Investigation of mechanical properties of additive manufactured graphene-reinforced aluminium composites

Li-Ya Liu, Qing-Sheng Yang

Department of Engineering Mechanics, Beijing University of Technology, Beijing 100124, China

1 Corresponding author, Tel./Fax: +86-10-67396333, E-mail: qsyang@bjut.edu.cn

Abstract. Graphene considered as an ideal reinforcement for metal matrix composites (MMCs) because of its excellent optical, electrical and mechanical properties has important application prospects in materials science, micro-nano processing and so on. Meanwhile, additive manufactured MMCs becomes the hotspot of current research due to the advantages of precise and controllable structure and easy implementation of modularization. In this paper, a two-dimensional rate-dependent crystal plasticity model using the numerical model is developed to simulate the mechanical behaviors of additive manufactured graphene-reinforced aluminium matrix composites (AMCs) under tensile load in a mesoscale. The mechanical properties are described by varying the volume fractions and distribution patterns of graphene. The results verify that graphene is the main load-bearing part of AMCs. The volume fraction and distribution pattern of graphene play an important role in the crystal dislocation strengthening of AMCs. Moreover, it is proved that the additive manufactured graphene-reinforced AMCs have a potential improvement with increased graphene volume fraction and optimized geometric graphene parameters.

Keywords: Additive manufacturing; Graphene/aluminum; Tension; Crystal plasticity

1. Introduction

AMCs have become one of the most important composites in MMCs due to their excellent properties of light weight, high specific strength and low thermal expansion coefficient, which are widely used in civil and military fields [1,2]. Additive manufacturing (AM) technology can achieve AMCs parts with lightweight structure and composite performance [3]. Common reinforcements such as silicon carbide, boron carbide and fibers need to improve the comprehensive properties of the materials, especially the plasticity of the composites, which limits the application of the AMCs [4,5]. Graphene is a two-dimensional material consisting of carbon atoms in sp² hybrid orbital with a honeycomb lattice monoatomic layer thickness [6]. Recently, graphene has attracted tremendous attention owing to its extraordinary mechanical properties, light weight, large surface areas, and unique electronic [7]. In addition, graphene can be prepared in large quantities by chemical exfoliation of graphite, which makes it an ideal reinforcement in AMCs [8]. However, the current research mainly pays close attention to on graphene/polymer and graphene/ceramic composites, there few efforts have been made to understand of graphene/MMCs [9]. Moreover, the experimental research of mechanical properties of additive manufactured graphene-reinforced MMCs has some limits, such as complicated experimental process, long time interval, high research cost and waste of raw materials, which seriously limits the applications of additive manufactured graphene-reinforced MMCs [10]. Therefore, the numerical simulation of additive manufactured graphene-reinforced AMCs products in a mesoscale is necessary, which can provide a theoretical basis for the manufacture and safety.
evaluation of AMCs and has great economic and time advantages, especially for rare and costful metal composites.

Recently, most of the simulating studies on MMCs are simulated by molecular dynamics. Long et al. [11] investigated the shock response of graphene/copper composites with molecular dynamics simulations and found that graphene formed wrinkles and folds under compression, while fractures upon release and tension for parallel shocks. It is proved that the interatomic interaction between aluminium and graphene is capable of exfoliating the graphene layers by Dixit et al. [12] with molecular dynamics simulations. Hereafter, Chu et al. [13] studied the anisotropic mechanical properties of graphene/copper MMCs. The results indicate that the strength of MMCs greatly increased with the addition of graphene parallel to the loading direction. However, the computational amount of molecular dynamics simulation is relatively large and can only be used to solve small-scale problems. Jia [14] simulated the tensile test with an equal proportion micro-finite element model of graphene-reinforced MMCs by developing a random algorithm in Abaqus, and analyzed the effect of processing parameters on the average cutting force of graphene-reinforced MMCs. Then, Su et al. [10] established a three-dimensional composite structure model of graphene/AMCs by using a self-developed composite structure modeling program based on Python language, and realized controllable reconfiguration distribution of graphene size, morphology, orientation, location and content. Nevertheless, the traditional finite element model mostly treat the metal as the isotropic material or use the improved constitutive model to study the mechanical properties of graphene-reinforced MMCs, so the whole deformation process of MMCs cannot be accurately described in a mesoscale. The above disadvantage can be solved by the crystal plasticity finite element method (CPFEM), a method combined by the crystal plasticity theory and the finite element method [15,16]. In recent years, CPFEM have been employed to study the mechanical properties of metal [17-20]. Yet, few studies on the mechanical behavior of additive manufactured graphene-reinforced MMCs have been reported so far, especially on the additive manufactured graphene-reinforced AMCs.

In this paper, a two-dimensional rate-dependent crystal plasticity model using the numerical model was developed to simulate the mechanical behaviors of graphene-reinforced AMCs under tensile load in a mesoscale in a mesoscale. Then the influences of graphene parameters on graphene-reinforced AMCs polycrystal were investigated.

2. Simulation process of additive manufactured graphene-reinforced AMCs

![Figure 1. Models of additive manufactured graphene-reinforced AMCs under uniaxial tensile. (a) AMCs reinforced by graphene within the grain; (b) AMCs reinforced by graphene at grain boundary (GB).](image)

The graphene nanosheets of additive manufactured graphene-reinforced AMCs observed by transmission electron microscope (TEM) morphologies were uniformly distributed in the whole aluminium (Al) matrix, and mostly located around the boundaries of the Al grains while a small portion located in other regions [21,22]. And the volume fraction of reinforcing phase is a significant
factor affecting the mechanical properties of composites. Therefore, there are different graphene-reinforced AMCs models in this work.

As shown in Figure 1, a two-dimensional graphene-reinforced AMCs polycrystal model with 49 grains and a size of 0.7×0.7 μm². Graphene dispersed respectively in boundaries and interior of Al grains at a certain volume ratio with a length of 100 nm and a thickness of 10 nm [23]. Young’s modulus and Poisson’s ratio of Graphene were taken as 1050 GPa and 0.186, respectively [24]. The static analysis of the interface of the metal laminates was carried out. In order to study the mechanical properties of graphene-reinforced AMCs polycrystal in tensile conditions, an uniaxial tensile boundary condition was applied. During the simulation process, the maximum deformation of the model was set up to 25%, while the elements of CPE4R were employed.

3. CPFE method of additive manufactured graphene-reinforced AMCs

The nature of metal deformation can be understood fundamentally at the grain scale from CPFEM [25]. The geometric relevance to deformation of crystal is based on the multiplicative decomposition theory of the deformation gradient in the crystal plasticity theory. The total deformation gradient of a single crystal can be decomposed as

\[ F = F^e F^p \]

in which, \( F^e \) and \( F^p \) denote the elastic and plastic part of the total deformation gradient respectively. Similarly, the total velocity gradient can be also decomposed into elastic and plastic parts of the velocity gradient and expressed as

\[ L = \dot{L} \cdot L^{-1} = L^e + L^p \]

The velocity gradient of plastic deformation related to the slip rate on the α-th slip system \( \dot{\gamma}^\alpha \) can be described by

\[ L^p = \sum_{\alpha=1}^{N} \dot{\gamma}^\alpha s^\alpha \otimes n^\alpha \]

where, \( s^\alpha \) and \( n^\alpha \) are the slip direction and the slip plane normal direction on the α-th slip system respectively.

The slip rate of slip system can be calculated according to the hardening equation.

\[ \dot{\gamma}^\alpha = \dot{\gamma}_0^\alpha \frac{\tau^\alpha}{\tau^e} \left[ \begin{array}{c} \tau^e \\ \tau^\alpha \\ \tau^e \end{array} \right]^{m-1} \]

where \( \tau^\alpha \) denotes the shear stress on the α-th slip system (Schmid stress), \( \dot{\gamma}_0^\alpha \) is the reference shear strain rate while \( m \) represents the rate sensitivity coefficient. The reference shear stress on the α-th slip system \( \tau^e_\alpha \), it can be expressed by

\[ \tau^e_\alpha = \sum_{\beta=1}^{m} h_{\alpha\beta} |\dot{\gamma}_0^\beta| \]

in which, the hardening modulus \( h_{\alpha\beta} \) is

\[ h_{\alpha\beta} = q_{\alpha\beta} h_0 \left( 1 - \frac{\tau^\beta}{\tau^s} \right)^\alpha \]

where \( h_0 \) represents the initial hardening rate, while \( \tau^s \) is the saturation flow stress and \( q_{\alpha\beta} \) denotes the latent hardening matrix.

Considered here the material performance of additive manufactured graphene-reinforced AMCs is the same as that of graphene-reinforced AMCs manufactured by traditional process, they are just different in the manufacturing process, thus the CPFEM can be used to study additive manufactured graphene-reinforced AMCs. And the material parameters used in this work are listed in Table 1 [26].
Table 1. The material parameters of CPFEM.

|   | $C_{11}$ (GPa) | $C_{12}$ (GPa) | $C_{44}$ (GPa) | $\dot{\gamma}_0$ | $m$ | $\tau_0$ (MPa) | $\tau_s$ (MPa) | $h_0$ (MPa) |
|---|----------------|----------------|----------------|-----------------|-----|----------------|----------------|-------------|
|   | 106.75         | 60.41          | 28.34          | 0.001           | 0.02| 12.5           | 75             | 60          |

4. Results and discussion
The results verify that the addition of graphene reinforcement enhances the strength of the additive manufactured AMCs greatly. As illustrated in Figure 2, the stress distribution of pure Al is very uneven. The reason is that the simulated model is based on the CPFEM and treats the metal as the anisotropic material. Then, the interactions among polycrystalline grains results in an uneven distribution stress and an orange peel phenomenon of the additive manufactured AMCs. Moreover, the stress in Al matrix is far less than that in graphene reinforcement, and graphene reinforcements bear more loads than Al matrix, which indicates that the load in Al matrix can be transferred to graphene reinforcement through the interface. As shown in Figure 3, the amount of deformation of the Al matrix is far greater than that of graphene. Therefore, geometrically necessary dislocation must be released during the deformation process to coordinate the non-uniform deformation between graphene and Al matrix, which leads to an increase in dislocation density and aggravates the polycrystalline orange peel phenomenon.

Figure 2. Equivalent stress distributions of AMCs. (a) Pure polycrystalline Al; (b) AMCs reinforced by graphene at GB.

Figure 3. Logarithmic strain distributions of AMCs. (a) Pure polycrystalline Al; (b) AMCs reinforced by graphene at GB.
4.1. The mechanical properties of AMCs reinforced by graphene at GB

The graphene nanosheets of additive manufactured graphene-reinforced AMCs are mostly located around the boundaries of the Al grains observed by TEM morphologies [21, 22]. In order to investigate the influence of graphene volume fraction on the mechanical properties of additive manufactured AMCs, other factors were assumed to stay constant, and the mechanical behavior of additive manufactured graphene-reinforced AMCs with volume fractions of 1%, 2% and 3% under tensile load were simulated respectively.

As illustrated in Figure 4, the stress of Al matrix decreases gradually with the increase of graphene volume fraction, and the maximum equivalent stress of Al matrix decreases from 66 MPa to 46 MPa. The reason is that the volume fraction of graphene increases so that more graphene is distributed in the composites, and the specific surface area of graphene increases. As a result, more loads are transferred from Al matrix to graphene, and the load-bearing capacity of graphene increases, which in turn reduces the stress on the surrounding Al matrix. From the Figure 5, the strain values of Al matrix near the left and right ends of graphene reinforcement are less than the other regions, and the strain gradient of Al matrix is formed. It can be seen from the figure that the logarithmic strain values of the graphene-reinforced AMCs does not change significantly with the increase of volume fraction of graphene reinforcement.

![Figure 4](image-url)

**Figure 4.** Equivalent stress distributions of AMCs for various graphene volume fractions. (a) Pure polycrystalline Al; (b) AMCs reinforced by graphene with volume fractions of 1%; (c) AMCs reinforced by graphene with volume fractions of 2%; (d) AMCs reinforced by graphene with volume fractions of 3%.
Figure 5. Logarithmic strain distributions of AMCs for various graphene volume fractions. (a) Pure polycrystalline Al; (b) AMCs reinforced by graphene with volume fractions of 1%; (c) AMCs reinforced by graphene with volume fractions of 2%; (d) AMCs reinforced by graphene with volume fractions of 3%.

Figure 6. The Stress-strain responses of AMCs for various graphene volume fractions.

Figure 6 shows the variation of nominal stress of AMCs polycrystal with applied strain for different volume fractions of graphene. By observing the elastic stage and yield point, the critical slip
stress of AMCs polycrystal increases with the increase of graphene volume fraction in the same strain. That is, the graphene-reinforced AMCs polycrystal with the larger graphene volume fraction is the more difficult to slip. It is also found that the AMCs polycrystal with the larger graphene volume fraction has the better resistance to deformation. This is due to the increase of graphene, which makes the deformation and dislocation movements of AMCs polycrystal are more difficult, and thereby the material strength will also increase. In summary, the addition of graphene reinforcement enhances the strength of the additive manufactured AMCs to a certain degree.

4.2. The mechanical properties of AMCs reinforced by graphene within the grain

The prediction of the relationship between the graphene location and the mechanical properties of AMCs has theoretical and practical significance for the effective utilization of additive manufactured graphene-reinforced AMCs. In order to better study the mechanical properties of AMCs reinforced by graphene within the grain, the graphene reinforcements are assumed to be placed in the grain interior and at GB respectively, while the mechanical properties of the two were compared.

Graphene reinforcements bear more loads than aluminium matrix in both cases, which obtained by stress distribution of Figure 7. Then, the AMCs with graphene within the grain have a larger stress values than those at GB. Besides, from the strain nephogram (Figure 8), the polycrystalline orange peel phenomenon of AMCs with graphene within the grain is more serious than that at grain boundaries. This is because the grain refinement due to the addition of graphene within the grain could further hinder the propagation of dislocation resulting in the matrix being less prone to deformation. Thus, the stress values of the AMCs with graphene within the grain increases. In turn, the started slip systems are concentrated around the graphene, which can lead to the localization of damage and failure.

**Figure 7.** Equivalent stress distributions of AMCs for various graphene locations. (a) Pure polycrystalline Al; (b) AMCs reinforced by graphene within the grain; (c) AMCs reinforced by graphene at GB.

Figure 9 demonstrates that the critical slip stress value of AMCs polycrystal with graphene reinforcement within the grain is the largest in the same strain. In other words, the slip systems of AMCs polycrystal with graphene reinforcement within the grain is most difficult to start by observing the yield point. Meanwhile, the resistance to deformation of intragranular graphene is higher than that of graphene location at the GB, as illustrated in plastic stage of the curve. The reason is that adding graphene within the grain can increase the number of grain boundaries, which hinders the movement of dislocations and the grain growth, while the polycrystal is harder to deform. Nevertheless, the capacity to resist to deformation enhances. Moreover, the mechanical properties of additive manufactured graphene-reinforced AMCs could be improved better by placing graphene within the grain compared to the other location.
Figure 8. Logarithmic strain distributions of AMCs for various graphene locations. (a) Pure polycrystalline Al; (b) AMCs reinforced by graphene within the grain; (c) AMCs reinforced by graphene at GB.

Figure 9. The Stress-strain responses of AMCs for various graphene (Gr) locations.

From the Figure 6 and Figure 9, it is found that the addition of graphene reinforcement enhances the strength of the additive manufactured AMCs to a certain degree. And the graphene has huge potential applications as the ideal reinforcement in ACMs. This result is consistent with the conclusion that the tensile stress of ACMs increases with the addition of graphene [21]. While the present calculation show that the enhancement effect of graphene reinforcement is not particularly obvious compared with the experiment. The reason is that the absolute length of graphene in this work is at the grain level, hence it is small. Moreover, the graphene contents and the angle between graphene and loading direction also affect the enhancement effect of graphene reinforcement. In order to obtain additive manufactured graphene-reinforced AMCs with higher tensile strength, methods such as increasing the content and size of graphene can be used.

5. Conclusions
In this paper, a rate-dependent crystal plasticity theory was applied to characterize the mechanical behavior of additive manufactured graphene-reinforced AMCs under tension. The effects of graphene parameters on graphene-reinforced AMCs polycrystal were discussed. The findings of this paper can be summarized as follows.

(1) The addition of graphene reinforcement enhances the strength of the additive manufactured AMCs greatly. Moreover, the graphene-reinforced AMCs polycrystal with the larger graphene volume fraction is the more difficult to slip and has the better resistance to deformation.
(2) The graphene location has a great influence on the mechanical behavior of polycrystal of graphene-reinforced additive manufacturing AMCs. The slip systems in polycrystal with graphene reinforcement at the GB are more prone to start than with graphene reinforcement within the grain.

(3) The Al matrix near graphene reinforcement is prone to stress concentration and the large plastic deformation, which is easy to cause the formation and expansion of crack. Attention should be paid to the preparation of graphene-reinforced additive manufacturing AMCs.

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