

Containerless Measurements of Density and Viscosity of Fe-Co Alloys

Jonghyun Lee¹, Paul Choufani¹, Richard C. Bradshaw², Robert W. Hyers³, Douglas M. Matson¹

¹Tufts University;

Department of Mechanical Engineering;

200 College Avenue; Medford, MA 02155, USA

²Bradshaw Consulting

108 Harnden Avenue; Watertown, MA 02453, USA

³University of Massachusetts;

Department of Mechanical Engineering;

160 Governors Drive; Amherst, MA 01003, USA

Keywords: Fe-Co Alloys, Density, Viscosity, Containerless measurements, Double recalescence

Abstract

During the past years, extensive collaborative research has been done to understand phase selection in undercooled metals using novel containerless processing techniques such as electrostatic and electromagnetic levitation. Of major interest is controlling a two-step solidification process, double recalescence, in which the metastable phase forms first and then transforms to the stable phase after a certain delay time. The previous research has shown that the delay time is greatly influenced by the internal convection velocity. In the prediction of internal flow, the fidelity of the results depends on the accuracy of the material properties. This research focuses on the measurements of density and viscosity of Fe-Co alloys which will be used for the fluid simulations whose results will support upcoming International Space Station flight experiments.

Introduction

Molten metallic alloys can be undercooled if the preferred sites for nucleation such as crucibles and impurities are effectively removed. One effective way of achieving undercooling is containerless processing. When the undercooled liquid solidifies, the heat of fusion is rejected and the temperature increases suddenly. This phenomenon is referred to as recalescence. The solidification of undercooled alloys can occur in two ways: either the stable phase nucleates directly from the liquid (single recalescence) or the metastable phase forms first followed by the growth of the stable phase into the mushy zone (double recalescence). The metallography reveals that alloys which have undergone double recalescence show fine and globular grains, which can result in better mechanical properties. Since the growth rate of the metastable phase is lower than that of the stable phase, it is important to attain a certain amount of delay time between the

nucleation events. If there is enough delay time, most part of the alloy will double recalesce, otherwise, the stable phase begins to grow and overtakes the metastable phase solidification front as shown in Figure 1 [1]. Previous research has shown that the delay time is greatly affected by the internal convection in the molten alloy. If the convective velocity is large, the primary arms of dendrites can be bent by the drag forces and eventually collide with each other. The collision between neighboring dendrite arms can serve as a preferred site for the unwanted early nucleation of stable phase [2]. Therefore characterization of fluid flow in the molten alloys is important in understanding the phase selection during solidification.

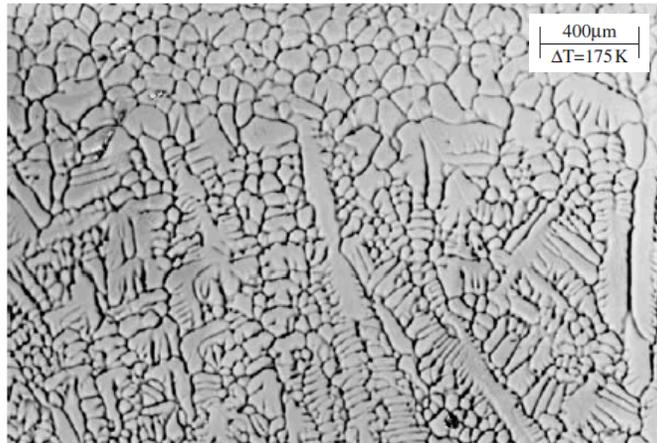


Figure 1. Microstructure of steel alloy: The solidification proceeded from top to bottom. The upper portion of the figure underwent double recalescence and thus shows globular and fine microstructures. As the solidification proceeded, the stable phase overtook the metastable solidification resulting in the dendritic structure shown in the lower portion of the figure. This corresponds to single recalescence behavior.

In the near future, a series of solidification experiments will be performed in space over a wide range of internal convective velocities to acquire data for comprehensive understanding of solidification processes. For the success of the International Space Station (ISS) mission, the identification of the correlation between test parameters and internal fluid flow is of great importance.

Hyers *et al* [3] have developed mathematical models which describe the convection inside the droplet while processed in both electromagnetic (EML) and electrostatic (ESL) levitators. These models can be solved using appropriate numerical techniques. To obtain satisfactory numerical results, it is important to secure reliable material properties. Using containerless measurement techniques [4], material properties of Fe-Co alloys, one of the candidate alloys for the ISS mission, are measured. These values will be used for the computational fluid dynamics simulations in support for the ISS mission.

Experimental Apparatus

The experiment was performed with the electrostatic levitation (ESL) facility at NASA Marshall Space Flight Center (MSFC) in Huntsville, AL. The weight of the sample was cancelled by Coulomb forces exerted by a pair of vertical electrodes. Two additional pairs of electrodes on the horizontal plane were used to stabilize the sample using the PID (Proportional, Integral, and Derivative) control scheme. After levitation and stabilization, the sample was heated by a 200 W YAG laser and the temperature was monitored by optical pyrometers (Mikron Infrared Inc.) which detects the wavelength of emitted light from the sample. The processing was achieved in a high vacuum chamber ($\approx 10^{-8}$ torr) to avoid chemical reactions. Figure 2 shows a schematic of ESL apparatus in NASA MSFC.

During the processing, images of the sample were captured by a high speed camera (Redlake Motion Pro 1000) and analyzed to obtain measurement of density and viscosity as a function of temperature. For analysis of images, the edges of the sample in the images were detected and fitted with sixth order Legendre polynomials for the calculations of projected areas and volumes of samples. The details of image processing techniques can be found in [5].

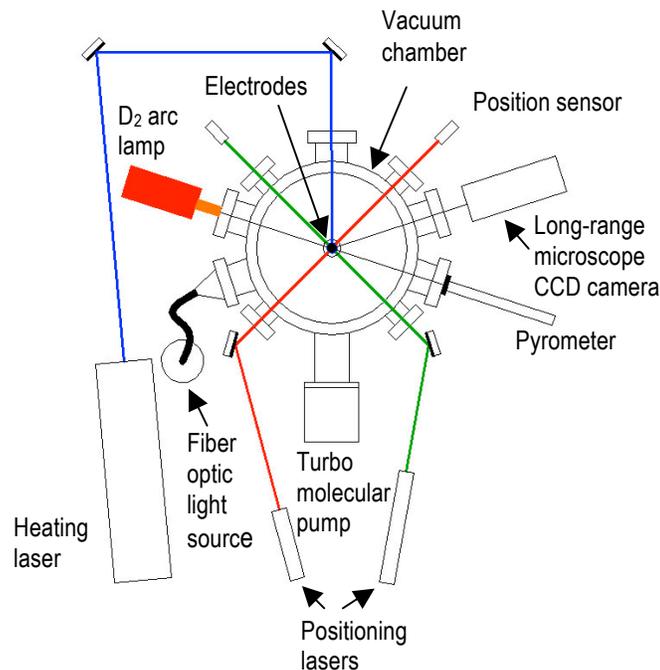


Figure 2. A schematic of ESL apparatus in NASA MSFC.

Measurement of Density

To estimate the density of the sample, it is essential to know the volume and mass of the sample for each time step in a thermal cycle. As the molten sample free cooled video images were acquired at a rate of 25 Hz and edge-detection software was used to tract sample shape. While the volume can be easily obtained by integrating the fitted functions (6th order Legendre polynomials), a care must be taken in dealing with the mass. Throughout the density test, the reduction in mass due to evaporation was observed. Adopting the Langmuir's equation for evaporation, the loss of mass for a given time was calculated.

$$\frac{\dot{m}}{A_s} = \frac{\alpha(p_{\text{vapor}} - p_{\text{atm}})}{\sqrt{2\pi kT}} \quad (1)$$

where \dot{m} is the rate of loss of mass due to evaporation, A_s is the surface area, p_{vapor} is the vapor pressure, p_{atm} is the partial pressure of the atmosphere, α is the correction factor to account for the mass transport diffusion layer resistance, k is the gas constant, and T is the absolute temperature. Having the volume and mass as functions of time, the density of the sample could be estimated as a function of time. Combining the estimated density and temperature as functions of time t , density-temperature plots were generated.

$$\rho(T(t)) = \frac{m(T(t))}{\text{volume}(T(t))} \quad (2)$$

Measurement of Viscosity

Since the viscosity acts as a resistance to fluid motion, the decay time for an oscillating sample can be used to measure the viscosity. The sample was excited by applying periodic changes in the vertical electrostatic field. The excitation started with the lower frequencies and was gradually increased until the sample showed noticeable oscillations; the excitation frequency was near the sample natural frequency ($\approx 200\text{Hz}$). Once the desired level deformation is achieved, the excitation was stopped and the sample was allowed to dampen out freely. The sample oscillations were captured at the rate of 1000 Hz.

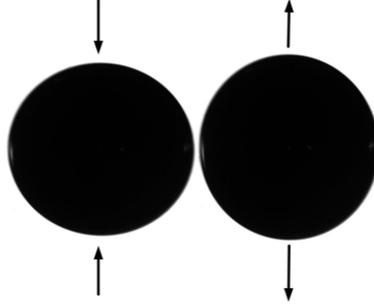


Figure 3. Shapes of an oscillating sample.

By analyzing the captured images, the areas of the oscillating sample were calculated for each frame. After the area of the oscillating sample was plotted in the time domain, the Fast Fourier Transform was applied to extract the natural frequency of the sample and then a sinusoidal curve as expressed in Equation (3) was fitted to the decaying portion of the amplitude $y(t)$.

$$y(t) = \cos(\omega t + \phi) \exp\left(\frac{t}{\tau}\right) \quad (3)$$

where ω is the natural frequency, ϕ is the phase angle, and τ is the damping coefficient which characterizes how fast the sample dampens out. The viscosity can be determined using its relationship with the damping coefficient as below.

$$\mu = \frac{\rho R_0^2}{(l-1)(2l+1)} \cdot \frac{1}{\tau} \quad (4)$$

where ρ is the density, R_0 is the radius of the undeformed droplet, l is the mode of oscillation ($l=2$).

Results and Discussion

The density of $\text{Fe}_{45}\text{Co}_{55}$ was measured as a function of temperature and plotted in Figure 4. As indicated above, during the experiment 6.86% of mass was lost due to the mass evaporation. According to the temperature-time plot, the sample was melted and superheated up to $T_m+130\text{K}$ for 66 seconds and cooled for 6 seconds during which the density was measured. Considering the fact that the time duration for the density measurement is only 9% of that for melting and superheating, it is reasonable to assume that the majority of mass evaporation had occurred before the measurement of density. Thus, the post-mass was used for calculation of density and this technique should result in a slight underestimation of density results. For comparison, the red line in the graph indicates the predicted values for density using rule of mixtures (ROM). For the

prediction, the density and the rate of change in density with respect to temperature ($\partial\rho/\partial T$) of Fe and Co shown were taken from the literature [6] and these values are shown in Table 1.

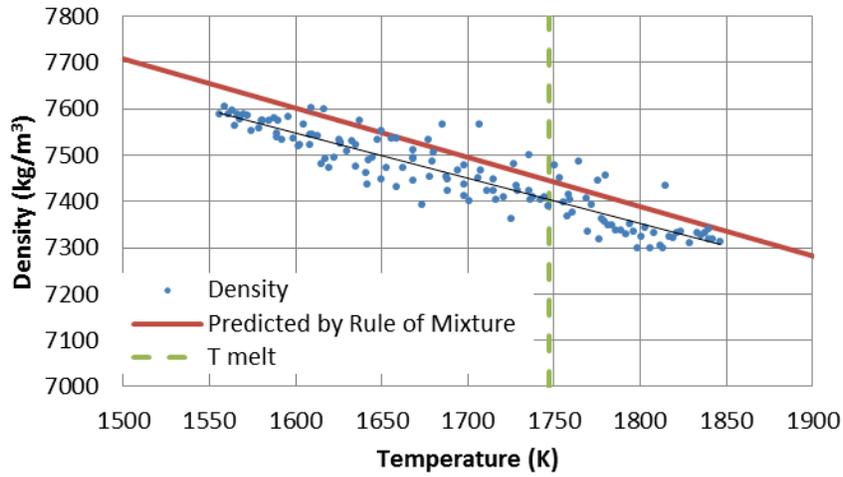


Figure 4. Variation of density as a function of temperature. The black line is a least-squares line fit to the measurements.

Table 1. Densities and the slopes of density with respect to temperature of Fe, Co, and Fe₄₅Co₅₅.

	Fe	Co	Fe ₄₅ Co ₅₅
T_m (K)	1808	1765	1747
ρ at T_m (kg/m ³)	7030	7750	7458
$-\partial\rho/\partial T$ (kg/m ³ ·K)	0.880	1090	0.996

The density at the melting point was estimated to be 7405 kg/m³ and it was slightly greater than the predicted value by 0.7%. The rate of change in density with respect to the temperature at the melting point was estimated to be -0.9741 kg/m³·K and the difference from ROM predicted slope was thus 2.1%. It can be stated that the experimental results and ROM predictions agree well with each other and.

Two samples of Fe₄₅Co₅₅ were used for the measurements of viscosity. Figure 4 shows the viscosity of Fe₄₅Co₅₅. The viscosity data was fitted with an Arrhenius relation.

$$\mu = \mu_0 \exp\left(\frac{E}{k_b T}\right) \quad (5)$$

where E is the activation energy, k_b is the Boltzmann constant (8.61730×10^{-5} eV/molecule-K). The fitted values are $\mu_0 = 4.912 \times 10^{-6}$ Pa·s and $E = 1.24891$ eV/molecule. The viscosity at the melting point was estimated to be 0.0197 Pa·s.

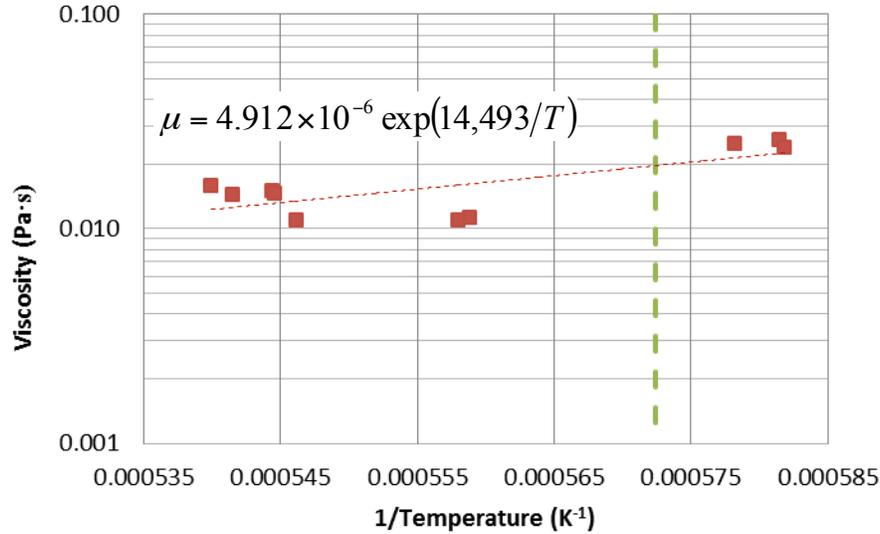


Figure 5. Variation of viscosity as a function of temperature.

Summary

As support and preparation for ISS mission, the density and viscosity of $\text{Fe}_{45}\text{Co}_{55}$ were measured with the novel containerless processing technique. The measured density and its rate of change with respect to the temperature at the melting point ($\partial\rho/\partial T$) were $7,405 \text{ kg/m}^3$ and $-0.9741 \text{ kg/m}^3\cdot\text{K}$ and these values show an excellent agreement with the predicted values by the rule of mixtures and their differences were to be 0.7% and 2.1% respectively. The viscosity of $\text{Fe}_{45}\text{Co}_{55}$ was measured and fitted with the Arrhenius relation as $\mu = 4.912 \times 10^{-6} \exp(14,493/T)$. The measured density and viscosity values will be used for the simulations of convection inside the droplets.

Acknowledgements

This project is sponsored by NASA under grants NNX08AL21G and NNX10AV27G. Experimental portions of this research were performed at NASA Marshall Space Flight Center. The authors wish to thank Trudy Allen and Glenn Fountain for extensive technical advice.

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