Experiments using a Ground-Based Electrostatic Levitator and Numerical Modeling of Melt Convection for the Iron-Cobalt System in Support of Space Experiments

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Materials research is being conducted using an electromagnetic levitator installed in the International Space Station. Various metallic alloys were tested to elucidate unknown links among the structures, processes, and properties. To accomplish the mission of these space experiments, several ground-based activities have been carried out. This paper presents some of our ground-based supporting experiments and numerical modeling efforts. Mass evaporation of Fe₅₀Co₅₀, one of the flight compositions, was predicted numerically and validated by the tests using an electrostatic levitator (ESL). The density of various compositions within the Fe-Co system was measured with ESL. These results serve as reference data for the space experiments. The convection inside a electromagnetically-levitated droplet was also modeled to predict the flow status, shear rate, and convection velocity under various process parameters, which is essential information for designing and analyzing the space experiments of some flight compositions influenced by convection.

INTRODUCTION

Since 2015, several materials experiments have been performed using the materials science laboratory electromagnetic levitator (MSL-EML) aboard the International Space Station (ISS). More than 50 principal investigators from 13 different countries have been participating in this space program. Thermophysical properties, transport phenomena, and glass/quasicrystal formation of various alloys are being studied using MSL-EML. To increase the chance of success in the space experiments, the team has meticulously planned and prepared for their science experiments. The safety aspects, mass evaporation [1] and thermophysical properties [2-4] of each sample have been measured as part of the ground support program.

The measurement of mass evaporation is important. Evaporation often causes composition shifts of tested alloys resulting from preferential evaporation of volatile species. Such composition shifts can be reduced by limiting superheating and time for thermal hold during property measurements. On the other hand, evaporated materials may end up as aerosols, which can be hazardous to the health of the astronauts. Otherwise the evaporated materials will deposit on the optics, obscuring measurements, or on the coils. Excessive coil deposition can cause flaking of deposited materials, cross-contamination, short circuiting, and arcing. Each batch has 18 samples, and the amount of mass evaporation of each sample is different depending on the composition and thermal history. After discussion among scientists, the thermal cycles of each samples are adjusted such that the total amount of mass evaporation does not exceed a designated value.

For flight samples whose internal convection is of interest, numerical models were developed [5] and used [6]. The developed numerical models have been used to predict the convection status under an anticipated combination of process parameters. The status of convection affects phase selection during solidification of metal alloys which exhibit a two-step solidification behavior [7] and the formation of quasicrystals. This paper presents a few examples on how the ground-based experiments and numerical modeling were performed to support the current materials research in ISS.

EXPERIMENTAL

ESL facility at NASA Marshall Space Flight Center

Levitation or containerless processing represents an important tool in materials research. Levitated specimens are free from contact with a container, which permits studies of deeply undercooled melts, and high-temperature, highly reactive materials. Containerless processing provides data for studies of thermophysical properties, phase equilibria, metastable state formation, microstructure formation, undercooling, and nucleation.

The NASA Marshall Space Flight Center (MSFC) electrostatic levitation (ESL) laboratory uses non-contact measurement methods to provide materials property and processing data [8]. Typical properties provided are the temperature dependence of thermophysical properties (e.g. density, surface tension, and viscosity), obtained via electrostatic levitation. An additional advantage of this containerless method is that these measurements can be performed on a single sample. Processing melts in a container can impair access to metastable states and deep undercooling that resulted from contamination. Containerless methods such as electrostatic levitation represent a novel approach to high temperature materials property measurement as well as the study of deeply undercooled melts and metastable states. The technique can be used to process a wide variety of materials including metals, alloys, ceramics, glasses and semiconductors [9].

Electrostatic levitation uses Coulomb forces introduced by a pair of vertical electrodes to counteract the sample's weight. The sample's lateral position is stabilized by another two pairs of horizontal electrodes. The sample's vertical and lateral position is controlled through proportional–integral–derivative (PID) control of the electrostatic field. The processing atmosphere of the chamber is high vacuum, typically in the 10^{-6} torr range. Although the sample is levitated, a laser heats the samples and the temperature is monitored by optical pyrometers [10, 11]. The emissivity value is determined through the melt plateau (due to the latent heat of fusion). The emissivity input of the pyrometer is adjusted until the melt plateau is observed at the correct temperature. Additional capabilities of the MSFC ESL lab include non-contact creep strength measurement [12, 13], triggered nucleation [14], and a rapid quench system [15].

Measurement of mass evaporation

At high temperatures, a material becomes more reactive and a small amount of oxygen will affect the measured properties of molten metals, such as surface tension. In order to minimize the disturbance caused by impurities including oxygen, materials processing in MSL-EML is performed in a vacuum condition or in an inert gas such as helium or argon. For processing using an inert gas, the chamber pressure is to be maintained below ambient cabin pressure of ~400 mbar so that evaporated particles cannot escape and contaminate the cabin of ISS. At a combination of high temperatures and low pressures, mass evaporation may become significant. Especially in a vacuum condition, a significant amount of mass evaporates for materials with high vapor pressure. For instance, a near-spherically shaped Fe₅₀Co₅₀ alloy tested in an ESL vacuum chamber at its melting point (1808 K) for 60 seconds lost approximately 5.9% of its mass and the Fe concentration was preferentially reduced by 2.2 %p. The change in mass and composition indeed affects the measured thermophysical properties. Therefore, an accurate calculation of mass evaporation is of critical importance when we are dealing with materials with high vapor pressure. The composition shift must also be incorporated in the analysis for alloys whose constituents have considerably different evaporation behaviors.

The rate of mass evaporation of a pure substance in a vacuum condition can be calculated using Langmuir's equation [12].

$$\dot{m} = \frac{AP_v}{\sqrt{2\pi MRT}} \quad (1)$$

where $\dot{m}$ is the rate of mass evaporation, A is the surface area, P_v is the vapor pressure, M is the molecular weight of the material, R is the universal gas constant, and T is the temperature. The calculation of mass evaporation of a pure substance is easier compared to that of alloys. For alloys, three additional factors – activity, surface segregation, and composition shift - must be considered [1]. Eq. 1 can be extended to apply to the alloys. The rate of mass evaporation for a constituent i can be expressed as

$$\dot{m}_i = \frac{A_i a_i P_{v,i}}{\sqrt{2\pi M_i RT}} \quad (2)$$

where a_i is the activity of the constituent i on the surface of the sample. A_i is the surface area occupied by the constituent i that varies significantly depending on the difference in surface tension among constituents. The integration of $\dot{m}_i$ over time and summation for all constituents will give a total amount of mass evaporation for a given thermal cycle.

To support the processing of Fe₅₀Co₅₀ in ISS, the rate of mass evaporation of six Fe₅₀Co₅₀ samples was measured using the ESL at NASA MSFC. Each sample was put on a different thermal cycle, and the amount of mass evaporation was calculated using Eq. 2 as listed in Table 1.

Fe and Co wires with 4N and 3.5N purity, respectively, were cut into desired masses to form near-Fe₅₀Co₅₀ composition and were arc-melted in a vacuum chamber to form near-spherical specimens of 2-3 mm in diameter. After a sample was levitated and stabilized, a 200W Nd:YAG heating laser was applied to heat the sample. After the sample melted and reached around 100 °C of superheating, the heating laser was regulated such that the sample found its predetermined thermal hold temperature for various time durations (at ~T_m for 60 s, T_m for 90 s, T_m-73 K for 50 s, T_m+20 K for 70 s, T_m+53 K for 38 s, T_m+80 K for 85 s). After thermal hold, the heating laser was shut off and the sample cooled radiatively in the vacuum chamber. Over the course of processing, the surface temperature of the sample was measured by an optical pyrometer (IMPAC IGA 140 by Mikron Infrared Inc.) and recorded as a function of time. A high-speed camera (Y4-S1 by Integrated Design Tools) captured the projected area of the sample at a rate of 30 frames per second, and these video images were analyzed to measure the volume of the sample for each time step. The change in volume was used to calculate the density as a function of temperature considering mass evaporation. The details on the density measurement will be presented in the next section. Before and after the test, each sample was massed to calculate the amount of mass evaporation. Using the temperature-time curve and the initial-final masses, the rate of mass evaporation was calculated explicitly for every 0.04 s. The activity of each constituent was calculated using a commercial thermodynamic software, ThermoCalc™. For all samples, the amount of mass evaporation and composition shift were predicted numerically with errors of less than 4.8% and 1.7%, respectively, as listed in Table 1. Such an accurate prediction is also important for the measurement of density of materials with high vapor pressure.

Measurement of thermophysical properties

The importance of accurate measurement of density cannot be overemphasized as it affects many other properties and modeling of processes including transport phenomena. For instance, density is used to determine other thermophysical properties such as viscosity. The abrupt change in density sometimes provides a critical clue for microstructural change resulted from liquid to liquid or solid to solid transformations. Density is also an essential property in the numerical simulation of transport phenomena. As mentioned, our space mission includes the measurement of the density of flight compositions. Measuring the density of the flight samples in ISS is important because the density data are to be used for numerical simulations of the corresponding sample. The validation of the measured density data including other thermophysical properties is as important as the measurement itself. To provide reference data, the density of five different compositions (Fe, Fe₅₀Co₅₀, Fe₆₀Co₄₀, Fe₇₀Co₃₀, and Co) of the iron-cobalt system was measured using ESL at NASA MSFC [2]. Initially, three samples for each composition were prepared by the same procedure described in the previous section and tested, but one Fe₆₀Co₄₀ and one Co sample were lost during processing so they were excluded for the density calculation.

The thermal cycles for density measurement are slightly different from those for mass evaporation measurements. Instead of thermal hold, the sample was allowed to free-cool once it reached 100 °C superheating. After solidification, the sample was reheated back up to T_m + 100 °C and the melting cycle was repeated for three to five times. The change in projected area of each sample was analyzed to trace the volume change as a function of time. And the volume change was combined with the temperature history to calculate the density as a function of temperature. When mass evaporation was not considered, the apparent density for each melt cycle increased as the number of melting accumulated. By the proper reflection of mass evaporation, the apparent liquid density became independent of melt cycle showing a consistent value. The increase in density by the preferential evaporation of iron is as small as 0.1 % per unit composition shift (1 %p). The measured liquid density of each composition was fitted with a linear function and rearranged in the form of

$$\rho(T) = \rho_{T_m} + a(T - T_m) \quad (3)$$

where $\rho(T)$ is the density as a function of temperature (T) in K, ρ_{T_m} is the density at the melting point (T_m), and a is the rate of change in density per unit temperature, which is directly related to the coefficient of thermal expansion. The values for ρ_{T_m} and a for each composition measured are given in Table 2.

These data have been serving as a reference values for the density measured in space, and the temperature-dependent density data are being used for the numerical modelling of mass transport phenomena described in the next section.

NUMERICAL

Magnetohydrodynamic modeling of EM-levitated molten metal droplets

Melt convection affects the kinetics during solidification of some alloys such as Fe-Co and Fe-Cr-Ni, which show a two-step solidification behavior [16]. When these alloys are rapidly solidified from a deep undercooled state, they preferentially solidify into a metastable body-centered cubic (BCC) phase that in turn transforms into a stable face-centered cubic (FCC) phase. In the second solidification step, some of metastable dendrites are remelted by the latent heat ejected by the second phase solidification and the microstructure becomes finer and more globular. This result implies that if the solidification process can be well-understood and controlled, the resulting microstructure can be tailored. Convection in the melt influences the time duration between the two solidification events (liquid to metastable BCC, metastable BCC to stable FCC), which is referred to as delay time. The solidification front speed of stable FCC phase is much faster by orders of magnitude than that of metastable BCC phase as a result of the difference in thermodynamic driving force. Therefore, to induce two-step solidification on the entire sample, the delay time must be long enough such that the whole volume of the sample is swept by metastable BCC phase prior to nucleation of the stable FCC phase. Containerless experiments both on earth and in space revealed that the delay time is a strong function of melt convection [3].

The influence of internal convection in molten Fe-Co alloys on their solidification behavior is being investigated using MSL-EML. For the success of the space mission, accurate prediction of convection in the sample during thermal cycle is indispensable. A magnetohydrodynamic (MHD) model that characterizes the convection inside an electromagnetically-levitated (EM-L) molten metal droplet was developed [4]. A molten metal can be assumed to be incompressible and Newtonian, and its flow is governed by Navier-Stokes' equations:

$$\begin{aligned} \nabla^* \cdot \vec{u}^* &= 0 \\ \frac{\partial \vec{u}^*}{\partial t^*} + \vec{u}^* \cdot \nabla^* \vec{u}^* &= -\nabla^* P^* + \frac{1}{Re} \nabla^{*2} \vec{u}^* + \vec{F}^* \end{aligned} \quad (4)$$

where $\vec{u}^*$ is the velocity in a vector form, t^* is the time, P^* is the pressure, Re is the Reynolds number that defines the ratio of inertial to viscous effects, and $\vec{F}^*$ is the body force vector that is electromagnetic forces per volume in this case. $\vec{u}^*$ and P^* are unknowns that this set of differential equations is to be solved for. It can be noticed from Eq. 4 that the flow is characterized by a single nondimensional number, Re .

These full Navier-Stokes' equations were solved using a commercial computational fluid dynamics package, ANSYS Fluent [5]. For the first quantitative validation of the developed model, a $\text{Co}_{16}\text{Cu}_{84}$ droplet was tested using a ground-based EML. This composition exhibits so called "metastable liquid phase separation" when it is undercooled below 1486 K. In the recorded high speed video, Co-rich particles were observed to drift along the flow of Cu-rich liquid. By analyzing the video, the convection velocity was measured as ~ 0.3 m/s. Under the same parameters, the developed model showed an excellent agreement with the experiment.

The validated model was applied to characterize the flow in the $\text{Fe}_{50}\text{Co}_{50}$ sample [6]. Flow pattern, maximum convection velocity, and Reynolds number (convection status: laminar or turbulent) were predicted to help design the space experiments. For instance, with the $\text{Fe}_{50}\text{Co}_{50}$ sample, we are studying the influence of convection on the phase selection during solidification. After the sample reaches a superheated region to be fully melted, it will be cooled down to a thermal hold temperature and will be allowed to solidify. For spontaneous solidification to occur, the thermal hold temperature must be lower than the maximum undercooled temperature of the sample. Otherwise, the sample will remain as liquid. Once the amount of undercooling is identified at the first melt cycle, thermal hold temperatures for the next cycles can be determined accordingly. The shaded area in Fig. 1 shows the predicted range of maximum convection velocity and the corresponding Reynolds number as a function of maximum undercooling of the sample. The vertical dotted line on the left was determined by the minimum undercooling for $\text{Fe}_{50}\text{Co}_{50}$ to solidify in two steps. With an undercooling shallower than 50 K, this composition solidifies directly into the stable FCC phase. The dashed line on the bottom shows the minimum convection that was induced by the current to maintain the stability of the sample during processing. If the current is reduced for less convection, the sample may escape the sample holder. The upper bold solid line, the maximum convection velocity, for a given undercooling, is

determined by the coil current which results in thermal equilibrium at that temperature. Using this area of accessible range of convection, the space experiments were designed and the results downloaded from ISS are being analyzed.

CONCLUSIONS

Ground support work in support of the current space experiments was introduced. Mass evaporation and thermophysical properties of Fe-Co alloys were measured using ground-based ESL at NASA MSFC. The internal convection of an EM-levitated Fe₅₀Co₅₀ droplet was characterized by a MHD model that had been validated by the ground-based EML experiment. The results of these ground support activities are being used to design and analyze the experiments performed using MSL-EML aboard the ISS. Similar work is being done for other flight compositions.

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Table 1 – Mass evaporation and composition shift of Fe₅₀Co₅₀ [1]

Sample	Thermal hold (T _m ± K)	Thermal hold duration (s)	Initial mass (mg)	Mass evaporated (mg)		Error (%)	Mole fraction of Fe (at%)			Error (%)
				Measured	Predicted		Initial	Measured final	Predicted final	
1	+20	70	50.016	1.878	1.887	0.49	0.4906	0.4906	0.4859	-0.96
2	-73	50	42.185	2.525	2.441	-3.32	0.4963	0.4817	0.4804	-0.28
3	~0	60	39.334	2.324	2.297	-1.15	0.4965	0.4744	0.4798	1.13
4	+80	85	40.839	3.763	3.920	4.17	0.4911	0.4703	0.4624	-1.67
5	~0	90	38.774	1.725	1.807	4.77	0.4880	0.4949	0.4803	-1.58
6	+53	38	41.488	1.810	1.729	-4.45	0.5017	0.4890	0.4903	0.25

Table 2 – Liquid density of Fe-Co system [2]

Composition (at %)	T _m (K)	Number of Samples	ρ_{T_m} (kg·m ⁻³ , ρ at T _m)	a (kg·m ⁻³ ·K ⁻¹ , $d\rho/dT$)
Fe	1808	3	6983.74	-0.581
Fe ₇₀ Co ₃₀	1763	3	7242.17	-0.569
Fe ₆₀ Co ₄₀	1753	2	7352.53	-0.612
Fe ₅₀ Co ₅₀	1750	3	7423.47	-0.618
Co	1768	2	7774.30	-0.779

Figure 1 - Accessible range of convection velocity and the corresponding Reynolds number as a function of undercooling of an Fe₅₀Co₅₀ droplet of 6.5 mm in diameter [4].