Metal matrix nano composites using graphene nano platelets indented on copper particles in aluminium matrix

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Abstract

Aluminium matrix composite was prepared by using an innovative approach of using graphene indent on copper particles as inforcement material. The reinforcement was mixed and ball milled for 30 and 60 min respectively to get proper sight of copper fracture where graphene (GNP) can be embedded. The reinforcement was also milled for 30 minutes to get uniform distribution of reinforcement in powder state. Casting technique was used with argon gas environment and mechanical stirrer to get final composite material. The morphological analysis has shown proper indent of graphene with Al-Cu and Cu-GNP interface. The composite so formed has micro hardness of 87 HV with an increment of 36.78% whereas yield strength and ultimate tensile strength have increased by 36.67% and 37.162% respectively. Copyright © 2018 VBRI Press.

Keywords: Metal matrix, nanocomposites, graphene nanoplatelets(GNP), aluminium matrix.

Introduction

The production of novel aluminum composites with lower specific density and high mechanical strength and properties is a prime objective of various material science researchers. The need is fulfilled by using various reinforcements such as carbon with various allotropic modifications (CNT, Graphene, Graphite, Carbon fibres etc.), ceramics (Al_2O_3, SiC, B_4C, WC) etc. among which CNT and graphene has achieved greater fame due to its higher Young's modulus of 1TPa, density of 2.3g/cm^3, thermal conductivity of 5000 Wm^{-1}K^{-1}, fracture strength of 125 GPa etc.[**1**]. Graphene, one atomic thickness sp^2 hybridized carbon atoms which are densely packed in honeycomb crystal structure[**2**], has all properties compared to CNT and has higher specific surface area than that of CNT, Therefore, it can replace CNTs in all respect[**3**].

There is improvement in the strength of graphene-based Al and Mg matrix composite when mixed in lower amount, it is basically due to its load transferability. Li and Xiong have investigated the microstructure and tensile properties of GNP/Al composites by using 0.25, 0.5 and 1 wt. % grapheme respectively. There was an increment of 38.27% and 56.19% in yield and ultimate tensile strength when using 0.25 wt% graphene but the value decreased when the percentage of graphene was put beyond 0.5 wt% [**4**]. Wang et al. have shown that in similar experiment having 0.3 wt.% graphene reinforcement composite exhibits 62% enhancement in tensile strength over unreinforced Al matrix[**5**]. Asgharzadeh and Sedigh while evaluating the thermal and mechanical properties on Al2024 matrix composite with 0.5 wt.% few-layer grapheme, have found that the composite has yield stress as two times higher and compressive yield strength as three times higher than that of monolithic Al at a temperature of 350^{0}C[**6**].

The presence of brittle phase aluminum carbide formed during the synthesis of composite material effects its mechanical prosperities. The interfacial phase and interfacial bonding between the GNSs and Al matrix is very important issue for mechanical properties of composites and good interface is always favourable for the transfer of load between GNSs and Al matrix and inhibit movement of dislocations. Zhang et al. have successfully shown that the brittle phase of Al_4C_3 was successfully avoided by transferring Mg on to the surface and, hence, avoiding the direct contact of aluminum with carbon thus excluding the formation of Aluminium carbide[**7**].

A new approach was also utilized to avoid agglomeration and carbide formation by encapsulating SiC nano-particles with graphene sheets during ball milling. The approach thus provides 45% and 84% improvement in yield and tensile strength respectively[**8**].

In this work, we have considered an innovative approach of composite formation by using graphene surrounded by copper and magnesium particles as reinforcement in an aluminum metal matrix in order to avoid the agglomeration tendency of graphene and subsequently eliminating the chances of formation of aluminum carbide. The composite was manufactured by using casting and mechanical stirring method using ball milled reinforcement and matrix materials.

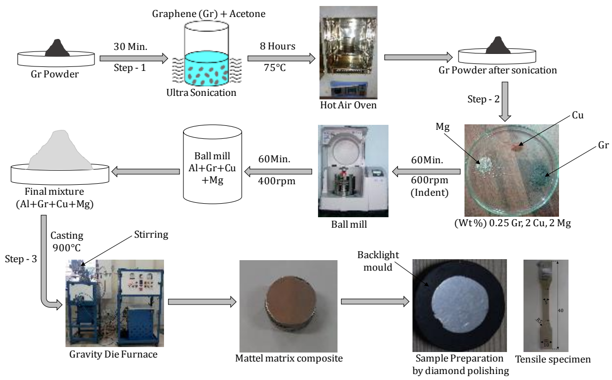

Fig. 1. Experimental procedure.

Material and method

The primary material used are (a) graphene nanoplatelets (GNP) having 2-10nm thickness, supplied from M/s ACS material (b) copper powder (99.5% pure) having mesh size as 200 microns (c) pure aluminum metal. Aluminium metal chip was obtained by turning of the pure aluminum rod and then converting into a fine powder by ball milling it for 3 hrs at 600 RPM with 10 min interval after every 30min of ball mill.

Fig. 1 shows full experimental procedure in which ball milling was performed in Retsch PM 100 planetary ball mill with interruption of over 10 min at every 30 min of milling time. The grinding jar was sealed airtight with an elastomeric O ring. The ball mill was carried out with the ball to mill ratio of 20:1 at 600 RPM for 30 and 60 min milling time respectively incorporating 0.25 wt.% of graphene and 2 wt.% of copper along with 1 wt.% of magnesium as a wetting agent in final aluminum matrix composite. The milling was done to get proper indent of graphene into the voids of fractured copper powder at different time intervals. Subsequently, the powder was collected and again mixed with rest of aluminum powder via ball mill for 30 min at 400 RPM to get uniform matrix reinforcement mixture.

Fig. 2 shows TEM image of graphene powder as taken from M/s ACS Material. The graphene has a thickness of 2-10 nm and specific surface area of 20-40 m^2/g.The initial step was to indent the graphene particles into the copper and magnesium powder for which first graphene was mixed into acetone and ultra-sonicated for 20 min followed by drying it for approx. 8 hours in an oven at 75ºC. This process ensures the separation of graphene layers which have accumulated due to weak Vanderwall force among graphene particles over time.

The composite preparation was carried out by casting process in an argon gas environment followed by compression of semi-solid material to get disc like composite materials. The vicinity for casting was initially cleaned to avoid the contamination of composite while preparation. The furnace specially designed for aluminum composite preparation was first sealed and partially vacuumed. Argon gas was further allowed in the chamber to get inert environment. Initially, the die was pre-heated to over 900ºC and, therefore, mixture was slowly dipped into die from the lid. The mixture was kept for 20 min at the elevated temperature and stirring was carried out for over next 40 min at 400 rpm so that graphene does not settle or agglomerate. Finally, the composite slurry was poured into the preheated die and compressed to get metal alloy.

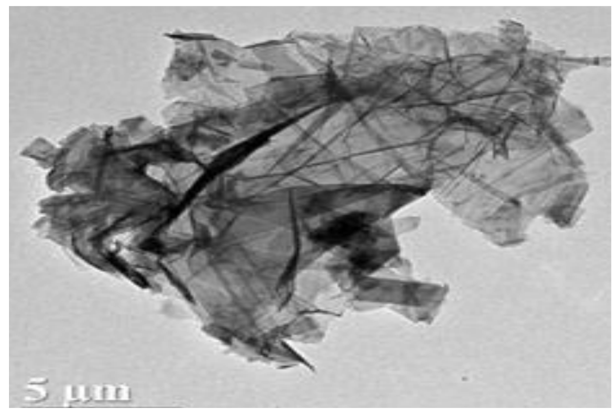

Fig. 2. TEM image of graphene (Courtesy : M/s ACS Material).

The morphological evolution and fracture behaviour of powder after milling were studied using field emission scanning electron microscope (FESEM) performed by ZEISS MERLIN Compact Gemini and electron probe micro analyser (EPMA) performed by CAMECA SX Five was used to characterize the composition and distribution of graphene into prepared composite.

Microhardness of Al/Cu-graphene sample was measured from Economet VH-1 MD Digital Automatic Turret Micro Vickers Hardness Tester. The tensile test was conducted on HOUNSFIELD tensile testing machine.

Result and discussion

Powder characterization

The graphene, copper, and aluminum were characterized using FESEM after ball milling process. **Fig. 3 (a)** and **(b)** show micrographs of mixed graphene and copper powder after ball milling process of 30 and 60 min respectively at 600 rpm. Both images show fracture onto the surface of copper particles creating voids on the surface of the particle.

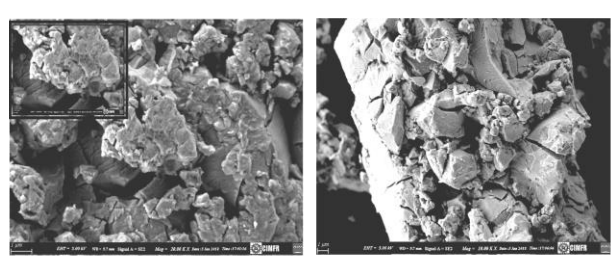

Fig. 3. FESEM image of graphene and Cu powder after ball mill (a) 30 min milling time (b) 60 min milling time.

The deposition of graphene surrounded by these copper particles can be clearly seen by black spots on the fractured surface. The properties of MMCs are highly affected by the characteristics of theinterface between the matrix and reinforcement as if the graphene particles can easily react with the aluminum particles at elevated temperature to produce aluminum carbide which has a brittle phase and easily decorates the properties of the composite[**8**]. These voids thus are filled with the graphene nanoplatelets creating barrier between the direct contact of aluminum and graphene particle.

Fig. 4. Layered Al/GNP/Cu powder.

The **Fig. 3(b)** with 60 min milling time easily demonstrates the larger number of voids than that of 30 min sample demonstrated in **Fig. 3(b)** black spots shows that graphene has strongly settled in the cracks of the surface of the copper powder. The detection of aluminum carbide is a difficult task, however, the result obtained can easily eliminate the chances of formation of these carbides. **Fig. 4** shows the image of 30 min milled 0.25wt.% graphene, 2 wt.% Cu, 1 wt.% Mg, Al powder. The figure illustrates the layered structure of particles in which graphene wassuccessfully embedded in between the copper particles. The powder characterization also shows uniform mixing of graphene particle into the aluminum matrix.

Characterization of Al/Cu-Gr Composite

The uniform distribution of graphene plays a key role in the modification of the properties of the composite. Lower density, weak Vanderwall attraction force and non-wetting characteristics of graphene in the aluminum matrix at a temperature below 1300ºC try to agglomerate the particles when dispersed in liquid metal[**9**], [**10**]. **Fig. 5(a)** shows EPMA characterization image of graphene in the composite material.

The uniform distribution of graphene into the aluminum matrix was observed having Al-Cu and Cu-GNP interface. This interface plays a strong interlink between the grains of composite material enhancing the strength of the composite. Though there was a uniform distribution of graphene in the composite material as confirmed by the test results but some bigger spots indicate the gathering of graphene particle was also seen. The reason of this gathering of graphene platelets was because the graphene which is not able to adhere properly with copper particles or was loosely linked with the copper particles may get separated due to stirring action during the casting process. Thus, showing the brighter image at certain locations. EPMA images have also shown the presence of a copper particles in between the aluminum matrices having dark spaces in it showing the successful presence of graphene between the copper metal matrices.

Microhardness test

Microhardness test was considered at 5 different locations with a gap of 1mm within each run. The microhardnes of formed Al/Cu-Graphene composite was 36.78% higher than that of the aluminum sample. **Fig. 5(c)** shows the diamond shape formed in the composite sample after a particular experimental run whereas, **Fig. 5(d)** shows the indent in the pure aluminum sample. The hardness observed was in the range of 78HV to 95HV. with an average of 87HV at the load of 100 grams. Whereas, microhardness in the pure aluminum sample was found in the range of 52HV to 60HV with an average of 55HV.

Tensile test

Tensile test specimen made out of composite sample having a width of 10mm and length of 40 mm was in accordance with ASTM standard shown in **Fig.1**. The tensile test result shows an increase in yield strength and ultimate tensile strength for the composite sample by 35.67% and 37.162% respectively as compared to unreinforced sample.

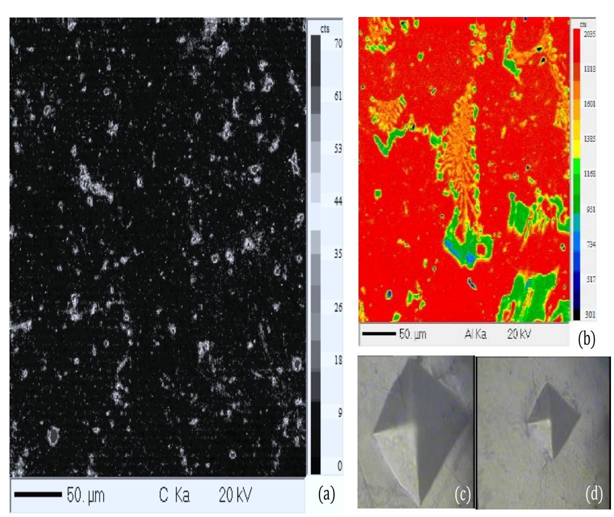

Fig. 5. (a) and (b) EPMA result of a composite showing graphene particles and aluminum matrix (c) and (d) Microhardness test result of composite and pure aluminum metal.

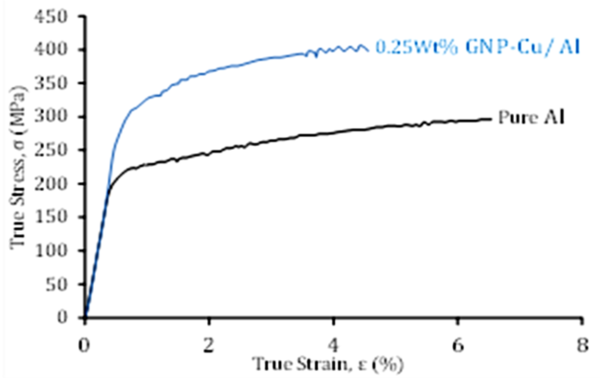

Fig. 6. Stress-strain graph for Al/Cu-GNP composite.

Fig. 6 shows the true stress-strain curve of the composite sample in comparison with the pure aluminum sample. The average value of yield and ultimate tensile strength is given in the **Table 1**. The increase in the strength values is significantly good because 0.25% of grapheme platelets dispersed easily in the matrix rather than the coagulation. The effect was also good because the graphene platelets were trapped inside copper particles and hence, stayed uniformly dispersed into aluminum matrix. The increase in tensile properties of the Gr/Al composite can be explained by the load transfer ability of graphene in the matrix material. During electrostatic self-assembly, the larger graphene particles are protected which in turn increases the contact area between the matrix and graphene resulting in the increased load transfer sites[**11**]. Previous literature has also shown that with an increase of graphene content also increases the tensile strength but rather the decrease in elongation is also dominant. The decrease of elongation is because the graphene is mainly distributed along the boundary of grain which causes loss of association among the aluminum powder thus decreasing the elongation properties of the composite.

Table 1. Mechanical properties of 0.25 GNP-Cu/Al composite at room temperature.

MATERIAL	TENSILE	
	YIELD STRENGTH (YS)	ULTIMATE STRENGTH (UTS)
PURE	222 MPa	296 MPa
0.25 GNP-Cu/Al Composite	301 MPa	406 MPa

Conclusion

The morphological and mechanical behavior of graphene aluminum composite has been studied by using an innovative approach of indenting graphene particle into the copper particle as reinforcement by using high energy ball milling process with sampling time as 30 min and 60 min respectively. The composite was prepared by casting process using argon gas environment. The FE-SEM analysis has shown various cracks in the surface of copper powder with graphene platelets in between the fracture. The composite sample formed has copper particle surrounding the graphene platelets which in turn has created an Al-Cu and GNP-Cu interface.

Mechanical characterization of the sample has shown increase in yield strength of composite from 222 MPa to 301 MPa and ultimate tensile strength from 296 MPa to 406 Mpa. The Vickers hardness of composite was found to be 87 HV which was 36.78 % higher than the pure aluminum sample.

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