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ELECTROSTATIC LEVITATION TECHNIQUE FOR THERMOPHYSICAL PROPERTY MEASUREMENTS OF MOLTEN MATERIALS ON EARTH AND IN SPACE

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Thermophysical properties of high temperature molten materials are measured using an electrostatic levitator in combination with several diagnostic devices. An objective of this study is to find the limitation of the earth-based measurements in order to evaluate the necessity of similar measurements in space. It is found that the hemispherical total emissivity, specific heat, specific volume, density and surface tension can be satisfactorily measured by the earth-based measurements. The viscosity measurement is not conclusive because of the suspected interference by the levitation force on the measurement. Further investigation is necessary to elucidate the degree of the interference by performing the same measurement under a short duration microgravity environment realized on earth.

1. INTRODUCTION

Thermophysical properties of high temperature molten materials have not been determined accurately because of the experimental problems associated with taking measurements at high-temperatures. However, the demands for accurate thermophysical property values have been strong in recent years for applications in both scientific and engineering. For instance, the structures of liquids are less understood in comparison to those of solids and gases. The theoretical studies of simple liquids will significantly advance if certain thermophysical properties of molten metals are accurately determined since the metals form the simplest liquid structures. The electronics industry constantly demands high quality semiconductor materials for high density integrated circuit devices. In order to simulate the crystal growth for optimization of the growth conditions, the accurate thermophysical properties of molten semiconductors are essential input parameters.

Among the problems associated with taking measurements at high temperatures, the sample contamination due to reaction with the container materials is a common problem for any property measurements. The contamination can be totally avoided if the material is processed in a containerless manner. Several techniques have been developed to achieve this objective on earth through levitation of a material in air. An additional advantage of levitation is its ability to let the liquid sample to be undercooled below the melting point. Thus the properties of the undercooled liquid can be measured. An obstacle to the levitation techniques is gravity which sets a limitation on materials which can be levitated. Low density materials are generally easy to levitate and process. To overcome this

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limitation, it is natural to seek a microgravity environment to perform the measurements. A short duration of the **microgravity** environment can be realized on earth by free fall of the sample. For a long duration, a satellite orbiting around the earth is an ideal place. Since access to the space environment is very limited and expensive at present, it is important to make best effort to perform earth-based measurements first. The space-based measurements should be reserved only for measurements which cannot be performed on earth.

At the Jet Propulsion Laboratory, we have developed a high temperature electrostatic levitator (HTESL) which can be utilized for the **thermophysical** property measurements. Advantages of the electrostatic levitation technique over other levitation techniques, especially the electromagnetic levitation is **decoupling** of levitation and heating elements and a wide selection of samples to be levitated. **Both** conductive and non-conductive (including semi-conductive) materials can be levitated. The sample can be levitated at any temperature between room and maximum temperatures. An objective of this paper is to find the limitation of the earth-based measurements in order to evaluate the necessity of similar measurements in space. First, we briefly describe the HTESL system. Secondly, we describe the techniques for acquiring the data and processing them to obtain the **thermophysical** property values. Thirdly, we present some of the results which are relevant for discussing the necessity of a microgravity environment to improve the accuracy. Finally, future plans to upgrade the system for enhancing the capabilities are presented.

2. ELECTROSTATIC LEVITATION SYSTEM

Electrostatic levitation is achieved by applying an electrostatic field to a charged sample to counterbalance gravity. Since the electrostatic field does not have a potential minimum position, the sample position must be actively controlled by a feedback system. Figure 1 shows a schematic diagram of HTESL. A sample (typically 2- 3 mm in diameter) is levitated between the bottom and top (not shown) electrodes which **are** usually separated by 10-12 mm. Two pairs of the side electrodes are used to control the lateral motion of the sample. The electrode assembly is placed inside of the vacuum chamber which is typically evacuated to 10^{-8} Torr. Two position detectors provide information for active position control. The sample position is usually updated 480 times per second. Heating of the sample is accomplished by a focused 1 kW xenon arc lamp which can raise the sample temperature up to 2300 K.

Single color pyrometers are used to measure the sample temperature. A long distance microscope is used to take magnified images of the levitated sample. A photo detector with a slit window is used to observe the sample oscillation. The system is run by several desktop computers which are assigned to specific tasks. The sample **position** is controlled by an algorithm which tracks the position, velocity and acceleration of the sample and determines the voltage to be applied. The sample temperature is displayed in real time and also stored for the post experiment analysis. The sample images are stored in video tapes for later processing.

3. DIAGNOSTIC TECHNIQUES

We have developed the diagnostic techniques such as a fast temperature acquisition and **igital** image processing. These techniques allow us to obtain the **thermophysical** properties

which include the specific **volume**², **density**², hemispherical total emissivity³, specific **heat**³, surface tension⁴, and viscosity.

3.1. Hemispherical Total **Emissivity** and Specific Heat

Since a molten sample is levitated in a high vacuum environment and is kept at a high temperature, cooling of the sample after the xenon ramp is suddenly blocked is purely radiative. Thus, the cooling rate, dT/dt of the sample is given by

$$\frac{dT}{dt} = -\frac{\sigma_{SB}\epsilon_T A}{mC_p}(T^4 - T_0^4) \quad (1)$$

where m is the sample mass, C_p is the specific heat, σ_{SB} is the **Stefan-Boltzmann constant**, ϵ_T is the hemispherical total **emissivity**, A is the surface area and T_0 is the temperature of the surroundings. By rearranging Eq.(1),

$$\frac{C_p}{\epsilon_T} = -\frac{\sigma_{SB} A}{m \frac{dT}{dt}}(T^4 - T_0^4). \quad (2)$$

The dT/dt value can be determined from the experimental temperature-time curve by taking a derivative. Thus, by knowing either C_p or ϵ_T from an independent measurement, the other quantity can be obtained by Eq.(2)³. It is found that the numerical determination of dT/dt is sensitive to the quality of the temperature data. Therefore, an accurate temperature measurement with a high speed pyrometer is essential for this technique.

3.2. Specific Volume and Density

A levitated molten sample is slightly elongated along the gravitational axis due to an electrical charge but maintains an **axisymmetric** shape. Therefore, the volume, V , of the sample can be determined from a single image taken from a direction perpendicular to the **symmetry axis** by the **CCD camera**. The **technique** involves digitizing the image and fitting the image edge with the spherical harmonic functions. Once the volume as a function of temperature is determined, the thermal volume expansion coefficient, β , is calculated by

$$\beta = \frac{1}{V} \frac{\partial V}{\partial T}. \quad (3)$$

The volume can be converted into the specific volume, v , or the density, p , by knowing the sample mass as

$$v = \frac{1}{p} = \frac{V}{m}. \quad (4)$$

The sources of errors are the image resolution, sample temperature and sample mass. The overall error of the current technique is estimated to be 0.2 %.

3.3. Surface Tension and Viscosity

A molten sample held at a constant temperature may induce a small oscillation when a small sinusoidal electric field whose frequency is close to the resonance frequency of the sample is superimposed to the levitation electric field for a short time⁵. The decay of the resonance oscillation after the imposed oscillation can be monitored by an opto-electronic device to determine the frequency, f , and the decay constant, τ . The fundamental frequency, ω (mode $n=2$), can be related to the surface tension, σ , through the following equation,

$$\sigma = \frac{r_0^3 \rho}{8} \left(\omega^2 + \frac{Q^2}{8\pi^2 r_0^6 \rho \epsilon_0} \right) \quad (5)$$

where r_0 is the sample radius, ρ , is the density, ϵ_0 , is the permittivity of vacuum and Q is the uniform surface charge. The surface charge can be approximately determined by

$$Q = mg \frac{L}{V_p} \quad (6)$$

where g is the gravitational acceleration, L is the gap between the electrodes and V_p is the potential difference. The effects of non-uniform charge distribution and non-sphericity are corrected using the method given by Feng and Beard⁶:

$$\omega_{ob}^2 = \omega^2 [1 - F(\sigma, q, e)] \quad (7)$$

where ω_{ob} is the observed frequency and q and e are defined as

$$q^2 = \frac{Q^2}{16\pi^2 r_0^3 \epsilon_0} \quad (8)$$

$$e^2 = E^2 r_0 \epsilon_0 \quad (9)$$

where E is the applied electric field. $F(\sigma, q, e)$ is defined as

$$F(\sigma, q, e) = \frac{(243.31\sigma^2 - 63.14q^2\sigma + 1.54q^4)e^2}{176\sigma^3 - 120q^2\sigma^2 + 27\sigma q^4 - 2q^6} \quad (10)$$

For a small amplitude oscillation, damping of the oscillation can be written in a weak damping limit as⁷

$$\frac{1}{\tau} = \frac{5\eta}{\rho r_0^2} \quad (11)$$

where η is the viscosity. Rewriting Eq. (11)

$$\eta = \frac{\rho r_0^2}{5\tau}. \quad (12)$$

Eq. (12) is only valid when the sample is nearly spherical. This condition is satisfied with the electrostatic levitation but not with the electromagnetic levitation in which the sample is deformed by the strong magnetic field. For this reason, Eq. (12) has never been applied to the measurements with the electromagnetic levitation technique.

4. EXPERIMENTAL RESULTS

We have applied the above techniques to various metals, metallic alloys and semiconductors. In this paper, we present the results from the measurements on the **Ni-Zr** alloys and Si. For these results, independent results obtained under a microgravity environment are available for comparison.

The surface tension of the **Ni₃₆Zr₆₄** liquid alloy was measured during the **Spacelab IML-2** mission in 1994 using the electromagnetic **containerless** processing facility **TEMPUS⁸**. The measuring technique was the oscillating drop method. This **eutectic** alloy is an easy glass former and shows relatively good undercooling ability. The result is reproduced in Figure 2. For the direct comparison, the result of the present study is also shown in Fig. 2. The linear regression gives the following equations:

$$\sigma = 1.545 \cdot 8.0 \times 10^{-5} (T - 1010) \text{ (N/m) (Egry et al.)} \quad (13)$$

$$\sigma = 1.511 + 3.8 \times 10^{-5} (T - 1010) \quad \text{(Present study).} \quad (14)$$

Considering the errors involved in the measurements, both results agree well.

Figure 3 shows the viscosity of the alloy obtained by the present study. The result can be fitted with the **Arrhenius** equation as

$$\eta(T) = 0.0028 \exp\left(\frac{95800}{RT}\right) \text{ (mPa s)} \quad (15)$$

where T is in Kelvin and R is the gas constant. Corresponding results from the **space-based** measurement are not available.

The surface tension of Si was measured by an oscillating drop method using a ground-based electromagnetic levitation system^{9,10}. The data was corrected to simulate the results from the space-based measurement by removing the effects of magnetic and gravitational forces¹¹. The result is reproduced in Figure 4. The result of the present study is also shown in Fig. 4. Note that most data points are in the undercooked region. The maximum undercooking level of Si processed by the levitation technique is approximately 300 K. The results disagree in the undercooked region. The Przyborowski et al. result shows a linear temperature dependence which is extended into the undercooked region. On the other hand, the present result shows a temperate independence in the undercooked region.

Figure 5 shows the viscosity of Si obtained by the present study. The viscosity rapidly increases below 1350 °C. Corresponding results from the electromagnetic levitation technique are not available.

5. DISCUSSION

As mentioned before, an objective of the present study is to find the limitation of the earth-based measurements in order to evaluate the necessity of similar measurements in space. For the hemispherical total emissivity, specific heat, specific volume and density, the only limitation of the measurements is the sample density. Heavy materials are difficult to levitate and process. However, this limitation should be overcome with the refinement of the system.

Levitation on ground requires a strong force to counterbalance gravity. The interference by the force may partially or completely disable the techniques for some other property measurements. For the surface tension measurement by an oscillating drop method, if the sample is deformed due to the force, the fundamental frequency may be split. The deviation from the sphericity of the electrostatically levitated sample is approximately 1-2 % of the diameter. However, we did not observe the apparent frequency split due to deformation. In some instance, we observed the frequency shift due to sample rotation. We discarded the data from the rotating sample. More serious interference may occur when the sample oscillation is coupled with the dynamic variation of the force. We observed the modulated decay of the oscillation. In extreme cases, the amplitude of the oscillation was amplified until the sample could not stay in position. The observable coupling occurs within certain ranges of parameters which include the temperature, sample mass, applied voltage and sample position relative to the electrodes.

We noticed that the coupling with the levitation force did not significantly alter the fundamental frequency. In other words, even when the damping of the oscillation was obviously modulated, the sample was still oscillated with the fundamental frequency. Therefore, the effect of the coupling is minor in the surface tension measurement, Fig. 2 is the only true comparison of the surface tension results between the space-based and ground-based measurements. By considering the errors involved in the measurements, both results seem to agree each other. On the other hand, the simulated space-based result in Fig. 4 disagrees with the present result. If the difference in the levitation techniques is not the source of the disagreement as indicated by the results in Fig. 2, the discrepancy must come from other sources such as the sample purity and the degree of oxidation. In any case, this discrepancy does not suggest the necessity of a microgravity environment for the surface tension measurements.

For the viscosity, direct comparison with the result of the space-based measurement is unavailable. The earth-based electromagnetic levitation technique cannot perform the viscosity measurement because of a large deformation due to the levitation force. The results in Figs. 3 and 5 were selected from the measurements **where** no obvious coupling was observed. The data above the melting point fairly agrees with the reported **values**¹² measured by the non-levitation techniques, However, it does not give any assurance for the data in the **undercooled** region since the temperature is a coupling factor which might induce coupling in the undercooled region. For further investigation of this subject, we have initiated a collaboration with a group of researchers who will perform similar measurements under a short duration microgravity environment realized on earth. An outcome will clarify whether the elimination of the levitation force improve the accuracy of the measurement. It may reveal that the accurate measurement requires a long duration microgravity environment since the decay of low viscosity materials takes a longer time.

At the same time, we will work on enhancing the present capabilities of the system. We will install devices which will allow us to raise the upper limit of the temperature, measure the spectral **emissivity** of the sample and manipulate the sample rotation. These enhanced capabilities will allow us to improve the accuracy of the measurements. We envision adding the capabilities which allow us to measure the thermal and electrical **conductivities** and to determine the liquid structures in the near future.

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Captions

- Figure 1. Schematic diagram of the **HTESL** system, (a) vacuum chamber, (b) diode laser, (c) **HeNe** laser, (d) xenon lamp, (e) position detector, (f) **CCD** camera with a long distance microscope, (g) beam splitter, (h) oscillation detector with slit window, (i) position detector, (j) focusing lens, (k) sample, (l) bottom electrode and (m) side electrode.
- Figure 2.** The surface tension of the **Ni₃₆Zr₆₄** alloy as a function of temperature measured by the space-based and ground-based experiments. The melting point (**MP**) of the alloy is shown by the arrow.
- Figure 3. The viscosity of the **Ni₃₆Zr₆₄** alloy as a function of temperature measured by the ground based experiment.
- Figure 4. The surface tension of Si as a function of temperature measured by the simulated space-based and ground-based experiments. The *melting* point (**MP**) is shown by the arrow.
- Figure 5. The viscosity of Si as a function of temperature measured by the ground based experiment,.

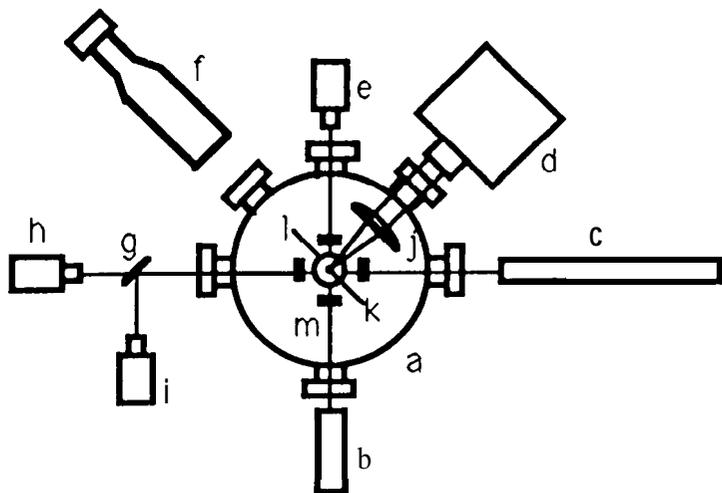


Fig. 1

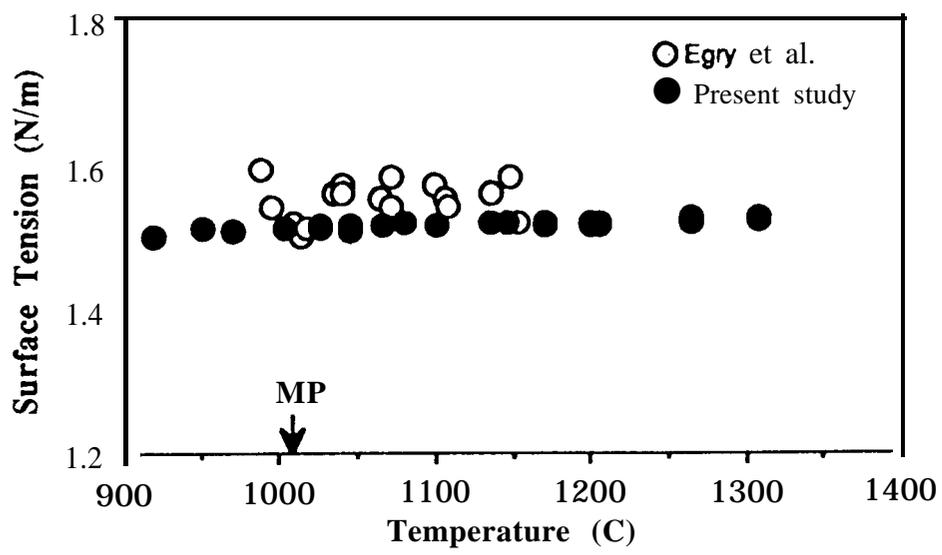


Fig. 2

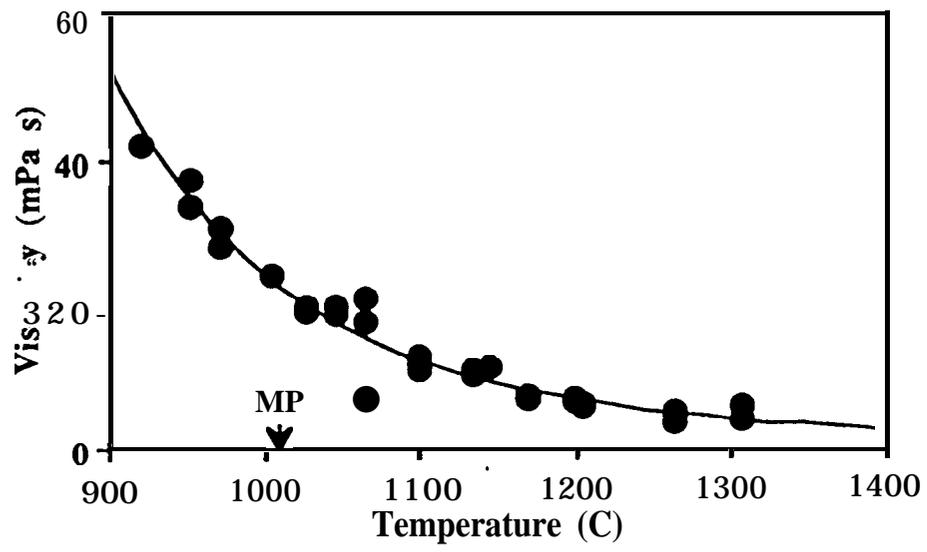


Fig. 3

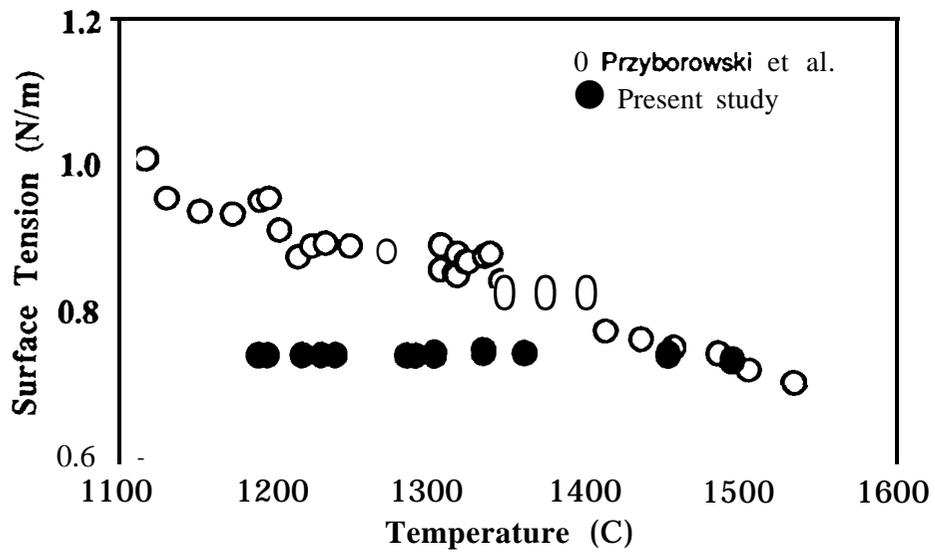


Fig. 4

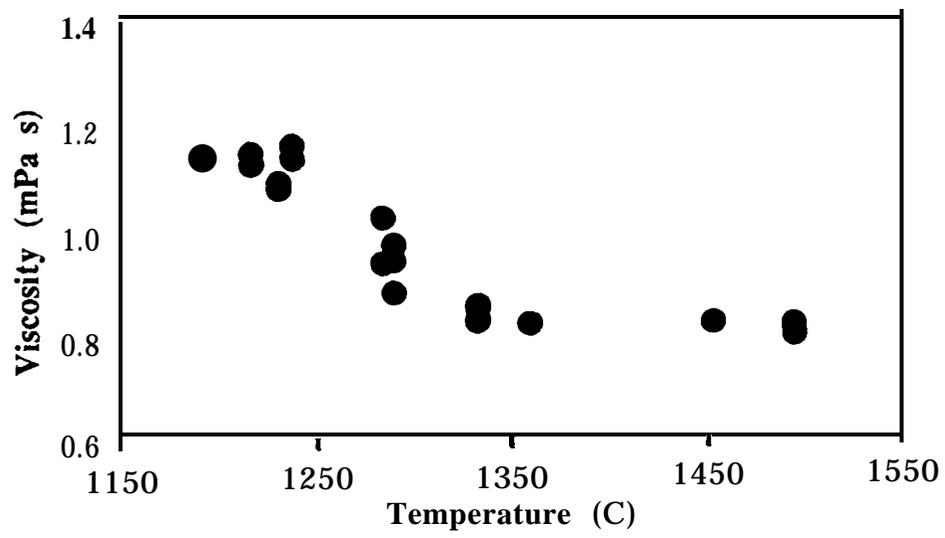


Fig. 5