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Three-dimensional analysis of particle coarsening in high volume fraction solid–liquid mixtures

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Abstract

The three-dimensional microstructure of 78 and 52 vol.% Sn-rich particles coarsened within a liquid Pb–Sn matrix were determined by the reconstruction of serial sections. The three-dimensional particle size distribution (PSD) and the particle–particle contact distributions were determined. The three-dimensional PSDs do not match those predicted by particle coarsening theory, but there is reasonable agreement with the grain size distributions predicted by a grain growth simulation. In addition, when the particle–particle contact distribution is normalized to the average number of particle contacts, the distribution is statistically invariant with volume fraction. At 52 vol.% it is found that the number of contacts is proportional to the square of the average particle size, but this is not true for 78 vol.%. This is attributed to the increased shape distortions of the particles that are present in the higher volume fraction samples.

Keywords: Coarsening; Grain growth; Image analysis

1. Introduction

Ostwald ripening occurs in multi-phase materials and is a diffusional process in which a system lowers its total energy by reducing the total interfacial area. In a two-phase system, large particles grow at the expense of smaller particles. As the system coarsens, the average particle size increases, while the total volume fraction of particles remains constant. Thus, there is a reduction in the average particle density within the system. Ostwald ripening occurs in many two-phase mixtures and can have a major effect on the properties of a material. For example, coarsening in precipitation-hardened alloys has a significant effect on materials properties, since the decrease in particle density leads to a degradation of the mechanical strength of the alloy.

Initial investigations into the kinetics of Ostwald ripening of particles within a matrix were performed by Greenwood [1], where the growth rates of individual particles within a dispersion of particles were determined theoretically, assuming that the particles are infinitely separated from each other. The results compared qualitatively well to experimental results, but the theoretical treatment did not address the evolution of the size distribution of particles.

The original description of the kinetics of Ostwald ripening of a distribution of particles was proposed by Lifshitz and Slyozov [2] and Wagner [3]. They examined the coarsening of infinitely separated, spherical particles. In the limit where $t \to \infty$, a similarity solution was found, wherein the microstructure should become self-similar when scaled by the average particle size and the average particle size increased with time as $t^{1/3}$.

While this theory captured some more important underlying physics of the coarsening process, it suffered from an unrealistic assumption of a zero volume fraction of the coarsening phase. Much of the later theoretical work has examined the effects of interparticle diffusional interactions that occur in systems with a finite volume fraction [4–13].

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The difficulty in developing such a theory lies in the need to account for the overlapping diffusion fields surrounding the particles. Because of this, most theories assume that the particles are spherical and noncontacting, which limits most theories to small volume fractions ($\lesssim 0.3$), with the notable exception of the effective medium theories [8,9,14].

Experimental aspects of coarsening in the solid–liquid system have been limited as well. The characterization of the microstructures for comparison to theory has been mainly confined to high volume factions of coarsening phase using two-dimensional (2D) sections [15–20]. However, recent experiments [21–23] performed as part of the coarsening in solid liquid mixtures (CSLM) project, a microgravity experiment performed on the space shuttle on missions STS 83 and STS 94, investigated the kinetics and microstructural evolution of Sn-rich particles dispersed in a Pb–Sn eutectic liquid matrix over a wide range of volume fractions. At low volume fractions a reasonable agreement between coarsening simulations, but not coarsening theory, and experiment was obtained. The coarsening models make predictions for three-dimensional (3D) systems, thus the comparison between theory and the data taken from 2D plane sections requires that the 3D predictions of theory be recast to make predictions of the microstructure that can be measured on 2D plane sections [24]. This was done assuming that the particles are spherical, an approximation that is valid only at low volume fractions where there are few particle–particle contacts.

In contrast, at higher volume fractions there was a significant disagreement between the predictions of mean field coarsening theory and the results from the experiment [22,23]. These discrepancies were mainly attributed to the increased number of particle contacts that occur at a high volume fraction of particles. However, the coordination number, C, defined as the number of particle–particle contacts per particle, could not be determined from 2D sections. Furthermore, the coordination number has been identified as an important microstructural parameter for microstructures with a high volume fraction of particles. For example, the nature of the particle contacts has been shown to affect the thermal expansion [25] and the mechanical properties of liquid phase sintered materials [26]. In addition, the number of grain faces in single-phase polycrystalline materials is also an important microstructural parameter for understanding the microstructural evolution [27,28]. One can measure the number of contacts per particle using a 2D plane section, but in order to convert this information to the 3D coordination number, one must assume that the particles are spherical and either monodisperse [29], or make assumptions about the dihedral angle and the 3D particle size distribution [30,31]. Thus, only by obtaining 3D data, can one avoid the many assumptions and approximations inherent in interpreting measurements made on 2D sections and achieve a more direct comparison to theory.

Within the last 10 years, there has been a dramatic increase in the amount of 3D data published in materials science investigations [27,32–38]. This has been due mostly to advances in computational power and software development, but also due to new experimental techniques for collecting 3D data. Several of these investigations have concentrated on microstructural evolution during grain growth [27,36]. Liu et al. investigated the 3D grain structure of an austenitic steel alloy using serial sections, with more than 1000 grains sampled. They found a near-linear relationship between the number of faces per grain and the spherically equivalent radius of the particle. Krill and Chen [39] analyzed a large volume of grains in Al–Sn system using X-ray microtomography. They found reasonable agreement with phase-field grain growth simulations, but poor agreement with more common 3D Poisson–Voronoi tessellation, which is often used to describe grain structures. Grain growth is similar to particle coarsening in that the overall energy of the system is lowered by reducing the total interfacial area, and it can be viewed as the limit of 100 vol.% coarsening phase. However, grain growth occurs in a single phase system, and the kinetics of the process are controlled by diffusion of atoms across a grain boundary, giving rise to a different temporal exponent of 1/2 for the average grain size, and to a different steady-state grain-size distribution.

To the best of our knowledge, there has been only one other investigation of the 3D microstructure of a large set of particles dispersed within a liquid matrix. Tewari et al. [37] used serial-sectioning to analyze the 3D microstructure of a liquid-phase sintered 83 wt.% W–Ni–Fe alloy under varying gravitational conditions. They measured both the particle size distribution and the particle–particle contact distribution for samples sintered in normal gravitational conditions and sintered in a microgravity environment. By montaging many 2D images into one continuous section, they could create a large 3D volume. However, because the height of the analyzed volume was relatively small compared to the particle diameter, many of the particles intersected the outside walls of the analyzed volume, which significantly limited the total number of particles that could be fully analyzed. Interestingly, they found that, when normalized to the average number of contacts per particle, the contact distribution was invariant with the gravitational environment present during the sintering process. In addition, it was found that the number of contacts per particle was proportional to the square of equivalent particle radius.

In this investigation, we will present the analysis of the 3D structure of coarsened Sn-rich particles dispersed in a Pb–Sn eutectic matrix over a range of volume fractions. Sedimentation of the particles at lower volume fractions is avoided by processing the samples in a microgravity environment, as part of the NASA CSLM project. A method of efficient serial sectioning is used to collect a statistically significant number of particles (>1000) for each volume fraction. Using these sections, the 3D microstructure is reconstructed and the particle sizes and number of contacts per particle are measured. Furthermore, using