etc. As aluminium is metal, so researchers use an inert atmosphere like Ar during milling for avoiding any chance of oxidation in the composite. Ball milling is carried out under dry or liquid media. The milling process is often carried out in the presence of chemical agents such as stearic acid, methanol, acetone, and ethanol. These control agents avoid agglomeration. The energy generated in ball milling is sufficient for eliminating agglomeration and bonding between graphene and Al. It will not allow you to form aluminum carbide. The output energy can be optimized by controlling milling time, rotation speed, and ball-to-charge (powder) weight ratio. The impact forces in the ball mill work against van der Waals forces exist in the bonds. The uniform dispersion of graphene in the composite is attributed to the rotation per minute (RPM) of the milling jar. Ball milling carried out at liquid media offers a greater degree of control on producing Al/graphene composite. Liquid media helps to avoid graphene agglomeration. Maintaining cryogenic temperature, grain growth and nanocrystalline structure of Al/graphene composite can be controlled. Compared to dry milling, some literature claims that the distribution of graphene within the matrix is more uniform in the presence of liquid media. It is also possible that graphite can be exfoliated to form graphene via mechanical milling. The size of graphene layers can come down to the nanometric level once after ball milling over. It is one of the economical techniques and a single-step way to prepare graphene and also it's composite with Al or any metal or ceramics. Dash, et al [19] had exfoliated graphite by milling between 2 and 24 hrs. By following beneficiation and chemical leaching processes low-grade graphite is converted into high pure graphite with a carbon value up to 99 percentages. The ball milling of the high purity graphite samples was carried out between 2 and 24 hrs. Among the ball-milled samples 16 hrs. A ball-milled sample was found to show improved morphology with better exfoliation of grains. The multilayer graphene was obtained in that work. The intensity ratio of the first order-disorder peak (D) of carbon to graphite lattice peak (G) of carbon was found increasing with increasing milling time. The 1st order-disorder peak of carbon (D) arises due to defects and oxygenated functional groups associated with the Bernel plane of carbon.

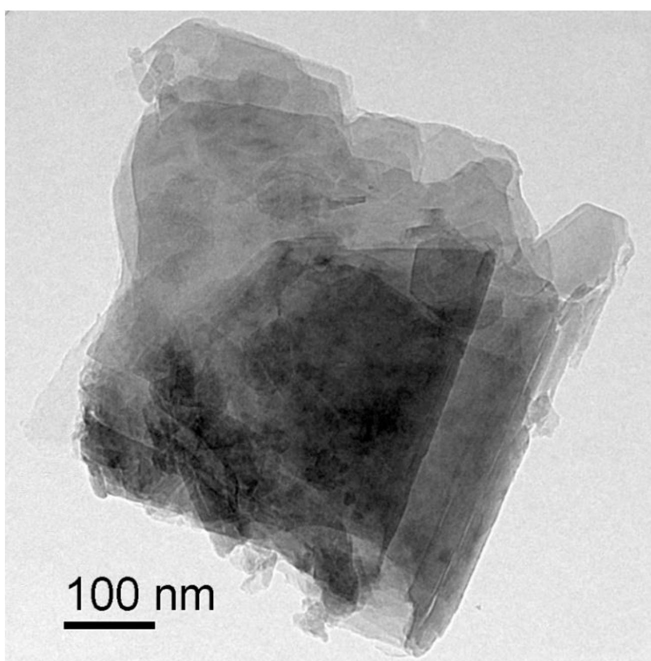

Fig. 3: *TEM image of 8 hr ball milled graphene sample*

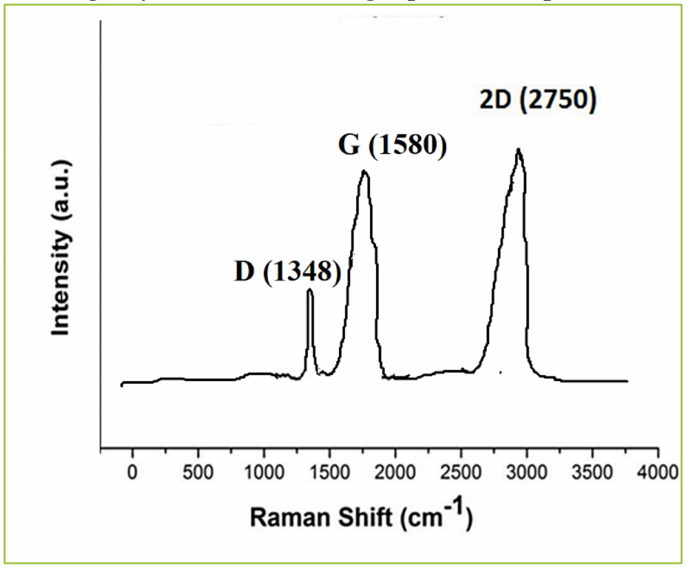

Fig. 4: *Micro Raman result of 8 hr ball-milled graphene sample*

The present authors carried out dry planetary ball milling of high purity graphite under argon atmosphere for 8 hrs. The microstructure of the sample was found to show a clear image of the exfoliated behaviour of graphene (Fig. 3). The micro Raman spectra of the ball-milled sample shown the bi-layer nature of graphene (Fig. 4). Peak broadening in micro Raman analysis is a clear indication of the formation of nanostructured grain size of graphene. As reported in literature here also three peaks are observed: "D (1^{st} order-disorder peak of graphite) at 1348 cm^{-1}, G (peak due to graphite lattice) at 1580 $cm^{-1}$and 2D (2nd order-disorder peak of graphite) at 2750 cm^{-1}".

The dry planetary ball mill technique is also found more efficient for Al/graphene composite to improve the properties of aluminium. Here no chemicals are used which avoids the chance of effect the purity of graphene. The present authors have prepared aluminium/graphene composites by varying graphene % from 0.5 to 1.5% by using planetary ball milling technique at dry condition followed by sintering at 550 ^oC. The ball milling was carried out for 6 hrs. Ball to sample ratio, 9:1 was maintained during ball milling. The sintering time was maintained for 5 hrs. The XRD analysis of samples is presented in Fig. 5. The ball-milled pure Al shows the peak of Al(111), Al(200), and Al(311). But 1.5 wt% added graphene shows an addition peak of C(002) which may be attributed to the presence of graphene. There is no other chemical bonding such as aluminium carbide is not formed. The above good result is attributed to the optimized ball milling condition used in preparing composite. Nayak et al.[29] evaluated microstructure and mechanical properties of Al/graphene (1 wt%) composite prepared by mechanical milling via a self-developed special ball mill. Experimentally samples were prepared by 8 hrs of milling. The samples were taken for sintering at 560 ^{o}C for 6 hrs after ball milling over. The composite formation was confirmed by employing various advanced techniques. From the FESEM analysis (Fig. 6) it is observed that in this work, graphene sheets are irregularly distributed over the sample. No oxidation phase is developed because of the preparation of composite under the Ar atmosphere. Al-graphene with 1 wt% shown a hardness value of 58 VHN. Microstructural and microhardness results indicate that graphene addition improves the structural and mechanical properties of the composite.

It has been seen that by continuous ball milling particles in spherical shape converted into a flaky type of shape. Indeed, the flaky type of shape in the graphene-based composite is most desirable for developing better interfacial bonding which helps in transferring stress from the matrix to the reinforcement. This result may enhance the properties of metal matric composite [41-43]. The milling technique is used for mixing a mixture of metal powders and alloying the metal powder mixture before making a composite with graphene. In literature, a group of researchers reported that via SPEX ball milling successfully aluminum-graphene nanoparticle (0.25, 0.5, and 1 wt%) composites can be prepared [43]. They carried out milling between 1 and 5 hrs. Even in this small duration of milling agglomeration in the composite could not be detected in the SEM study. From Raman spectra, it was observed that the intensity of G, D, and 2D was found to be decreased with an increase of milling time. This gave a clear indication of the increasing defect rate in the composite. But the intensity ratio of D to G was found to be increased with increasing milling time which further supported the claim of increasing defects or disorders with increasing milling time. The decreasing of 2D peaks supports the dislocation developed in the Bernel stacking. But the dispersion of graphene was found to improve with increasing milling time. Hence an optimized condition is always suggested to develop keeping in view balancing dispersion, cold welding, and fracture in the Al/graphene composite. Bastwros et al. [41] studied the microstructure of Al-1 wt% graphene samples after ball milling for 0.5-1.5 hr. It was found that aluminium particle size increase whereas graphene particle size decreased. The intensity ratio of D peak to G peak increased following increment of milling time which indicated more and more defects formed in the composite. But the composite prepared after 5 hrs milling did not follow the trend. Possibly graphene sheets were protected in the cluster of aluminium. But at the same time intensity of 2D peak to G peak (I_{2D}/I_G) was found to increased which indicated more exfoliation and reduction of the number of layers of graphene. 10 min ball-milled was found no effect on graphene dispersion but it lowers mechanical property of graphene. The flexural strength was reported to be increased with increasing milling time from 1 to 1.5 hr.

2.2 Stirring and sonication

Solution-assisted mixing techniques can be processed in two ways (i) stirring or (ii) sonication. Stirring is a phenomenon of mechanical agitation. Sonication is a result of sound applied. At frequencies of sound greater than 20 kHz have been used for developing homogenous mixing of the Al/graphene composites. The applied frequency helps in braking van der Waals interlayer forces that exist in different layers of graphene. This technique consumes very lower energy in comparison to the ball milling technique. As a result of which it induces significantly fewer defects in the Al/graphene composite. But this technique is found to show less effective behaviour on particle size or microstructure of composites. But it has an advantage over contamination in comparison to ball milling techniques. Hence both sonication and ball milling has distinct advantages and complements each other. Nowadays researchers are using both the techniques [27, 29, 44] simultaneously to develop new materials of Al/graphene composites for various advanced applications.

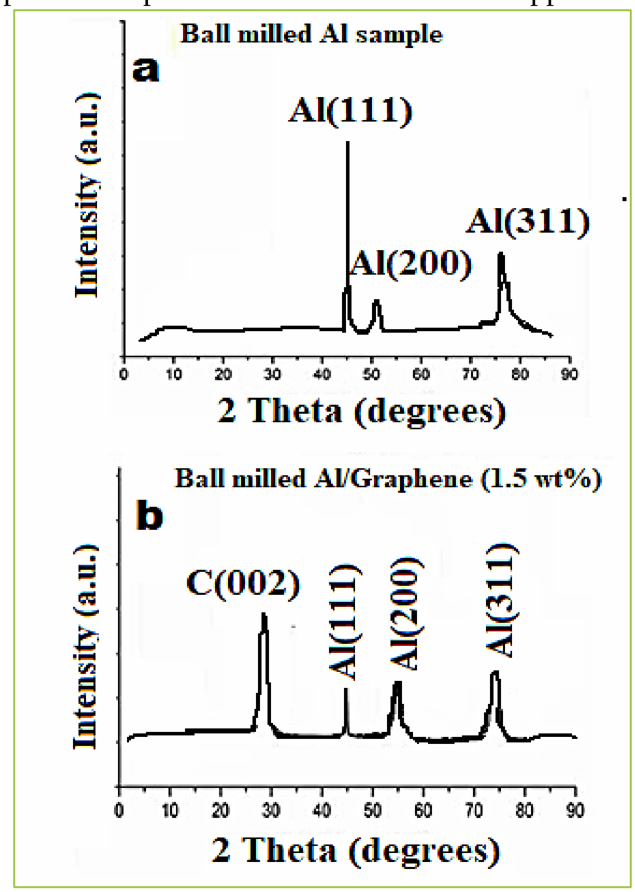

***Fig. 5** XRD spectra: a-aluminum sample; b-aluminum-graphene (1.5 wt%) (8 hrs) composite.*

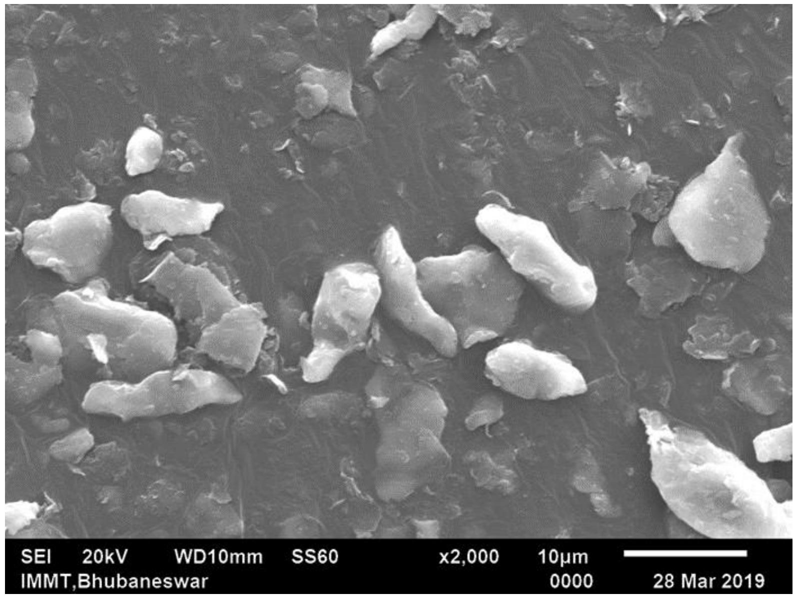

Fig. 6 *FESEM microstructure of aluminium-graphene (1 wt%) [29]*

2.3 Consolidation

After the successful formation of the Al/graphene mixture, compaction is almost essential before going its properties evaluation and application. Compaction can be carried out with or without the presence of applied or external heating. Hence there are two basic kinds of the route for processing composite, (i) "pressureless sintering" and (ii) "pressure-assisted sintering". In pressure less sintering, to develop the mechanism of diffusion, improve the strength of bonds, and improve the mechanical properties, sintering was carried out after compaction. In the "pressure-assisted sintering" route in a single step, both pressure and sintering are carried out which can help in improving properties especially the mechanical property of Al/graphene composites. Some examples of sintering with pressure are "Hot pressing, hot isostatic pressing (HIP), spark plasma sintering (SPS), hot extrusion, hot rolling, and sinter forging". Among the above methods spark plasma sintering (SPS) has been efficiently using as the most prominent one for developing a super composite of Al/graphene for various industrial applications.

2.4 Potential applications of Al/graphene composites

Aluminium is a very popular material having a huge scope of applications. Its potential uses became limited because of its poor mechanical and limited electrical properties. These disadvantages of aluminium are overcome by reinforcing super material graphene successfully in the matrix of aluminium. But still, there is no commercial application of this composite to date. Many fields will be benefited once such a composite will be developed. Because of high strength, low density, and having excellent electrical conductivity, Al/graphene composites can be used in the automotive industry, piston combustion face for lowering engine emissions at elevated temperatures, aerospace industries, transportation materials, preparing defence components, energy carrier material, material for consumer industries, etc. [27, 29].

3. Conclusion

The outstanding properties of graphene promote the field of application of aluminium in the form of composites. In recent years it has been seen that among the metal matrix composite, Al/graphene is present in the front row of the focus of researchers. The graphene which is used in Al should be of high quality which depends on its processing. The reinforcement of graphene in aluminium matrix found significantly improves the physical, thermal, mechanical, and electrical properties of the composite. Inside the chapter, we have discussed the uniform distribution of graphene in aluminium matrix. The composite with more uniformity of graphene has more possibility to exhibit more target outcomes. Different techniques have been developed in recent years to prepare high-quality composite. Irrespective of any techniques, actually process parameters such as the amount of graphene, medium, experimental condition, etc. decide the development of optimizing composite of Al/graphene. As we know the theoretical single layer of graphene distributed uniformly helps to achieve these exceptional physical, mechanical, and electrical properties but this is far from reality at present. Among the different process techniques developed, the powder metallurgy route found cost-effective, less energy-consuming, single step in preparing Al/graphene composite. Different characterization techniques such as X-ray diffraction (XRD), field emission scanning electron microscopy (FESEM), X-ray photoelectron spectroscopy (XPS), energy dispersive spectroscopy (EDS), transmission electron microscope (TEM), high-resolution transmission electron microscope (HRTEM), selected area diffraction pattern (SAED), micro Raman spectroscopy, electrical conductivity, and microhardness are required to properly optimize and validate the product of Al/graphene composites. This review chapter infers that the Al/graphene composite has great scope to impact and full fill the outstanding requirement of the material world to make our life smart and advance.

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