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Shape effects on nanoparticle engulfment for metal matrix nanocomposites

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ABSTRACT

Obtaining a uniform dispersion of the nanoparticles and their structural integrity in metal matrix is a prominent obstacle to use the intrinsic properties of metal matrix nanocomposites (MMNCs) to the full extent. In this study, a potential way to overcome the scientific and technical barrier of nanoparticle dispersion in high performance lightweight MMNCs is presented. The goal is to identify the shape and size of Al_2O_3 nanoparticle for its optimal dispersion in Al matrix. Critical velocity of solidification is calculated numerically for spherical, cylindrical and disk-shaped nanoparticles using an analytical model which incorporates drag force, intermolecular force and inertia effect. The results show that it is possible to reduce the critical solidification velocity for nanoparticle capture by 6 times with proper shape modification.

1. Introduction

Incorporation of nanoparticles (NPs) into lightweight metals such as Al and Mg alloys can lead to unusually enhanced chemical, physical, and mechanical properties, which offer significant advantages over iron-based alloys to improve the energy efficiency of numerous products. Due to metal matrix nanocomposites' (MMNCs) great potential in aerospace, automotive and military applications, it is important to establish a reliable and cost effective processing technology to enable a high volume and net shape production of complex MMNC structural components with reproducible structures. Traditional fabrication methods, such as high-energy ball milling, electroplating, and sputtering, cannot be used for this purpose without significant post-processing [1–5]. More recently, there have been several processing technologies available to fabricate these MMNCs, such as powder metallurgy [6], severe plastic deformation [7,8], friction stir processing [9], and solidification processing (liquid infiltration [10,11], disintegrated melt deposition [12], spray atomization [13], and ultrasonic-cavitation based technique [14–16]). Of these processing methods, the solidification processing is a highly adaptable and cost-effective method to produce large volume of near-net-shape MMNCs with complex features in bulk for various industrial applications.

In order to attain the desired properties of MMNCs fabricated with the solidification processing method, NPs should be incorporated and dispersed uniformly in the molten metal. However, it is extremely challenging to achieve the uniform dispersion of NPs in molten metals since there is no repulsive force to balance the attractive van der Waals force between NPs for their thermodynamically stable dispersion in the melt. Moreover, aggregation of NPs during the solidification processing is another significant obstacle to produce reliable MMNC products with uniformly dispersed and distributed NPs. The advancing solidification front would push the NPs during solidification because of the repulsive interaction forces, thereby causing re-agglomeration of NPs in the matrix.

The solidification front–particle interaction has been widely studied both experimentally [17–21] and theoretically [22–27] in the past few decades. The experimental studies show that there exists a critical velocity, v_{cr}, which determines whether the particles are pushed away from the solidification front or engulfed in the metallic solid [28,29]. Usually, the theoretical studies aim to find the critical velocity of the solid–liquid interface and most of them utilize the physics governing the interface–particle interaction. The force balance on the particle among the attractive and repulsive forces at the solid–liquid interface is established to determine the critical solidification velocity. Due to the molecular interactions between the particle and solidified metal through metal melt, forces such as van der Waals forces [30–32] (or disjoining pressure [18,33]), and interfacial forces [22] arise.

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Convective [34,35] and Brownian forces [36] due to the collision of randomly moving liquid particles with NPs and viscous drag force [37,28,38,39] due to motion of the NP in molten metal are other forces acting on the particle.

The analytical models developed for solidification front–particle interaction can be classified as (i) steady-state models [28,19,30,23,21,40] and (ii) dynamic models [29,34,35]. Steady-state models determine the critical interface velocity from the steady-state balance of the forces acting on the particle. These models assume that there exists a critical steady-state velocity of the solidification front which results in continuous pushing of the particle ahead of the front if the solidification front velocity is less than this critical velocity. To capture the particle, the velocity of the solidification front has to be higher than this value. However, it has been shown that the process of particle pushing/engulfment has a transient nature and dynamics of the system has to be considered in the model for accurate description [29].

All of the models mentioned so far consider micron to millimeter sized large particles with spherical shapes. A few number of models exist which consider nano-sized particles but these models consider only spherical shapes as well [36,41,42]. By examining the physics and mechanics of the interaction between the solidification front and NPs, it can be shown that for a given solidification front velocity, both the repulsive and attractive forces are dependent on the size and shape of the NPs. Therefore, it is important to understand the relationship between the critical velocity and NP morphology. Such understanding will facilitate the selection and optimization of MMNC systems for uniform distribution of NPs.

The study presented in this paper aims to improve the NP capture during solidification through modifying the shape and size of NPs. We study the kinetics of the solidification front–NP interaction by using a dynamic model for three different NP shapes, namely sphere, parallel cylinder, and horizontal disk/perpendicular thin cylinder. Critical velocities of solidification are calculated numerically using the analytically derived force expressions for each NP shape at different sizes and a comparison is given for achieving a better NP distribution in the solidified metal matrix.

2. Theoretical analysis

2.1. Model description

The crystal growth in MMNCs is typically dendritic or cellular. When compared to the size of the nanoparticle, the dendritic/cellular solidification front can be assumed planar for solidification modeling. In addition, molecular interactions between NPs and solidification front become significant when the distance in-between is very small. Therefore, the analytical model assumes a plane solidification front and the distance between the front and the particle is the thickness of several layers of liquid metal atoms which is much smaller than the size of particle. The model also assumes that the interaction between NPs is negligible due to the long distance between them since the volume fraction of the NPs is considered to be less than 5%. Therefore, it is assumed that no aggregation of NPs occurs, and a single NP is assumed to interact with the solidification front. The nanoparticle shapes (sphere, parallel cylinder, and horizontal disk) that are considered in the model are shown in Fig. 1, where R is the radius of the shape, t is the thickness of the disk, L is the length of the cylinder, and D is the distance between the particle and the solidification front.

In this research, we focus our investigation on the Al–Al_2O_3 MMNCs for the following reasons: (1) Al and Al_2O_3 have been studied extensively, their material properties and physical behavior are well understood; (2) due to the availability of the component materials, Al–Al_2O_3 MMNCs provide excellent economic advantage and are suitable for producing large structural components; (3) synthesis techniques have been developed for manufacturing Al_2O_3 nanoparticles with all 3 types of nanostructures with tunable geometric characteristics (Fig. 1).

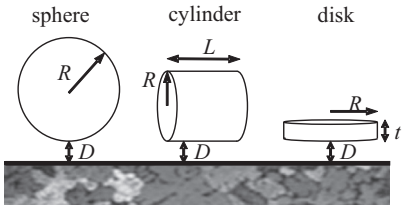

Fig. 1. NP shapes considered in the model.

In the analytical model, the Brownian force, which is the force acting on the NP caused by collision of liquid atoms with NP, is not considered since its magnitude and direction changes randomly in time and for different particles. Moreover, very small distance between the NP and solidification front induces confined Brownian motion and therefore causes less significant Brownian forces [43]. As shown in [34,35], lift force (Saffman force) due to liquid flow parallel to the interface (convection effects) plays a significant role in particle pushing/engulfment process. Because of the velocity gradient in the boundary layer, particle experiences a lift force from the lower velocity region to the higher velocity region. However, the spherical particle considered in [34,35] has a diameter of 250 μm which is three orders of magnitude higher than the particles considered in our study. Therefore, it is reasonable to assume that the velocity gradient across a NP is small and the Saffman lift force is negligible. For metallic nanocomposites, the electrostatic forces between the liquid and solid metal is neutralized due to electron interchange between them [36] and thus omitted from the model. During the motion of NP, it is assumed that there is no adhesion of extra fluid or gas film on the NP and no chemical reaction between the NP and the metal melt. The gravity and buoyancy forces are neglected as well since these forces are much smaller than the van der Waals force and drag force for Al–Al_2O_3 nanocomposite, as shown in Section 3. Therefore, only the non-retarded van der Waals forces and drag forces are taken into account in the model.

2.2. Forces acting on a NP

2.2.1. The van der Waals force

The van der Waals forces are interaction forces between atoms or molecules originating from electromagnetic fluctuations. Although they are attractive between objects of the same materials, they can be repulsive depending on the material system (object–medium–object) [44,45]. These forces have been studied extensively and explicit expressions have been derived for several interacting bodies [44]. Magnitude of the van der Waals force depends on material properties, shape and size of the interacting bodies, material properties of the medium they are interacting through, and the distance between the two bodies.

The van der Waals potentials and forces between several common shapes (e.g. sphere/cylinder/disk-plane wall) are given in [44]. The expressions for forces are obtained by derivation of the corresponding potential with respect to the separation distance, D. This potential/force is called non-retarded van der Waals potential/force at small separations ($D < 10$ nm).

Spherical particle: The non-retarded van der Waals interaction potential between a sphere and a semi-infinite plane wall (Fig. 1) is given as [44]

$$W_{vdW}^{sphere} = -\frac{A}{6}\left(\frac{R}{D}+\frac{R}{2R+D}+\ln\frac{D}{2R+D}\right) \quad (1)$$