49th AIAA Aerospace Sciences Meeting including the New Horizons Forum and Aerospace Exposition
4 - 7 January 2011, Orlando, Florida

AIAA 2011-1346

Coarsening in Solid-Liquid Mixtures: Effect of Microgravity Accelerations on Particle Sedimentation

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Coarsening is the growth of larger particles at the expense of smaller ones in a two-phase system of varying particle sizes. Solid-liquid mixtures are ideal systems with which to study the process of coarsening. When the experiments are conducted on Earth, gravity leads to particle sedimentation. Conducting the experiments in a zero-gravity environment would eliminate the particle sedimentation. A series of samples consisting of solid tin particles embedded in a tin-lead liquid matrix were coarsened at the Microgravity Glove box at the International Space Station (ISS). The experiment of interest was conducted in June 2010. The effects of the residual microgravity acceleration on particle sedimentation are investigated. It is observed that there is significant sedimentation, compared to the average particle size, as a result of the residual microgravity acceleration. We also find that the mass transport during coarsening was diffusion driven despite the sedimentation.

Nomenclature

V	Velocity, m/s
g	Gravitational Acceleration, g
R	Particle Radius, μm
P	Density, Kg/m^3
η	Kinematic Viscosity , Kg/(m·s)
Pe	Peclet Number
t	Time, s
D	Mass Diffusion Coefficient, m^2/s^2

Subscript

avg	average
min	minimum
max	maximum
S	Tin, Sn
L	Lead, Pb

I. Introduction

Given a two-phase system with varying particle sizes, the larger particles tend to grow at the expense of the smaller ones. This process, called coarsening or Ostwald ripening, is a way for the system to minimize its energy by decreasing the total interfacial area through mass transport. As a result, the average particle size within the system increases over time. Coarsening is observed during processing of metal alloys and therefore is a common problem when these alloys are used in applications that require elevated temperatures. The theory of coarsening was established by Lifshitz and Slyozov,[2] and Wagner[3] in 1960s. The theory predicts the coarsening rate of the system in the limit of infinite coarsening time. In this limit the system attains a steady state wherein the kinetics are defined by temporal power laws and time independent scaled particle size distributions.

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Solid-liquid mixtures are ideal systems for tests of coarsening theory. For examples, the interfacial energy is isotropic and the rapid diffusion through the liquid phase assures that there will be significant coarsening in reasonable experimental time. However, experiments involving solid-liquid systems usually suffer from a density difference between the solid and the liquid. Accordingly, the gravity of the Earth causes the Sn particles to sediment to the top of the sample in a short time. Since testing the theory of coarsening calls for longer experiments, it is not possible to conduct them on Earth. A ground processed sample is shown in Figure 1(a), where the sedimentation of the particles to the top of the sample is clearly visible.

The ideal environment for such an experiment is a zero-gravity environment, under which the particle sedimentation would disappear and the solid particles would be evenly distributed in the liquid until the steady state is reached. The Microgravity Glove Box at the International Space Station (ISS) provides a near-zero gravity environment, minimizing the effect of gravity on the system and therefore making testing the theory possible. However, there are small residual accelerations acting on the system. In order to support the scientific experiments and to better determine the experimental conditions, the International Space Station is equipped with the Microgravity Acceleration Measurement System (MAMS). MAMS measures residual microgravity accelerations that occur as a result conditions such as general experiment operation, crew activity and rotational effects.[1] Being a bigger force, the effects of Earth's gravity on the experiments is obvious. On the other hand, the effects of this residual microgravity acceleration on the coarsening experiments and if they lead to particle sedimentation is unknown.

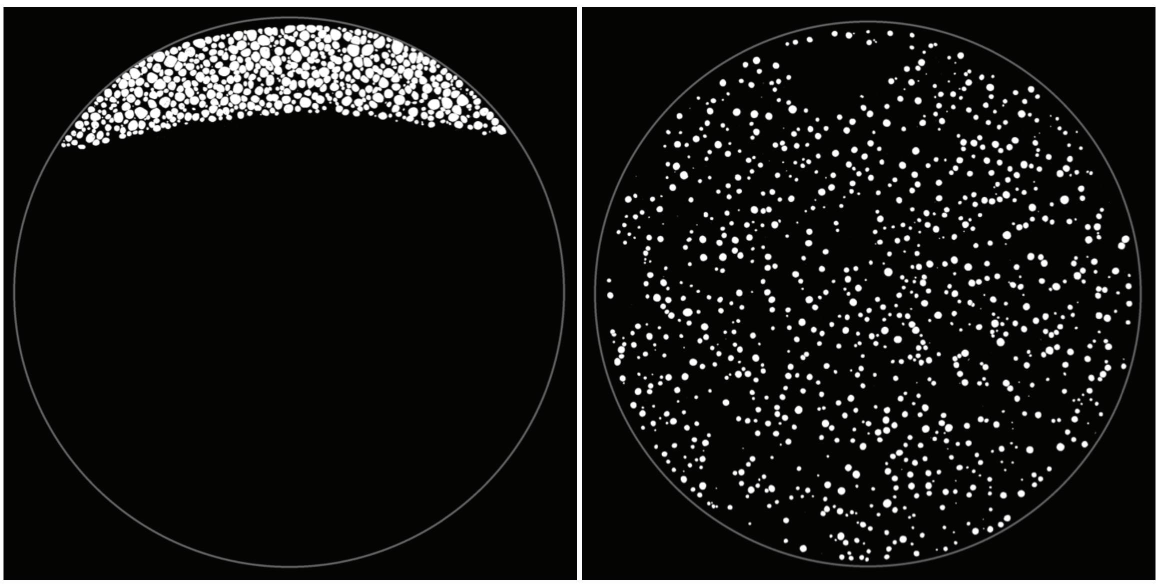


Figure 1. Effect of gravity on particle sedimentation: Sample with 10% solid Sn-phase (white) held for 10 hours and 10 minutes at 185°C in a liquid (black), (a) on ground, (b) at the Microgravity Glove box at ISS during CSLM I. The Sn-particles are less dense than the liquid, so they sediment to the top of the sample.

II. Experimental Procedure

Pb-Sn alloys are well studied, mostly due to their low melting point (183°C), isotropic interfacial energy and rapid coarsening rates, making it ideal for coarsening studies. The samples were prepared using a method previously used by Hardy and Voorhees.[4] The samples consisted of solid, almost spherical Sn particles embedded in a Pb-Sn eutectic matrix. A number of experiments were conducted at the Microgravity Glove Box at the ISS. The particular experiment analyzed was from June 2010, sample processing unit 5, which had a length of approximately 34 hours, and was from GMT174/17.45 to GMT 176/04.26. In the microgravity environment, the samples were first heated up to solid-liquid regime. They were then held at 185°C, 2°C above the eutectic temperature, for varying coarsening times and quenched. An ISS processed sample is shown in Figure 1(b). Some of the results were discussed in an earlier paper by Kammer et. al.[5]

The residual microgravity acceleration data from MAMS were studied for the sample with 30% volume fraction of solid. Using the mapped acceleration data for this experimentation time from Erik Kelly at Zinn Technologies at the Microgravity Glove Box location, and interpolation to bridge data gaps during times where there was a loss of signal we computed the sedimentation of the particles in the system.

Using the mapped MAMS acceleration data, the Stoke's sedimentation velocity can be determined at each time step using,

$$V = 2gR^2(P_S - P_L)/9\eta. \tag{1}$$

In equation 1, P_S, P_L and η are constants and their values summarized in Table 1. Results from CSLM-II

Table 1. Values of the Constants

Variable	Value
P_S	7088 Kg/m^3
P_L	8074 Kg/m^3
η	0.00265 $Kg/(m \cdot s)$
D	$6.2 * 10^{-10}$ m^2/s

had shown $t^{1/3}$ dependence of R.[5] Furthermore, the particle size distribution is not monodisperse. Given the strong dependence of the sedimentation rate on particle radius, we examine the velocities and displacements for representative particle radii in the distribution. These relations are summarized by equations 2. Using the relations shown in equation 2 along with V, the displacement was calculated in X, Y and Z directions separately, assuming, for each, minimum particle size, average particle size and maximum particle size.

$$\begin{aligned} R_{avg} &= 1.34 * 1.387 * t^{1/3} \\ R_{min} &= 0.1 * R_{avg} \\ R_{max} &= 1.6 * R_{avg} \end{aligned} \tag{2}$$

The total displacement was determined by integrating 1.

III. Results

Figure 2 shows the residual microgravity acceleration with respect to time. The accelerations vary strongly with time and are less than 1 μg most of the time. The resulting displacements of each particle with respect to time are also shown. It is observed that while the body of particles tend to remain stable with respect to the acceleration in X and Y directions, they do have a relatively significant displacement resulting from the acceleration in the Z direction. Despite the small accelerations, over the length of the experiment significant particle sedimentation is possible. The particle displacement is smooth compared to the acceleration data because the duration of these larger accelerations is very small and thus induces nearly no change in the particle location.

These findings are also supported by the results shown in Figures 3 and 4. Figure 3 shows the magnitude of velocity at each time step for R_{avg}, R_{min} and R_{max}, in blue, green and red respectively. The smaller particles, shown using the green line, tend to be more stable than the bigger particles, with a velocity close to 0. The overall velocity is increasing over time and although it is very small, is on the order of $10^{-8} m/s$. This velocity still gives rise to a relatively significant displacement, shown in Figure 4.

Figure 4 shows the magnitude of displacement at each time step for R_{avg}, R_{min} and R_{max}, again, in blue, green and red respectively. This plot supports the observation that the smaller particles remain stable, with almost no change in overall displacement, while the larger particles tend to move within the matrix. The overall displacement values are on the order of $10^{-4}m$. Given that the average particle radius is around 85 microns, the displacement is significant with respect to the particle sizes. Thus, these larger particles sediment through a comparatively stationary array of smaller particles.

These findings raises the question of whether the mass transport during coarsening is via diffusion or convection. The dimensionless Peclet number can be calculated using equation 3. The value of the constant D was given in Table 1.

$$Pe = VR/D \tag{3}$$

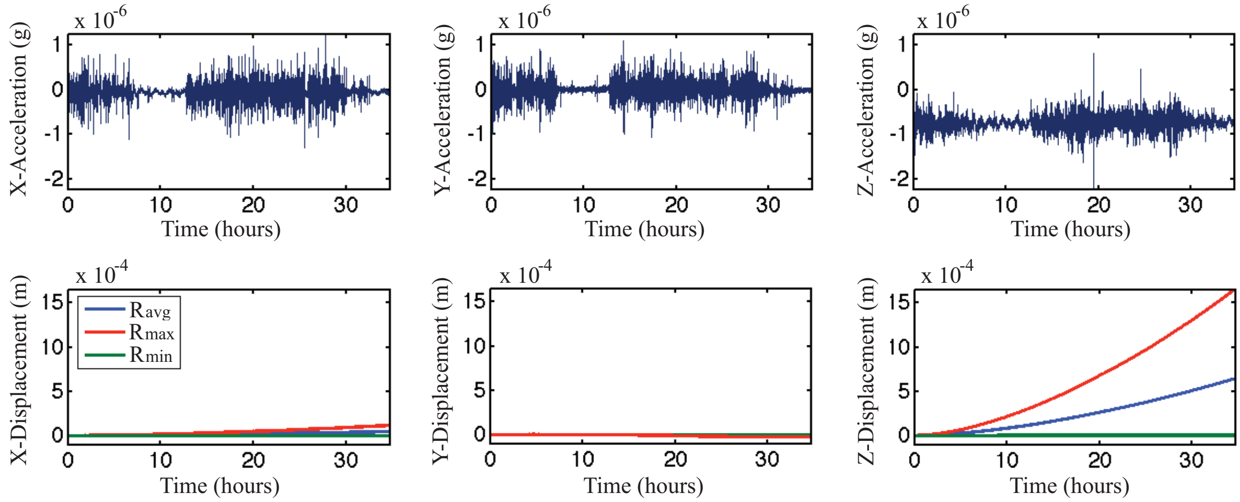


Figure 2. Residual acceleration data for the sample coarsened for 34 hours, plotted for X, Y and Z directions, shown along with the displacement calculated using equation ??.

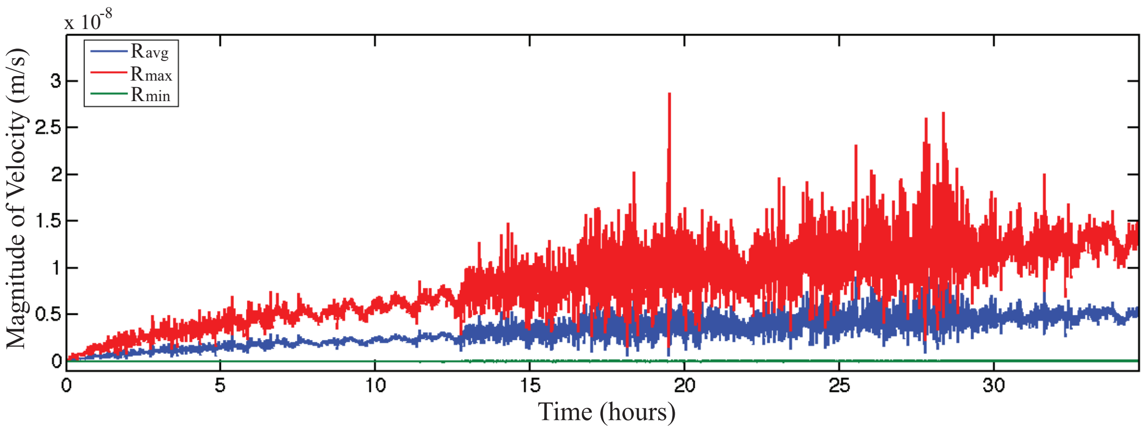


Figure 3. Magnitude of Stoke's velocity with respect to time

A smaller Peclet number points to a diffusion driven mass transportation while a larger one signifies a convection dominated mass transport process.[6] Although showing a slightly different trend for R_{avg}, R_{min} and R_{max}, the Peclet numbers calculated from this data are on the order of 10^{-3}, and are therefore small. It can be concluded that although the residual microgravity acceleration does effect the displacement of the larger particles, the mass transport during coarsening remains diffusion driven.

IV. Summary

A 30% volume fraction Pb-Sn sample was coarsened in the Microgravity Glove Box on the International Space Station for 34 hours. During the experiment, the residual microgravity acceleration data was also recorded using Microgravity Acceleration Measurement System. It was found that the residual microgravity acceleration had significant effect on the location of larger particles, but the coarsening process remained diffusion driven.

Acknowledgments

We are grateful for the financial support of NASA, contract NNX10AV36G. We thank Walter Duval from NASA Glenn and Tibor Lorik from Zinn Technologies for their help in planning and performing the

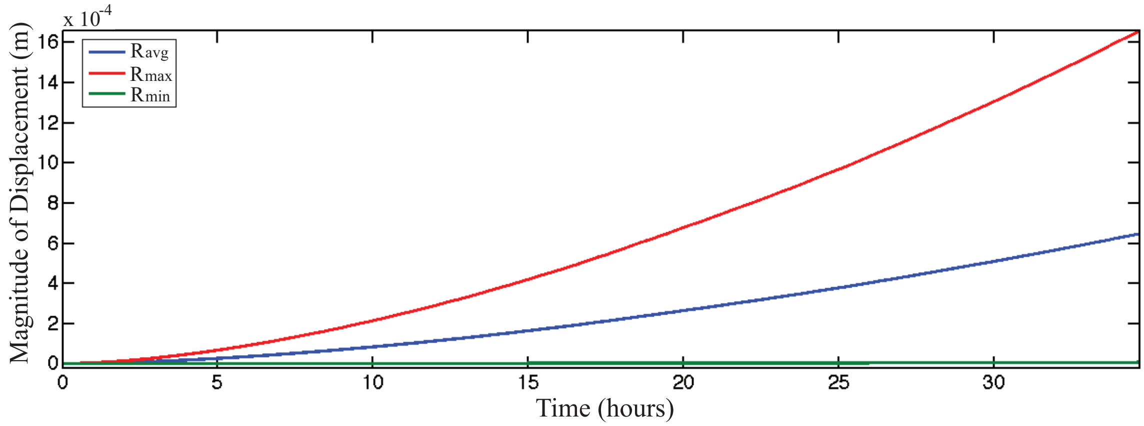
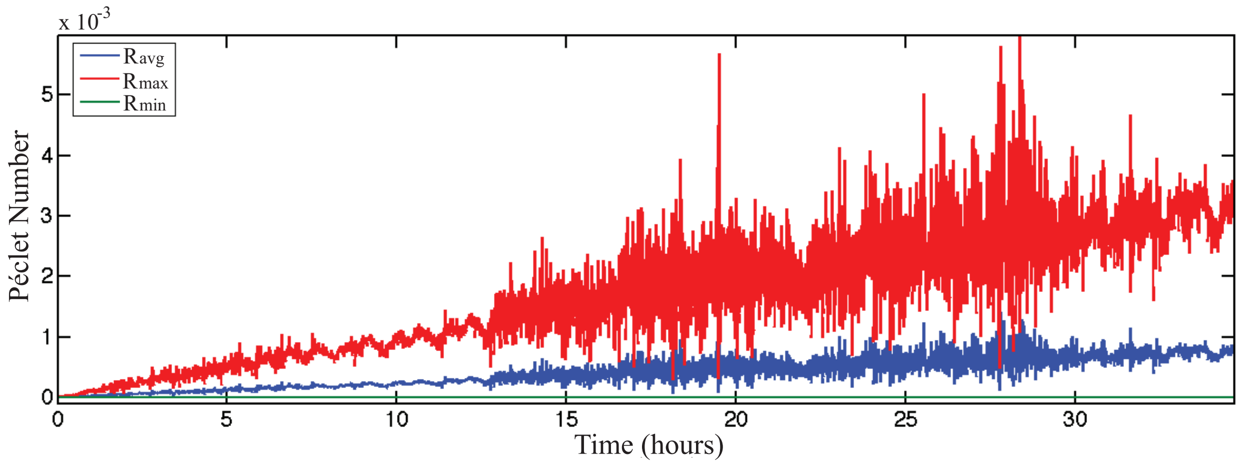


Figure 4. Magnitude of displacement with respect to time



experiments.

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