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Enhanced tensile properties of aluminium matrix composites reinforced with graphene encapsulated SiC nanoparticles

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Abstract

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Keywords

encapsulated, graphene, nanoparticles, reinforced, sic, composites, matrix, aluminium, properties, tensile, enhanced

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Enhanced tensile properties of aluminium matrix composites reinforced with graphene encapsulated SiC nanoparticles

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Abstract

Due to a high propensity of nano-particles to agglomerate, making aluminium matrix composites with a uniform dispersion of the nano-particles using liquid routes is an exceptionally difficult task. In this study, an innovative approach was utilised to prevent agglomeration of nano-particles by encapsulating SiC nano-particles using graphene sheets during ball milling. Subsequently, the milled mixture was incorporated into A356 molten alloy using non-contact ultrasonic vibration method. Two different shapes for graphene sheets were characterised using HRTEM, including onion-like shells encapsulating SiC particles and disk-shaped graphene nanosheets. This resulted in 45% and 84% improvement in yield strength and tensile ductility, respectively. The former was ascribed to the Orowan strengthening mechanism, while the latter is due primarily to the fiber pull-out mechanism, brought about by the alteration of the solidification mechanism from particle pushing to particle engulfment during solidification as a consequence of high thermal conductive graphene sheets encapsulating SiC particles.

Keywords: Metal-matrix composites (MMCs) (A); Graphene sheets (A); Mechanical properties (B); Casting (E)

1. Introduction

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1 The achievement of both strength and ductility for aluminium alloys reinforced with ceramic
2 particulates is very useful for a wide range of safety applications. However, although aluminium
3 alloys reinforced with ceramic particulates enhance the tensile strength, they suffer from inadequate
4 ductility [1, 2]. This is, in fact, a well-known bottleneck that limits the widespread engineering
5 application of micro-composites especially when there is high ceramic particle content [3, 4].
6 To attain a higher strength and retain ductility of the composite, nano-sized ceramic particles are often
7 used [5, 6]. The result is a metal matrix nano-composite (MMNC) [7, 8]. In such cases, it is extremely
8 challenging to distribute and disperse the nano-particles uniformly in the metal matrix. This is
9 especially true for using liquid based processing routes such as casting, because of the large surface to
10 volume ratio and the poor wettability of nano-particles in most metallic melts [9, 10]. It was found
11 that composites manufactured specifically by liquid methods such as stir casting suffer from particle
12 pushing [11], resulting in rejection and agglomeration of particles from the growing solid/liquid
13 interface during solidification. These problems easily induce the agglomeration and clustering of the
14 nano-particles in the matrix, prompting low tensile properties, especially ductility, [associated with](#)
15 [intergranular fracture mode in the final solidified material \[12-14\]](#).
16 The key to prolonged ductility is, therefore, to disperse these nano-particles into the grain interior,
17 rather than having them agglomerated and concentrated at the grain boundaries, which can endanger
18 elongation by causing cracks in nano-particles settled at the grain boundaries [13]. The former is
19 accomplished by manipulating the shape of the interface and its curvature, controlling the interaction
20 between the particle and the interface. It has also been suggested that the shape of the interface behind
21 the particle is also dependent on the thermal conductivity of the particle and the melt [15, 16]. Khan et
22 al. [16] showed that when the thermal conductivity of the particle is greater than that of the melt, the
23 shape of the interface behind the particle changes from convex to concave. [This difference at interface](#)

curvature brings about engulfment of particles in the matrix instead of segregation and pushing into the interdendritic regions, conferring improved tensile properties on the produced composites [17].

Many researchers have focused on using innovative methods to incorporate nano-particles into the molten aluminium, making an MMNC with a suitable dispersion and avoiding nano-particle agglomeration [10]. The milling of nano-particles with metallic powders such as aluminium and ultrasonic assisted casting methods, especially in the semi-solid state, are the most important methods which have been proposed in this regard. However, these methods are still in development, especially where a high reinforcement loading is sought [3, 14, 18] in the implementation of thermal models to predict the interaction at the solid/liquid interface during solidification.

In addition to the unprecedented characteristic of graphene, i.e. a single-atom-thick sheet of sp^2 hybridised carbon atoms, as a strengthening nanofiller in the world of polymer matrix composites [19, 20], recent studies have shown that graphene can also be considered as an effective reinforcement for metal matrices [21]. However, to the best of our knowledge, most of these studies [21, 22] have only considered the mechanical property changes after the implantation of graphene nanosheets (GNSs) as a new nano filler for metal matrix composites, but exploiting the promising characteristics of GNSs, such as the high thermal conductivity and anchoring nano particles, to harness the structural uniqueness and full potential of utilising nano-particles during the fabrication process remains largely unexplored. For instance, it has been reported that graphene sheets possess the unique feature of having a two-dimensional shell(s) which can nucleate and anchor nano-particles on the edges and surface[23, 24], however, the ability of graphene sheets to alleviate the agglomeration of nano-particles during solid and liquid processing for the production of metal matrix composites has hitherto not been reported.

In this study, an innovative nanocapsulating route is implemented to exploit the high thermal conductivity and flexibility of graphene sheets as wrapping shells to diminish the pushing and

1 agglomeration of SiC nano-particles during milling and the subsequent semi-solid casting process.
2 The effect of using 1 Wt. % GNSs to alleviate the agglomeration of nano SiC particles and subsequent
3 enhancement in tensile properties of the composite, produced using a process encompassing milling
4 and semi-solid stir casting, was investigated using high resolution transmission electron microscopy
5 (HRTEM) and tensile tests, respectively. In order to gain insight into the relationship between the
6 microstructure of the fabricated composite and its tensile properties, two mathematical models were
7 developed, making use of the Orowan strengthening mechanism, and the models were further
8 validated with tensile test results.

9 **2. Experimental Procedures**

10 In order to prepare a mixture containing nano SiC and GNSs reinforcements, high purity aluminium
11 powder (45 μm , supplied from Alpha Aesar Company with 99.5 % purity), a sufficient amount of
12 nano- β -SiC particles (45 nm, supplied from Nanostructured & Amorphous Materials, Inc.) and pure
13 pristine monolayer graphene with the average lateral size of 550 nm (supplied from Graphene
14 Supermarket) were used.

15 The ball milling process was performed in a Fritsch Pulverisette P5 planetary ball mill without
16 interruption under high purity (99.999%) argon gas in a liquid nitrogen environment (cryomilling).
17 During cryomilling, liquid nitrogen was constantly added to compensate for evaporation. The stainless
18 steel vial was sealed with an elastomeric O ring. The stainless steel balls to powder weight ratio is
19 15:1, and the rotation rate of the vial is 250 rpm under a total milling time of 2h. The amount of GNSs
20 and SiC nano-particles was adjusted to 83 Wt. % SiC and 17 Wt. % graphene. These components
21 were milled for 0.5h without aluminium powder. Subsequently, the milling was continued for 1.5h by
22 adding aluminium powder to the mixture, containing graphene and SiC, by setting the aluminium
23 weight equal to 45Wt. % of the total SiC and graphene powders, to enhance the incorporation of the
24 nano SiC particles into the molten aluminium.

1 Powder injection into molten A356 aluminium alloy was conducted with high purity (99.999%) argon
2 atmosphere (6 lit/min) in the semi-solid state. Table 1 demonstrates the chemical composition of the
3 A356 alloy used in this study. Two distinct methods including (a) as received SiC particles and (b)
4 composite powder containing aluminium powder and SiC particles encapsulated by graphene sheets
5 (prepared, as noted above, by the milling process) were utilised. After the entire alloy in the crucible
6 was melted, it was cooled to 605 °C and held at this temperature. This temperature lies in the solid-
7 liquid range and corresponds to a solid fraction of about 0.30 [25].
8 Then, stirring of the semi-solid alloy (using a graphite impeller) at 400 rpm was initiated, while
9 prepared powders were added to the uniformly formed vortex over a time period of approximately 5
10 min associated with adding 1Wt.% Mg as a wetting agent. Simultaneously, non-contact ultrasonic
11 casting was utilised to apply vibration to the prepared melt. This was done using an ultrasonic
12 chamber (Bandelin-Germany Make – Model: RK – 100H), which can vibrate at a frequency of 35
13 kHz. After the completion of particle feeding, mixing was continued for an extra 1 min. Finally, the
14 composite slurry was poured into a pre-heated cast iron mould using a bottom-pouring system. The
15 composites fabricated using as-received SiC particles and composite powder are denoted herein as
16 AR-SiC and preform samples, respectively.
17 The density of the samples was measured by the Archimedes method in order to calculate the porosity
18 of the samples. In addition, in order to investigate the formation of GNSs in the matrix and around the
19 SiC particles in the composite, HRTEM was employed using a Philips CM200 at an accelerating
20 voltage of 200 kV. The morphology evolution of the powders during milling and the fracture
21 behaviour of the fabricated composites were characterised using Field Emission Scanning Electron
22 Microscopy (FE-SEM) performed in a HITACHI S4160. Tensile testing was carried out using a
23 Hounsfield universal test machine at a cross-head speed of 0.5 mm s⁻¹. The dog-bone shaped tensile

specimens had a gauge size of 6 mm in diameter and 30 mm in length, according to ASTM: B557M-10.

3. Results and Discussion

3.1. Morphological characterization of powders

Fig. 1(a) shows the morphology of Al-55 Wt. % SiC-12 Wt. % graphene powder (preform), set to reach the final composition after casting. As shown in Fig. 1(a), the size of the agglomerated particles after milling is smaller than $1\mu\text{m}$, which can accommodate better dissolution and lower agglomeration during subsequent semi-solid stir casting. Basically, the dissolution of finer milled powders in the melt is easier than that of larger agglomerated ones. Three important phenomena are responsible for formation of nanodispersions before solidification. First, the dissolution of aluminium powders during injection of powders into the melt. In fact, aluminium powders act as a carrier for ceramic particles to the melt and protect them from any contact with the surface of the melt and alumina layer, and after their dissolution, the nanoparticles will be released in the matrix. Second, the power of ultrasonic vibration causes the agglomerated particles not being sintered. In fact, it is envisaged that aluminium powders which are in the inner part of an agglomerated nanoparticles act as a binder at $605\text{ }^{\circ}\text{C}$ to avoid particles separation and release, which only ultrasonic vibration could separate these nanoparticles during stirring. Finally, the importance of using semi-solid stirring must be considered. Indeed, a high amount of shear stress during mechanical blending in the semi-solid stirring causes the agglomerated particles to easily separate from each other.

Fig. 1(b) demonstrates the high magnification image of the agglomerated particles in Fig. 1(a). As can be seen in Fig. 1(b) and corresponding cross section as a right corner image, a uniform distribution of SiC particles inside the aluminium matrix was achieved after the milling process, due to the formation of graphene sheets around SiC particles in the first step of milling. In fact, it is believed that the

formation of graphene sheets can be a good strategy to diminish the high propensity of nano-particles for agglomeration during the milling process.

HRTEM experiments have been utilised to reveal the formation and dispersion of graphene sheets around SiC particles and inside the preform sample. Fig. 2(a) demonstrates the representative HRTEM image of graphene sheets encapsulating SiC particles during the milling process, appearing in the form of onion-like shells around the SiC particles. It can be seen that this innovative process has a unique capability to encapsulate SiC particles with bilayer or trilayer graphene sheets bent around the SiC particles.

As shown in Fig. 2(a), the outer graphene shells (white arrow) tightly surrounded SiC particles without any gap, following the surface curvature of the ceramic particles in order to minimise surface energy of system including particles and graphene shells. Basically, the impact forces produced during the initial step of milling is what bends the sp^2 around the SiC particles, thus forming these onion-like shells.

Fig. 2(b) demonstrates the TEM image of SiC particle distribution in the composite powder. As can be seen, a uniform distribution of SiC particles is achieved. In fact, besides the unique characteristic of this process for producing a bilayer or trilayer graphene shell encapsulating the SiC particles (Fig. 2(a)), this process is also able to prevent the agglomeration of nano SiC particles in the milled powder, as shown in Fig. 2(b), substantiating the results shown in Fig. 1(b). As shown in Fig. 2(c), the TEM elemental x-ray map also authenticates the formation of well-dispersed SiC particles throughout the composite powder.

3.2. Composite structure

3.2.1. FE-SEM investigations

Fig. 3 represents the FE-SEM microstructure of the AR-SiC (Fig. 3(a)) and preform (Fig. 3(b)) samples, respectively. There are four important points that can be deduced from Fig. 3. First, the

formation of agglomerated SiC nanoparticles is observed in the AR-SiC sample (Fig. 3(a)) compared with a uniform distribution of these particles in the preform sample (Fig. 3(b)), despite the fact that some clusters remain in the matrix (black arrows in Fig. 3(b)). As shown in Fig. 3(a) by black arrows, SiC nanoparticles in the AR-SiC sample have this propensity to be agglomerated within aluminium matrix associated with formation of microvoids at the interface of these particles and surrounding matrix, resulting in lower tensile properties.

Second, the remain of nano-gaseous pores after stirring seems to be inevitable during the semi-solid state process for both samples due to a high viscosity of the melt, even when ultrasonic waves were used. Third, the amount of nanoparticles is considerably higher for preform sample containing graphene nanosheets and aluminium powders. Forth, the presence of air gap in the agglomerated ceramic particles and formation of solidification shrinkages around these agglomerated particles could be seen in Fig. 3a. There is also some evidence of formation of nano pores within the matrix.

3.2.2. TEM investigations

In Section 3.2.1, nanostructures of the produced composites were characterised using TEM, associated with schematic illustrations to give a better insight into the effect of graphene sheets on the nanostructures of the samples. Additionally, the effect of graphene sheets on manipulation of solidification behaviour of the samples, playing an important role in manifestation of the aforementioned nanostructures, is investigated in Section 3.2.3.

Fig. 4 represents TEM nanostructure images of AR-SiC (Fig. 4(a, b)) and preform samples (Fig. 4(c-f)) after semi-solid stir casting. Fig. 4(b), (d) and (f) are schematic illustrations corresponding to the nanostructures seen in Fig. 4(a), (c) and (e), respectively. As shown in Fig. 4(a), nano SiC particles in the AR-SiC sample have a high tendency to agglomerate during stir casting in order to reduce their free surface energy. As can be seen in Fig.4 (a), the vast majority of SiC particles in the AR-SiC sample have a high propensity to agglomerate at grain boundaries instead of within the grain interior,

and this is attributed to the effect of the particle pushing mechanism during solidification, as shown by arrows in Fig. 4(b). However, as can be seen in Fig. 4(c), in the case of preform samples, the majority of well-dispersed SiC particles reside in the grain interior rather than at the grain boundaries, and this is ascribed to the effect of the particle engulfment mechanism during solidification, as demonstrated by arrows in Fig. 4(d).

In fact, the better distribution of SiC particles especially in the grain interior of the preform sample compared to the formation of agglomerated SiC particles at the grain boundaries of the AR-SiC sample can be related to various parameters involving manufacturing processes (e.g. ball milling and semi-solid routes) and other mechanisms controlling the solidification process after semi-solid casting.

First, regarding the role of milling, it is believed that graphene shells wrapping nano SiC particles can be in charge of the uniform dispersion of these particles in the milled composite powder, as shown in Fig. 2(c). From an energetic standpoint, the quasi-spherical carbon and onions are the most stable form of carbon particles [26]. When the highly curved graphene sheets are obtained as a result of high flexibility of graphene sheets and impact forces applied during ball milling, they are in a metastable state in the light of the theory of minimisation of surface area, so the need to minimise the energy drives this curling up to spherical forms like onions. Under this circumstance, nano SiC particles can be a favourable substrate to be wrapped by highly curved graphene sheets which in turn diminishes the total energy of system by alleviating the agglomeration of nano SiC particles.

Second, semi-solid processing also plays an important role in the uniform distribution of the SiC particles in the preform sample. In fact, it is believed that creation of a vortex in the semi-solid stirring process breaks solid dendrites at 605 °C because of higher friction between the SiC particles and the aluminium alloy matrix. This further induces a homogeneous distribution of the SiC particles in the matrix of the solidified A356 composite. In addition, the sedimentation of the SiC particles decreases

at the semi-solid state stirring temperature[27], which can also result in a homogeneous distribution of the SiC particles because of the presence of a solid phase]28[.

During the solidification process, instead, it is believed that the particle engulfment mechanism governs the solidification process of the preform sample, rather than the particle pushing mechanism, which is activated in the AR-SiC sample. Generally, it has been proven that the particle engulfment mechanism results in the formation of isolated particles inside the grains of metal matrix during solidification, and the particle pushing mechanism leads to the formation of agglomerated particles at grain boundaries and in the last freezing zones of the solidified material [29].

3.2.3. Solidification mechanisms

The current models addressing the incorporation of particles into a solidifying matrix can be categorised into three classes: (i) the kinetic models that predict the velocity of the solid/liquid interface which is critical for the transition from particle pushing to engulfment [30], (ii) the thermodynamic models [31] which are closely related to classical heterogeneous nucleation theory and (iii) the models based on the ratio of the thermophysical properties of the particles and the melt [32]. The latter model predicts the incorporation ability in general, that is, a dependence of the incorporation behaviour on the processing conditions. Any dependence on the morphology of the interface is not included.

The models based on the thermophysical characteristics rely on the thermal conductivity of particles and liquid using Equation (1) [33]. The subscripts p and l refer to properties of the particles and the liquid, respectively.

$$K_p > K_l \text{ for engulfment} \quad (1)$$

The models based on heat diffusivity [34] characteristics are based on Equation (2) using the thermal conductivity (k), the specific heat (c_p) and the density (ρ):

$$\sqrt{\frac{K_p \rho_p c_p}{K_l \rho_l c_l}} > 1 \text{ for engulfment} \quad (2)$$

These two models show that by enhancing the thermal conductivity of particles incorporated into the liquid matrix, the possibility of particle engulfment through the grains of the solidifying matrix increases owing to change of the interface shape from convex to concave [15, 16, 32, 33], facilitating the engulfment of particle through the matrix during solidification [15]. In essence, the lower thermal conductivity of the particles affects the temperature gradient ahead of solidification front and therefore acts as a barrier to the removal of the heat necessary for further solidification and consequently inhibits particle engulfment.

In this study, it is expected that the thermal conductivity of SiC particles encapsulated by graphene shells is much larger than those which are not encapsulated, as a result of high thermal conductivity of graphene shell covered SiC particles [35, 36]. This is because the thermal conductivity of graphene shells on SiC particles is better conserved in bilayer and trilayer GNSs than in single layer GNSs [37]. According to Equations (1) and (2), this high thermal conductivity of the SiC particles in the preform samples results in the incorporation of most SiC particles in grain interiors rather than at grain boundaries, as shown in Fig. 4(c) and corresponding schematic illustration (Fig. 4(d)).

Fig. 4(e) represents a high magnification TEM image of the preform sample containing engulfed SiC particles. The inset in Fig. 4(e) demonstrates the selected area diffraction (SAD) pattern obtained from the selected disk-shaped region of Fig. 4(e), demonstrating that this region contains GNSs with a preferred crystallographic relationship of $[111]_{\text{Al}} // [0001]_{\text{GNSs}}$ to the matrix. Fig. 4(f) shows the schematic demonstration of Fig. 4(e), representing the presence of SiC particles encapsulated by graphene shells and some disk-shaped GNSs dispersed in the matrix.

3.3. Tensile properties

Fig. 5 represents tensile properties of the preform and AR-SiC samples. Insets are sketches illustrating the microstructure corresponding to different samples. As shown in Fig. 5, the tensile strength and ductility of preform samples are improved by about 45% and 84% compared to AR-SiC samples,

respectively. These superior tensile properties are ascribed to the effect of three possible important strengthening mechanisms: (i) the Orowan strengthening mechanism related to uniform distribution of SiC particles not only at the grain boundaries but most importantly in the grain interiors as well, (ii) enhanced dislocation density as a result of the large mismatch between the thermal expansion coefficient of GNSs and the aluminium matrix and finally, and (iii) the fiber pull-out toughening mechanism.

As illustrated in the schematic image of Fig. 4, the microstructure of the preform sample contains SiC nano-particles encapsulated by graphene shells and some dispersed disk-shaped GNSs inside the grains of the aluminium matrix. In fact, graphene sheets play two important roles in enhancing the tensile properties of the preform sample. First, they reduce the susceptibility of SiC particles to cracking as a result of the onion-like shells around these particles, as shown in Fig. 4(e, f). This, in turn, enhances the threshold stress limit of the SiC particles cracking during tensile stress, resulting in higher tensile properties especially tensile elongation of the preform sample (Fig. 5). Second, these onion-like graphene shells around the SiC particles can block the movement of dislocation passing through the matrix, and make it difficult to reach the grain boundaries at the low stress levels associated with the higher yield stress of the preform sample (225 MPa) compared to the AR-SiC sample (155 MPa). Along with this unique role of the onion-like graphene shells in hampering the dislocation crossing the SiC particles through the matrix, disk-shaped GNSs dispersed within the matrix can also take a same synergic role in hampering dislocation movement within the matrix.

3.3.1. Strengthening models

Equation (3) can be employed to measure the increment in yield strength ($\Delta\sigma_y$) of the processed composite as a result of the interfering effect of disk-shaped GNSs with dislocations sliding on (111) the slip plane of the aluminium matrix, provided that the particles cannot be sheared by dislocations and are well-dispersed in the slip plane of the matrix phase[38]. In this equation, M (3) is the Taylor

factor for polycrystalline FCC alloys, G (25 GPa) is the shear modulus of the aluminium matrix, b (0.25nm) is the magnitude of the Burgers vector of dislocation, ν (0.35) is Poisson's ratio, f (0.01) is the volume fraction of the GNSs, d (10 nm) and t (3 nm) are the average diameter and thickness of at least 100 disk-shaped GNSs measured using HRTEM analysis, respectively.

$$\Delta\sigma_y^{Disk} = \frac{MGb}{2\pi\sqrt{1-\nu}} \left(\frac{1}{0.931 \sqrt{\frac{0.306\pi d_1 t_1}{f} - \frac{\pi d}{8} - 1.061 t_1}} \right) \ln \frac{1.225 t_1}{b} \quad (3)$$

Calculated results from Equation (3), using the above-noted values, demonstrate that enhancement in the yield strength ($\Delta\sigma_y^{Disk} = 229$ MPa) of the preform sample as a result of disk-shaped GNSs is significantly larger than the experimental values ($\Delta\sigma_y = \sigma_y^{Preform} - \sigma_y^{AR-SiC} = 70$ MPa) obtained from tensile tests (Fig. 5). This difference between predicted and experimental values for $\Delta\sigma_y$ as a result of the Orowan strengthening mechanism can be attributed to the formation of graphene with two different shapes including disk-shaped and onion-like morphologies. Indeed, the total volume fraction (f) of disk-shaped GNSs was set to 0.01 during the above-mentioned calculation; however, as shown in Fig. 4(e, f), graphene can also be found in the form of onion-like shells encapsulating SiC particles. So, in order to clarify the weight of onion-like graphene shells encapsulating SiC particles in yield strength increment compared to disk-shaped GNSs dispersed in the matrix of the preform sample, a modified version of the Orowan strengthening mechanism applicable for round-shaped particles is used in Equation (4)[38]:

$$\Delta\sigma_y^{Onion} = \frac{MGb}{2\pi\sqrt{1-\nu} \left(\frac{0.779}{\sqrt{f}} - 0.785 \right) d} \ln \frac{0.785 d}{b} \quad (4)$$

In this equation, all parameters have the same values as Equation (3) unless the d parameter, related to the size of the onion-like graphene shells encapsulating the SiC particles through the matrix, is set to 46nm, under this assumption that at least two graphene layers have covered the SiC particles. This was calculated by investigating more than 100 nano SiC particles, with a diameter of 45nm and

1 provided there is no agglomeration of the SiC particles, and all pure GNSs have been converted into
 2 onion-like shells. The results show that by considering these primary conditions, the maximum
 3 enhancement in yield strength of the preform sample, as a result of onion-like graphene shells around
 4 SiC particles, is limited to 58 MPa. This value is in good agreement with results of tensile tests with a
 5 better approximation, making Equation (4) the main governing model for predicting tensile properties
 6 of the preform sample. Indeed, these calculations demonstrate that the maximum increment is induced
 7 due to the fact that the formation of disk-shaped GNSs particles through the matrix can be about 4
 8 times ($\frac{\Delta\sigma_y^{Disk-shaped\ GNSs}}{\Delta\sigma_y^{Onion-like\ GNSs}} = 3.95$) bigger than the one endowed by onion-like graphene shells, evincing
 9 the major role of disk-shaped GNSs in enhancing the yield strength of the preform sample. Therefore,
 10 it can be seen that the achievement of a 45% improvement in the yield strength of the preform sample,
 11 over that of the AR-SiC sample is attributed primarily to the higher contribution of onion-like
 12 graphene shells encapsulating SiC particles. There are a number of reasons for the lower yield
 13 strength of the preform sample compared to the values predicted by Equation (3) ($\Delta\sigma_y^{Disk} = 229$ MPa):
 14 First, disk-shaped GNSs are assumed to have a habit plane perpendicular to the slip plane of
 15 aluminium (111) in the model presented. However, in fact, GNSs lie on the slip plane of the
 16 aluminium matrix. Hence, it is predicted that the amount of interaction between GNSs and dislocation
 17 gliding on the matrix slip plane is lower than in the suggested model. Nevertheless, SiC particles
 18 coated by onion-like graphene shells interact with slip planes more easily as a result of their three
 19 dimensional circular shape, which means that they have at least some sections which interact with
 20 most active slip planes during the deformation process. Second, it is also anticipated that the number
 21 of GNSs which are able to survive agglomeration, to form the undesirable graphite structure during
 22 the manufacturing process through Van der Waals interactions, is not as high as to have the strong
 23 effect suggested by the model presented (Equation (3)). Finally, as shown in Fig. 4(e, f), disk-shaped

GNSs have not been dispersed uniformly throughout the entire matrix, making it hard for them to interact with dislocation effectively.

On the other hand, it should be noted that in addition to the Orowan strengthening mechanism which confers higher yield strength on the preform sample, there are other strengthening mechanisms which play an important role in boosting the tensile properties of the composite produced. One of these is the thermally induced dislocation generated via a considerable difference between the thermal expansion coefficient of the aluminium and the graphene ($\alpha_{Al}=21.4\times 10^{-6}/k$, $\alpha_{graphene}=-6\times 10^{-6}/k$ at 300k [39], $\alpha_{graphene}=0.9\times 10^{-6}/k$ at 873-1073K [40]) during solidification. As shown in Fig. 4(e), there is a high possibility in the formation of thermally induced dislocation through the matrix during solidification.

In addition to the above-mentioned strengthening mechanism, the other reason for the higher tensile properties of the preform sample compared with the AR-SiC sample is related to the lower porosity content ($1.6\% \pm 0.3$) introduced in the sample compared to the AR-SiC sample ($4\% \pm 0.6$). This lower porosity content of the preform sample can be attributed to the milling of SiC particles with aluminium powder that causes close contact between these powders and removes the air gap between them.

3.3.2. Fracture behaviour

Fig. 6(a) and (b) show the fractographic image of the AR-SiC and preform samples after tensile tests, respectively. As can be seen in Fig. 6(a), the fracture surface of the AR-SiC sample contains cleavage surfaces (white arrow) and some big dimples (white circle), is responsible for the lower tensile ductility of the AR-SiC sample, as compared to the preform sample. In fact, the agglomeration of particles in the AR-SiC sample (Fig. 6(a) and (b)), especially at the grain boundaries, as a result of particle pushing, can facilitate the cracking of these agglomerated particles under lower stresses, and accommodate a low energy path along the grain boundaries (intergranular fracture) for crack propagation resulting in the lower tensile properties of the AR-SiC sample. In fact, externally applied

load is generally transferred from particle-lacking zones to particle-rich zones in a composite, with the damage being generated in particle-rich zones because of the higher stress concentration at small strains [28]. Furthermore, agglomerated particles are suitable sites for damage accumulation and local particle-rich zones are the most appropriate nucleation sites of cracks [1, 41, 42].

However, in the case of the preform sample, as shown in Fig. 6(b), no evidence of cleavage surfaces or big dimples was observed on the fracture surfaces, demonstrating the ductile failure of the transgranular mode. More importantly, as demonstrated by black arrows in Fig. 6(b), in addition to the unique characteristic of GNSs in blocking dislocation within the grains, they also have another interesting macroscopic feature for preventing crack propagation during the fracture process under the fiber pull-out toughening mechanism. Under such circumstances, growing cracks encounter pull-out fibers, increasing the energy needed for propagation by diminishing the load transferred from the matrix to the dispersed reinforcements resulting in more ductility [43].

3.3.3. Characterization of the SiC/Matrix Interface

The properties of MMCs are affected significantly by the characteristics of the interface between the matrix and reinforcement [44, 45]. Reinforcing particles are thermodynamically unstable and endure reaction at the SiC /matrix interface, depending on processing temperature and time. Interfacial reactions often cause the removal of desirable elements from molten alloy, increasing melt viscosity and reducing castability [46]. SiC reinforcement is not stable thermodynamically in contact with molten aluminium alloys and therefore, Al_4C_3 formation is expected depending on the temperature, environment and other parameters [1].

Fig. 7 (a) and (b) show HRTEM images of the SiC /matrix interface in the case of AR-SiC and preform samples, respectively. Formation of Al_4C_3 at 605°C and in the presence of about 7 Wt. % Si is difficult [43, 47], and consequently the detection of Al_4C_3 is not an easy task. However, the results obtained, as shown in Fig. 7(a), indicate its formation at nanoscale (2-5 nm) with a rod-like structure,

as shown in right corner inset of Fig. 7(a). As seen in Fig. 7(a), in the case of the AR-SiC samples, the Al_4C_3 layer is formed around the SiC particle while in the case of the preform sample (Fig. 7(b)), Al_4C_3 is formed only in some damaged areas of GNSs (white arrows). As demonstrated by inset of Fig. 7(b), the formation of Al_4C_3 has also been approved by SAD pattern taken from the damaged area of graphene sheets encapsulating SiC particles. Therefore, in the former, the ability for load transfer from the matrix to the SiC particles is reduced, while in the latter, it increases. Additionally, particle-rich zones produced as a result of severe agglomeration in the AR-SiC sample, limit the flow of liquid before complete solidification of the composite and therefore, the interfaces in particle-rich zones are defective. Since these zones rapidly lose their load-bearing capability, the strength of the composite containing agglomerated particles is lower than those without particle agglomeration [48]. In fact, agglomerated particles contain many inner smaller particles without any contact with the matrix. These inner fine particles are easily displaced on each other during loading due to a weak mechanical bonding, leading them to being conjoined. There is an air gap between these fine particles in which a defect-free interface could not be formed between agglomerated particles and matrix.

More importantly, it is anticipated that the interface between SiC particles and the surrounding matrix is stronger in the preform sample than in the AR-SiC sample as a result of the rough and wavy surface of the graphene shells encapsulating the SiC particles. Indeed, it should be noted that graphene will be wrinkled during heating, as shown by the black arrow in Fig. 7(b), due to its different behaviour and it will expand during cooling and solidification [49]. This rough topography of the interface augments interfacial friction in the interfacial zone of the SiC particles and the adjacent matrix during sliding of debonded particles in the fracture which, in turn, permits realisation of more toughness in the preform sample.

Moreover, the wrinkling phenomenon during solidification of the composite can lead to the formation of a low-defect interface between the aluminium matrix and the nano-particles, leading to an

1 increment in the tensile strength of the composite. Additionally, the Al_4C_3 layer around the SiC
2 particles in the AR-SiC sample cracks easily because it has a greater likelihood of containing a flaw(s)
3 which can initiate cracks and lead to a weaker interface.

4 **4. Conclusions**

5 This study proposes an innovative fabrication method, making use of encapsulating and high thermal
6 conductivity promising features of graphene, for manufacturing aluminium-based composite
7 reinforced with nano SiC particles. The former enables the agglomeration of nano SiC particles
8 during powder milling and subsequent liquid processing to be diminished, and the latter made a useful
9 change in the solidification mechanism from pushing to the engulfment of particles.

10 The augmented tensile properties of this composite can be ascribed mainly to the strengthening effect
11 of the graphene sheets manifested in the form of onion-like encapsulating shells for the nano SiC
12 particles dispersed throughout the aluminium matrix. Having considered the Orowan strengthening
13 mechanism, two different models were investigated, emphasising the major role of nano SiC particles
14 encapsulated by onion-like graphene shells in attaining greater tensile properties. This improvement is
15 also attributed to thermally induced dislocation brought about by considerable difference in thermal
16 coefficient expansion of the graphene and aluminium matrix.

17 The fractographic investigations have also demonstrated that higher tensile ductility of composites
18 containing graphene can be ascribed to lower agglomeration of SiC particulates as well as fiber pile-
19 up mechanism. The former reduces the dimple size and the latter augments the energy needed for
20 crack propagation through the matrix.

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Figure captions:

- Fig.1. (a) Morphology of Al-55 Wt. % SiC-12 Wt. % graphene powder and (b) high magnification image of (a).
- Fig.2. (a) HRTEM images of SiC particle encapsulated by trilayer GNSs, (b) TEM image of SiC particles in composite powder and (c) elemental TEM x-ray map of (b).
- Fig.3. FE-SEM micrograph of (a) AR-SiC and (b) preform samples.
- Fig.4. TEM micrograph of (a) AR-SiC sample, (b) schematic representation of (a), (c) preform sample, (d) schematic representation of (c), (e) high magnification image of preform sample associated with selected area diffraction (SAD) pattern of specified region, and (f) schematic representation of (e).

Fig.5. Stress-strain curve of AR-SiC and preform samples accompanied with schematic illustration of their nanostructures.

Fig.6. Fracture surfaces of (a) AR-SiC and (b) preform samples.

Fig.7. TEM microstructures of a SiC particle showing SiC/matrix interface in the case of (a) AR-SiC and (b) preform samples. The rod-like structure and SAD pattern of Al_4C_3 are representing in the insets of Fig. 7 (a) and (b), respectively.

Table1. Chemical composition of A356 alloy used in this study (Wt. %)

Al	Si	Fe	Cu	Mn	Mg	Zn	Ti
Bal.	7.0	<0.2	<0.2	<0.1	<0.1	<0.1	<0.2

1

2

3