Thermophysical Properties Measurement of High-Temperature Liquids under Microgravity Conditions in Controlled Atmospheric Conditions

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Abstract

Microgravity conditions have advantages of measurement of surface tension and viscosity of metallic liquids by the oscillating drop method with an electromagnetic levitation (EML) device. Thus, we are preparing the experiments of thermophysical properties measurements using the Materials-Science Laboratories ElectroMagnetic-Levitator (MSL-EML) facilities in the international Space station (ISS). Recently, it has been identified that dependence of surface tension on oxygen partial pressure (Po_2) must be considered for industrial application of surface tension values. Effect of Po_2 on surface tension would apparently change viscosity from the damping oscillation model. Therefore, surface tension and viscosity must be measured simultaneously in the same atmospheric conditions. Moreover, effect of the electromagnetic force (EMF) on the surface oscillations must be clarified to obtain the ideal surface oscillation because the EMF works as the external force on the oscillating liquid droplets, so extensive EMF makes apparently the viscosity values large. In our group, using the parabolic flight levitation experimental facilities (PFLEX) the effect of Po_2 and external EMF on surface oscillation of levitated liquid droplets was systematically investigated for the precise measurements of surface tension and viscosity of high temperature liquids for future ISS experiments. We performed the observation of surface oscillations of levitated liquid alloys using PFLEX on board flight experiments by Gulfstream II (G-II) airplane operated by DAS. These observations were performed under the controlled Po_2 and also under the suitable EMF conditions. In these experiments, we obtained the density, the viscosity and the surface tension values of liquid Cu. From these results, we discuss about as same as reported data, and also obtained the difference of surface oscillations with the change of the EMF conditions.

Introduction

Microgravity conditions have advantages of measurement of surface tension and viscosity of metallic liquids by the oscillating drop method with an electromagnetic levitation (EML) device [1]. Thus, we are preparing the experiments of thermophysical properties measurements using the Materials-Science Laboratory ElectroMagnetic-Levitator (MSL-EML) in the international space station (ISS). Recently, it has been identified that dependence of surface tension on oxygen partial pressure must be considered for industrial application of surface tension values [2]. Effect of oxygen partial pressure (Po_2) on surface tension would apparently change viscosity from the damping oscillation model. Because viscosity values are obtained from damping time of surface oscillations, the change of surface tension with oxygen partial pressure would change the damping time of surface oscillation. However, viscosity is bulk properties, so the values affected by oxygen partial pressure are not real properties. Therefore, surface tension and viscosity must be measured simultaneously in the same atmospheric conditions. In our project using the parabolic flight levitation experimental facilities (PFLEX), the effects of Po_2 and external forces by EML are systematically investigated for the precise measurements of surface tension and viscosity of high temperature liquids. From the above purpose, we performed the observation of surface oscillations of levitated liquid Cu and Ag in the controlled Po_2 atmosphere and various electromagnetic forces (EMF) under the microgravity conditions by parabolic airplane flight in order to decide the suitable conditions of thermophysical properties measurements by EML. On the suitable conditions, we measured the surface tension and viscosity of liquid Cu. From the results, we propose the refinement of the surface oscillation measurement under the microgravity for precise measurements of thermophysical properties of high-temperature liquid, such as industrial application materials and scientifically interested materials, on the future ISS experiments using MSL-EML facilities.

Experiments

a. Oxygen partial pressure control

To control Po_2 conditions in microgravity, we used Ar+H_2 gas as atmospheric condition gas around the levitated liquid samples. The control of Po_2 by Ar+H_2 gas is used the reaction of $H_2+1/2O_2=H_2O$, so Po_2 values has the temperature dependence. However, surface tension of liquid Ag is weak dependence on Po_2 in wide range of Po_2 (10^{-2}-10^{-20}Pa) [3], so we can separate between temperature dependence and Po_2 dependence of surface tension values. On the other hand, liquid Cu has large dependence of surface tension in the range from 10^{-10} to 10^{5}Pa [4]. In the rage from 10^{-20} to 10^{-10}Pa, the surface tension of liquid Cu does not depend on the Po_2 values. For liquid Cu we can perform the measurements of surface tension under the controlled Po_2 conditions below 10^{-10}. Therefore, the method using Ar+H_2 gas is not best way to control Po_2, so for the future experiments we must use oxygen pomp using electrochemical reaction based on the solid-state electrolyte.

b. Electromagnetic levitation experiments on board parabolic flight

We performed electromagnetic levitation experiments under microgravity by parabolic flight of airplane (Gulfstream-II) operated by Diamond Air Service Co. (DAS) using PFLEX. PFLEX has electromagnetic levitation coil in high vacuum chamber, 2kW radio frequency (RF) power supply with 300Hz, control system for electromagnetic levitation conditions, water cooling system for coil cooling, and sample droplet observation system. The observation system has single color pyrometer for temperature measurement of samples and high-speed camera with sampling rate of 250Hz and shutter speed of 100msec and also we monitor oxygen partial pressure in vacuum chamber by the zirconia based solid electrolyte sensor. The surface oscillation of levitated liquid droplets was observed from the top view by high-speed camera with the frame rate of 250kHz by the recording system of the flash solid-state drive (SSD) system. For electromagnetic levitation experiments under microgravity, TEMPUS facilities have been used for long time in European research group [5]. TEMPUS has two coils system for separation between heating and positioning. Thus, since the magnetic field shape is different in the one coil system used in ground experiments, we cannot identify the effect of electromagnetic force on modification of liquid droplet shape. From the above reason, we adopted the one coil system same as the ground experiments. Using one coil system, we can levitate a sample before microgravity conditions during parabolic flight, so the samples can be melted just at the start of microgravity conditions. However, for the one coil system, weak electromagnetic force applies to liquid droplet even in microgravity conditions in order to keep position of droplets, so we cannot reduce the electromagnetic force to completely zero. The effect of the weak electromagnetic force on liquid droplet oscillation, we checked the effect of electromagnetic force on the surface oscillation of liquid droplets before thermophysical properties measurements. For this purpose, we changed electrical current applying to the coil from 0 to 10A.

Experimental Results

We observed surface oscillation of liquid Cu in Ar+H_2 atmospheres under microgravity with different electromagnetic force conditions. The surface oscillation was obtained from the analysis of the change of liquid droplet shape with time. Figure 1 shows the result of the dumping of surface oscillation of liquid Cu droplet with change of electric current applying to levitation coil. From the results, on the large EMF case surface oscillation was long damping time and was not completely damped. This is due to that the EMF continuously generated the surface oscillation during sample levitation. For the case of long time dumping, we cannot obtain correct viscosity. Also, the EMF affects on the surface oscillation frequency shown in Fig.2. Figure 2 shows the power spectrum of surface oscillation of both conditions. From Fig.2, we found that the small EMF had single peak of surface oscillation but for the large EMF case multi peaks found and the main peak position was shifted. This means that the electromagnetic force by applying small EMF case did not modify the surface oscillation of liquid droplets. However, for the case of large EMF, the electromagnetic force modified the surface oscillation. The effects of gravity and electromagnetic force cause the modification of the surface oscillation of electromagnetically levitated liquid droplets in the ground. For the case of electromagnetically levitated liquid droplets under microgravity, only electromagnetic force caused for the effect of modification of the surface oscillation. Thus, details of the modification of surface oscillation are different between microgravity and ground conditions. The modification of surface oscillation of liquid droplets by gravity and EMF must be clarified for the preparation of ISS experiments in near future. Based on the conclusion we selected the levitation condition for precise measurement of surface oscillation for thermophysical properties measurements. Figure 3 shows the results of viscosity of liquid Cu measured with the change of EMF. In Fig.3, previous reported viscosity of liquid Cu [6] was also plotted. From the results, on the small EMF case viscosity values almost the same as the previous reported values. From our results, temperature dependence of liquid Cu viscosity was fitted by Arrhenius type formula with the activation energy of 25kJ/mol. From these measurements, in order to obtain correct viscosity we found the critial EMF conditions for keeping the sample position was below 3% of maximum EMF for heating.

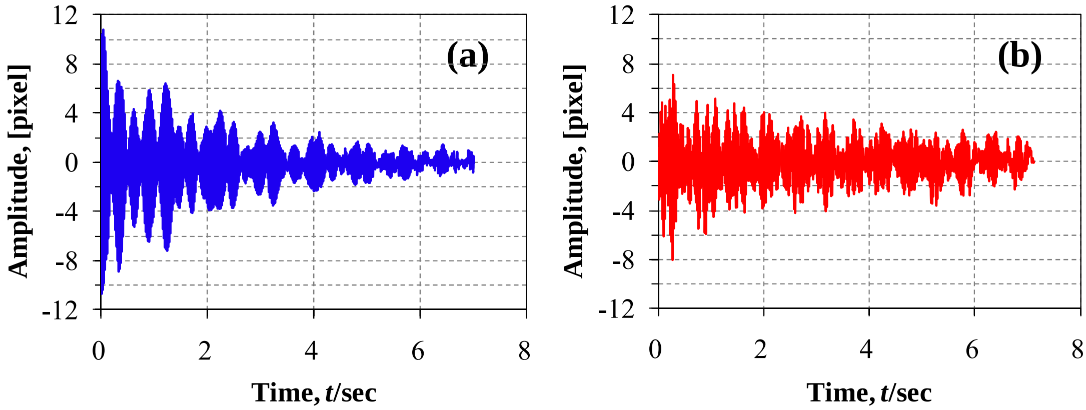

Fig.1 Surface oscillation dumping of liquid Cu at 1620K under microgravity conditions with different electric current applying to RF coil, (a) 0.2A and (b) 7A

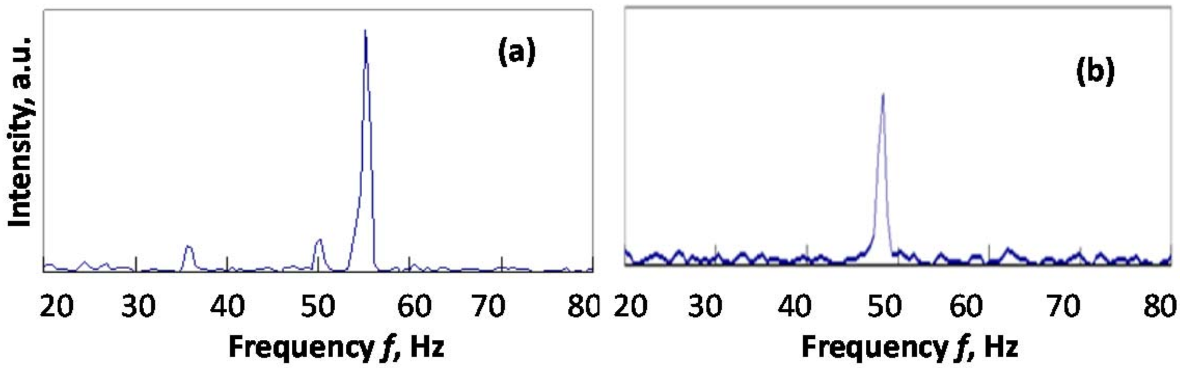

Fig.2 Power spectrum of surface oscillations of liquid Cu shown in Fig.1.

In order to find the affect of EMF on the surface oscillation frequency fro surface tension measurements, we calculated the surface tension values of liquid Cu from the surface oscillation frequency shown in Fig.2 with different EMF conditions. For these measurements of surface oscillations of liquid Cu, we controlled Po_2 condition in 10^{-20}Pa. Figure 4 shows the results of temperature dependence of surface tensions liquid Cu [7] with previous reported values. Figure 4 exhibits the surface tension of liquid Cu calculated from the single frequency peak of the droplet under small EMF case using the Rayleigh equation [8] (■) and that calculated from the frequencies of the m = 0, ±1, and ±2 oscillations for the l = 2 mode of the droplet on the ground using the Cummings and Blackburn calibration (▲) [9]. The surface tension is also calculated from the frequency of the main peak (●) in the spectrum which shows three peaks as shown in figure 4a. Even though the droplet oscillation does not degenerate completely under microgravity in the EMF conditions shown in figure 8a, the main peak may correspond to the frequency of the l = 2 mode if it is predominant in the oscillation of the droplet. For the case, the surface tension is calculated from the frequencies assigned same as the m = 0, ±1, and ±2 oscillations observed in the terrestrial experiment. The temperature dependence of surface tensions of liquid Cu obtained from single peak of surface oscillation agrees well with the literature values [10,11,12] and the surface tension decreases from about 1300mN/m to 1200mN/m as the sample temperature rises from 1515K to 1815K. However, the calculated surface tension shows a large scatter when it is calculated from the frequency of the main peak in the frequency spectrum that also has small two additional peaks.

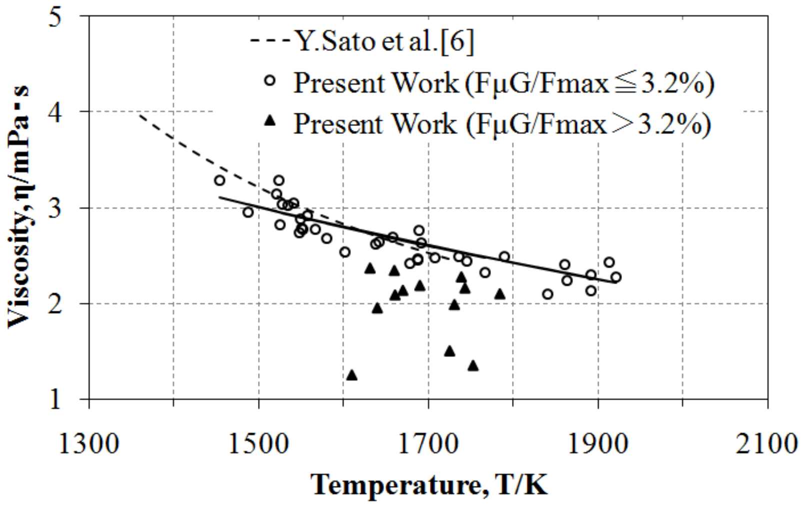

Fig.3 Viscosity of liquid Cu measured under microgravity with different EMF conditions.

This suggests that the main peak does not correspond to the frequency of the l = 2 mode. The surface oscillation of the droplet would not degenerate as long as a few additional peaks are observed in the frequency spectrum even if it is very small.
When the surface tension of the droplet is calculated from the single frequency peak of the surface oscillation observed under the microgravity experiment, it is almost the same as that measured in the terrestrial experiment though it shows a small scattering. This implies that the Cummings and Blackburn equation can calibrate the effect of the deformation of the droplet due to the gravitational acceleration and the external electromagnetic force.

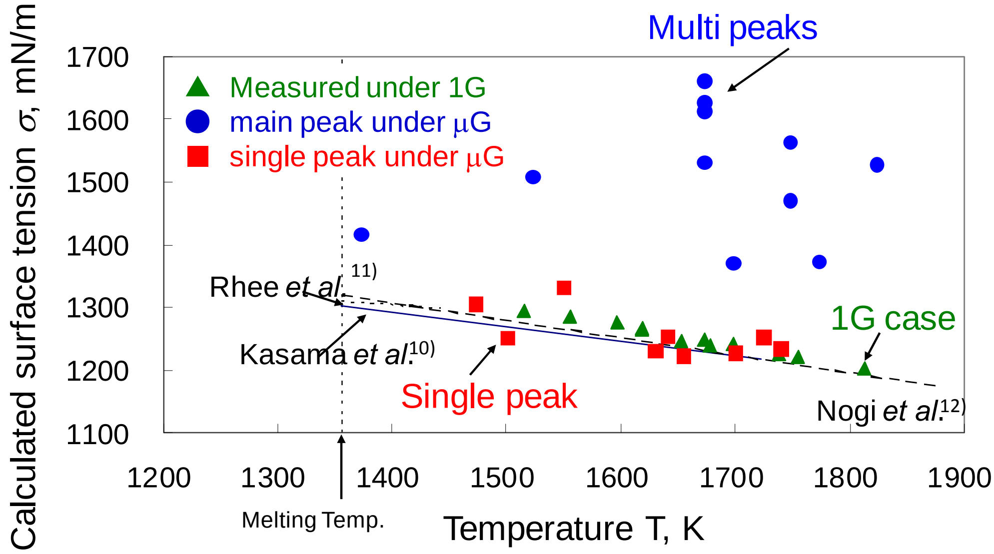

Fig.4 Surface tension of molten copper calculated from the single frequency of the surface oscillation observed under microgravity (■) and that calculated from the frequencies of m =0 , ±1, and ±2 oscillations under 1G (▲). The calculated surface tension from the main peak in the frequency spectrum which shows three peaks as shown in figure 6a is also exhibited (●).

Discussion

As mentioned above, we succeeded in a significant decrease in the electromagnetic force given to the droplet during the viscosity and the surface tension measurements by the oscillating droplet method using EML under microgravity. Furthermore, the oxygen partial pressure of the ambient atmosphere was monitored and controlled simultaneously. The surface tension of the liquid Cu calculated from the single frequency peak of the surface oscillation of l = 2 mode observed under the microgravity condition became almost the same as that calculated from the frequencies of the m = 0, ±1, and ±2 oscillations on the ground. However, the surface tension calculated from the frequency of the single peak for the microgravity experiment showed a small scattering due to the incorrect estimation of the sample mass because the sample could not be collected after the experiment. Furthermore, the calculation result of the surface tension of liquid Cu under microgravity included the influence of the temperature variation of the sample due to the decrease in the electromagnetic force. Since a large electromagnetic force was applied to the sample to levitate it under the high gravity condition of 1.5G before the microgravity condition starts, in many cases the surface oscillation did not degenerate within the short microgravity period prepared by the parabolic flight of the airplane. A long microgravity experiment without any gravity change is expected in the International Space Station (ISS). In this case it is easy to levitate the sample in a stable state. Furthermore, there is a lot of flexibility for the design of the RF coil because a large electromagnetic force is not necessary to levitate the sample. As a result, the sample would be heated and kept at certain temperature easily. Since the sample can be collected after its solidification due to a sufficient experiment time, it would become easy to estimate the change of the sample mass during the measurement. Further investigation is required under a long microgravity environment in the ISS so that the oscillation behavior of the droplet, which is used for calculation of surface tension, can be understood more clearly. We are planning the microgravity experiment in the ISS as collaboration of ESA (European Space Agency), JAXA (Japan Aerospace Exploration Agency), and NASA (the National Aeronautics and Space Administration). For ISS experiments, the present status of our planning was shown below. Our project of thermophysical properties measurements of high-temperature liquid using the materials-science-laboratory electromagnetic-levitator (MSL-EML) in ISS initially focus on the semiconducting materials liquid, so our project name is SEMITHERM. SEMITHERM is concerned with the THERMophysical properties of SEMIconductors, specifically Ge, and Si-Ge.

Conclusion

Using the parabolic flight levitation experimental facilities (PFLEX), the effects of Po_2 and external EMF on surface oscillation of levitated liquid droplets were investigated for the measurements of surface tension and viscosity of liquids Cu. The surface oscillations of levitated liquid Cu using PFLEX on board flight experiments by G-II were observed in $Ar+H_2$ and pure Ar gas atmospheric conditions. From these observations, we obtained the density, the viscosity and the surface tension values as same as reported data, and also obtained the difference of surface oscillations with the change of the Po_2. For these thermophysical properties measurements, we investigated the effect of EMF on the surface oscillations in controlled Po_2 conditions. This clarified the effect of EMF on the surface oscillation of levitated under microgravity. This is different from the ground conditions. Therefore, if large EMF applying cases, we need modified the surface oscillation analysis method from the present method based on the Cummings and Blackburn analysis. We future more systematically perform precise measurements surface oscillation in different Po_2 conditions. On the basis of present experimental results, we are discussing about ISS experiments of MSL-EML with controlled atmospheric Po_2 conditions with Investigator Working-Grope of EML (IWG-EML) in order to progress high temperature liquid science.

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