An Experimental and Modeling Study of Al-based Nanocomposites Fabricated by Ultrasonic Cavitation and Solidification Processing

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Abstract
In the present investigation, A356 nanocomposites were fabricated via ultrasonic stirring technology (UST) in a coreless induction furnace. SiC nanoparticles were used as the reinforcement. Nanoparticles were added into the molten metal and then dispersed by ultrasonic cavitation and acoustic streaming assisted by electromagnetic stirring. The UST was also applied during the solidification process. The nanocomposite microstructure was investigated by optical microscopy and SEM. The distribution of SiC nanoparticles in the A356 alloy matrix was also analyzed. The SEM and EDS analyses showed that both the matrix microstructure and the SiC particles dispersion into the matrix can be considerably improved when ultrasonic cavitation, induction melting and stirring, and solidification processing techniques are applied together. Molecular dynamics simulations were conducted to analyze the complex interactions between the nanoparticles and the S/L interface.

Keywords:
Al-based nanocomposites; Microstructure refinement; SiC nanoparticle dispersion, Molecular dynamics simulation, Particle pushing and engulfment.

Introduction
Al metal-matrix-composites (MMCs) have been widely used in aerospace, military application, automobile and so on, since their low cost and desirable properties, such as high wear resistance, and high specific strength, etc. [1-5]. Because normally the use of micro-size particles could jeopardize the ductility of composites, nano-size ceramic particles has been studied as reinforcement to fabricated metal-matrix-nano-composites (MMNCs). It was shown that MMNCs could significantly improve matrix properties while maintaining good ductility and high temperature creep resistance at the same time [6-8]. Mechanical stirring is widely used to produce MMCs. However, it is not suitable to fabricate MMNCs. Nanoparticles have high specific surface area, which means it is difficult for mechanical stirring to distribute and disperse them uniformly in the melt. In addition, nanoparticles will agglomerate again and float on the liquid surface after stirring due to their poor wettability with the metallic alloy melts [8].

Ultrasonic stirring treatment (UST) is a promising way to fabricate MMNCs, since it will induce nonlinear effects into the melt, such as cavitation and acoustic streaming, which can spread and break the nanoparticle clusters in the melt. Also, extremely high temperature (possibly larger than 5,000K) could help to enhance the wettability between nanoparticles and metal melts, which means the nanoparticles would not gather together or float on liquid surface when the UST processing is stopped [3-5, 9]. It was demonstrated that the application of UST during the melt solidification process refined the matrix microstructure [10]. Previous studies have indicated that the application of UST for A356-Al2O3 nanocomposites around liquidus temperature changed the matrix microstructure from dendritic grains to globular grains [3, 5, 11].

In this study, the effect of the UST processing on the microstructure of the A356-SiC nanocomposites has been studied. Additionally, the effect of nanoparticles size on their distribution in the matrix has been discussed. During the solidification process, the particles can be pushed, engulfed or entrapped by the solidification front, among which particle pushing will always lead to particle clustering, which is undesirable as it results in non-homogeneous and lower macroscopic mechanical properties [12]. The particle pushing and engulfment (PEP) is a complicated phenomenon and it is affected by many factors [13] including particle size and shape, interfacial energy between particle, liquid, and solid, as well as a temperature gradient in the melt ahead of the solidification interface.

Molecular dynamics (MD) simulations conducted at the atomic level were performed to study the interface dynamics. In the current study, the open source MD program LAMMPS (Large-scale Atomic/Molecular Massively Parallel Simulator) [14] was applied to conduct MD simulations to analyze the complex interactions between the nanoparticle and the liquid/solid interface.

Experimental Procedure
Aluminum alloy A356 was used as the matrix and SiC nanoparticles (spherical shape, average diameter range is about...
45-55 nm) were used as the reinforcement. Jia et al. [3] showed that the optimum amount of SiC is about 1.0 wt. %. An Inductotherm induction melting furnace was used to melt the alloy. When the melt temperature reached about 750°C ultrasound cavitation and stirring processing was performed onto the melt. 1.0 wt. % SiC was added into the cavitation area during a 15 min time-frame. During the UST processing, a 40 mm diameter Nb ultrasonic probe was inserted to about 50 mm beneath the melt surface. The sketch of the UST system is shown in Fig. 1. The system parameters are: amplitude is 20 microns, maximum applied power is 1.5kW and frequency is 18 kHz. The cast ingots were cylindrical in shape with a diameter of 85 mm and a height of 120 mm. To avoid the effects of different locations (cooling rates) on the microstructure [5], all samples were taken from the middle location of the ingots. Al samples were taken from a section below the probe insertion location. Optical microscopy (OM) (Nikon EPIPHOT 200) photos and scanning electron microscopy (SEM) (JEOL 7000) photos were prepared by grinding with 240, 600, 800, 1200 sand papers followed by polishing with 9 µm, 3 µm, and 1µm water based diamond suspensions.

![Figure 1. Sketch of the UST system.](image)

**Molecular Dynamics Modeling Procedure**

The dimensions of the MD geometry in the x, y, and z-direction are 40 nm × 40 nm × 40 nm. The diameter of the SiC nanoparticle is 10 nm. The simulated structure of pure aluminum is a face centered cubic (FCC) structure with a lattice parameter of 4.05 Å and with the <100> directions coincident with the Cartesian coordinates, since planes of looser packing, such as {100}, are better able to accommodate an atom that leaves the liquid to join the solid than a closer packed plane, such as {111}. The nanoparticle consists of a cubic crystalline poly-type of silicon carbide (3C-SiC) with a lattice parameter of 4.36 Å. In total, 3,900,208 atoms are generated. The MD simulation details are provided in [15, 16].

**Experimental Results**

Figure 2 shows the optical microscopy results in the middle part of the cast ingots [17]. As shown in Figs. 2(a) and 2(b), the A356 samples without UST during solidification have coarser microstructures than the ones with UST. Applying UST during solidification can break up the dendritic structure and thus finer globular grain structure can be obtained. Figure 2(c) shows the microstructure of A356 reinforced with SiC nanoparticles with UST during solidification. They have similar microstructures with those shown in Fig. 2(b), which means adding nanoparticles will not change the microstructure significantly, and the UST is the main cause of grain refinement. Thus, by applying the UST during the solidification (Figs. 2(b) and 2(c)) the matrix microstructure is refined and modified from a dendritic to a globular grain morphology. This can be explained as follows: First, ultrasound will increase the nucleation potential in the melt during solidification. This is because UST will cause the temperature and pressure characteristics of the melt to change periodically at high frequencies. As a result, the local number of nuclei into the melt are increased. Moreover, strong convection produced by UST will promote the diffusion of solute, which will also increase the number of nuclei into the melt. The second reason is because shock waves produced by ultrasonic cavitation will break the dendrite tips during solidification, which will contribute to the microstructure refinement and modification [5].

Figure 3 shows the SEM microstructures at high magnification of the A356 sample reinforced with 1.0 mass % SiC and UST during solidification [17]. Some nanoparticles in Fig. 3(b) were found in the grain boundary area. Since the particles are in nanometer size, the EDS accuracy is not high enough to identify the particle chemical composition, therefore Fig. 3(c) only shows the EDS mapping results at relatively low magnification. In the mapping photo, the red color represents C (carbon), the green color represents Al (aluminum). The EDS results indicate that some of C is located in the grain boundary area. By combining the EDS results with the SEM photo, it can be determined that some of the SiC nanoparticles are gathered around the grain boundary area.
Figure 2. Optical micrographs in the middle part of the cast samples: (a) A356 without UST; (b) A356 with UST during solidification and (c) A356 sample reinforced with 1.0 mass% SiC and UST during solidification.

Figure 3. SEM results and EDS mapping photo of the sample processed via UST+1.0 mass% SiC. (a) SEM picture at X500 and (b) SEM picture of the red rectangular area in (a) at X50000 and (c) EDS mapping results at X40.

Simulation Results

The two-phase Al system is established by holding the atoms in solid Al region of the original crystalline lattice and melting the liquid Al region by raising the temperature above the melting point. The whole system is equilibrated after 40ps, then the initial solid Al atoms are fixed at some temperature (500 K) below the melting point, and Si, C, and liquid Al atoms which will release heat through the solid atoms. Figure 4 shows the solid/liquid status of the system with a 10 nm SiC nanoparticle after 100 ps, 200 ps, 400 ps, and 600 ps, respectively [15, 16]. As it can be seen from Fig. 4(a), when the liquid/solid interfaces are far away from the SiC nanoparticle, both of the interfaces at the top and bottom are flat. When the interface at the bottom approaches the SiC nanoparticle, a trough is formed below the particle (see Fig. 4(b)). As time goes on, the interface at the bottom passes through the particle and the interface at the top approaches the particle (See Fig. 4(c)). Finally, these two interfaces meet each other, become one, and disappear (see Fig. 4(d)).

Figure 4. Solid/liquid interface evolution of the system with a 10 nm SiC nanoparticle.

Figure 5 shows the position of the 10 nm SiC nanoparticle with time [15, 16]. As it can be seen from Fig. 5, the horizontal movement (x and y) of the nanoparticle is insignificant. But in z direction (vertical), the particle moves down
towards the liquid/solid interface at the bottom in the beginning, and when the interface from the top approaches it, the particle moves up towards that interface again. This MD simulation confirmed that during the solidification process, the SiC nanoparticle will be engulfed by the solidification front instead of being pushed, as predicted by previous PEP models [18]. Similar conclusion was found for the solidification domain with a 20 nm SiC nanoparticle [15, 16].

Figure 5. Position evolution of a 10 nm SiC nanoparticle during solidification.

Conclusions

It was determined from this study that the application of UST during solidification plays a key role in the refinement and modification of the microstructure of the A356 cast ingots. Without UST processing, dendritic structures were obtained. By using UST during solidification, fine globular grain structures were obtained. The addition of SiC nanoparticles affects the formation of the microstructure of the A356 cast ingots only marginally. The SEM and TEM results indicated that the added SiC nanoparticles are dispersed reasonably well into the A356 matrix. However, some SiC nanoparticles are gather around the grain boundary area.

The complex interactions between a SiC nanoparticle and the Al liquid/solid interface were analyzed with a 3D molecular dynamics model using LAMMPS on the High Performance Computing Cluster (HPCC) at the University of Alabama. Molecular dynamics simulation results showed that SiC nanoparticles with diameters less than 20 nm will be engulfed by the solidification front instead of being pushed, which is beneficial for nanodispersion.

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