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FINAL TECHNICAL REPORT

MEASUREMENTS OF THE VISCOSITY OF THE UNDERCOOLED MELTS UNDER THE CONDITIONS OF MICROGRAVITY AND SUPPORTING MHD CALCULATIONS

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Executive Summary

This report covers the work done by MIT's Materials Process Modeling Group under grant number NAG8-970 from December 1, 1992, to September 30,1995, to support the measurement of the surface tension and viscosity of undercooled metals with the TEMPUS as a part of the IML-2 mission. This work involved close collaboration with the other TEMPUS PI's, including Prof. Egry of the DLR, Prof. Johnson of Caltech, and Prof. Fecht of TU-Berlin. This work resulted supported the Ph.D. thesis of Elliot Schwartz (MIT Ph.D. 1995) and partially supported that of Robert Hyers (MIT).

This work consisted of two parts. First was the ground-based research program, whose purpose was to establish the feasibility of the flight program and to establish the knowledge base necessary to plan a successful flight program. Analytical calculations established a fundamental understanding of the problem, and then a more rigorous program of numerical calculations was employed in the development of the experimental program.

The second part of the covered research was the actual flight program, developed in close collaboration with Prof. Egry. A metal sphere was levitated and melted, and then set oscillating by "squeezing" it with the magnetic field. The droplet's oscillations were recorded on videotape and digital image analysis was employed in reduction of the data.

The experiment was successful because surface tension measurements were obtained for the gold and gold-copper alloy samples. This data provides unique experimental evidence in support of theories about electromagnetic levitation on the ground. Because of unforeseen difficulties with stability of liquid samples in TEMPUS, the viscosity measurements were not possible, but the data collection and analysis techniques were well proven in this mission. Further research is being conducted regarding the measurement of viscosity by this technique.

Table of Contents

I. Introduction

It was proposed to use the TEMPUS electromagnetic levitator to employ the oscillating droplet technique to measure thermophysical properties of liquid metals. Containerless processing methods promise to allow measurements which are otherwise impossible in the undercooled regime to be carried out. This technique is also very promising where reactivity or sensitivity to contamination is an issue.

The theory behind the measurement of surface tension of an oscillating spherical droplet comes from Rayleigh (1879) [15]. Rayleigh's equation (eq. 1) gives the relation between the frequency of oscillation ω and the surface tension γ of a liquid sphere subject to no external forces and only small deformations. n is the mode of the oscillations, with n=2 being the first mode observed for axisymmetric incompressible droplets.

$$\omega_n = \sqrt{\frac{n(n-1)(n+2)\gamma}{\rho R_o^3}} \qquad \text{(eq. 1)}$$

An extension of this theory by Lamb (1881) [16] allows for the effect of viscosity on the oscillations. The viscosity μ of the sample determines the damping constant τ as described in (eq. 2)

$$\tau_n = \frac{\rho R_o^2}{(n-1)(2n+1)\mu} \qquad \text{(eq. 2)}$$

Lamb's equation postulates that the viscous dissipation due to the oscillatory flows is the only mechanism for damping this motion, and that the flows are simple and laminar.

Electromagnetic levitation provides a method to support and melt a droplet without contact with a container. However, in ground-based levitation, the need to provide a strong, unbalanced force field to support the weight of the sample excites large velocity internal fluid flows in a liquid droplet. For normal metals, the flows induced by earthbound levitation are turbulent.

The turbulent eddies constitute another mechanism of dissipation of kinetic energy, so Lamb's equation is not applicable droplets with strong internal flows. It is to achieve the simple laminar flows required by Lamb's analysis that we proposed to perform this experiment in microgravity, where the small accelerations allow the sample to be constrained with only small forces. These small forces will, in turn, cause only basic laminar fluid flow in the sample.

Surface tension measurements on the ground are possible; however the magnetic forces that support the weight of the droplet do change the frequency of oscillation of the droplet's free surface. With the publication of the correction formula by Cummings and Blackburn (1991) [17], the contribution of the magnetic field may be removed. This correction is based on theoretical calculations, and experimental verification of this correction requires the use of negligible magnetic forces, a condition that is only achievable in microgravity.

TEMPUS is an electromagnetic levitation system designed and developed for microgravity use. It consists of separate positioning and heating coils in a self-contained ultra-high vacuum system. The coil geometry used in IML-2 TEMPUS is shown in figure 1.

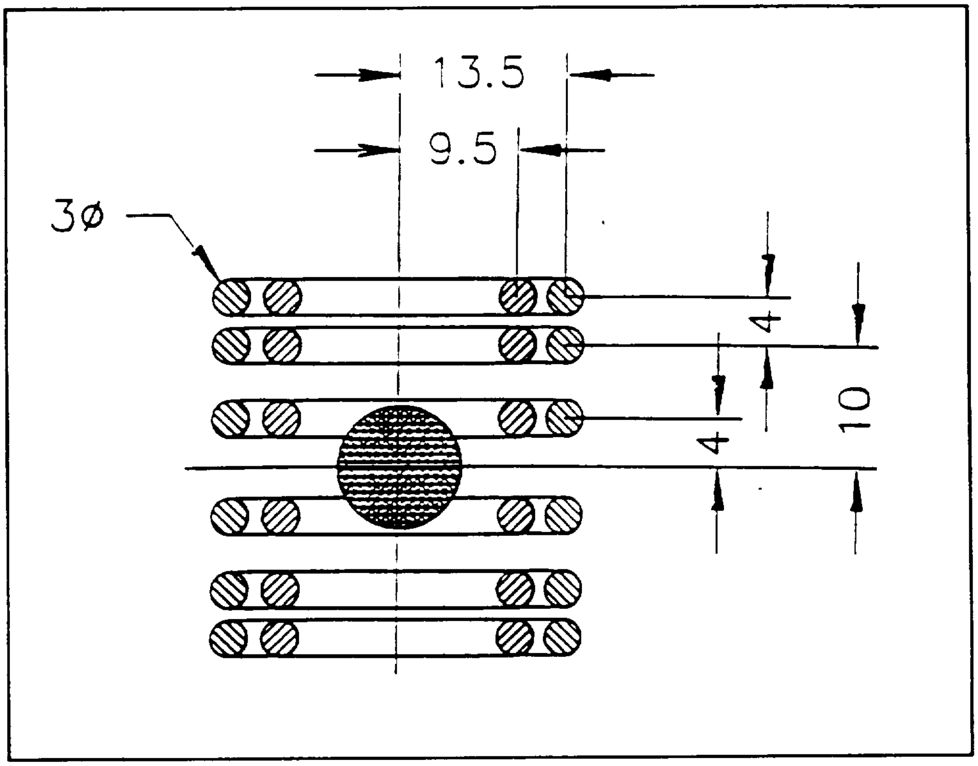

Figure 1: Schematic diagram of TEMPUS coil system and sample.[14]

II. Ground-Based Research

The ground based research program had two main purposes: to establish the feasibility of the proposed flight experiment, and to develop the operating parameters required to achieve the success of that flight research.

In pursuit of the first goal, a series of asymptotic calculations were made which provided encouraging results. These calculations included calculation of heating and cooling rates for various possible sample sizes and materials.

Following these asymptotic calculations, a more in-depth numerical analysis was undertaken. The details of the development of the methods involved in this analysis are described in [2-8], included in the Appendix. The methods employed are explained briefly below.

First, numerical methods for determining the positioning force and Joule heating of the sample were developed [2-4,7] and tested experimentally with solid samples suspended on a wire [6]. These calculations used the method of mutual inductances to solve Maxwell's equations (eq. 3) and the constitutive equations (eq. 4) to determine the current density and magnetic field throughout the sample. Here, E is the electric field, H the magnetic field, B the magnetic flux, J the current density, and σ the conductivity of the sample.

$$\begin{aligned} \nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t} \\ \nabla \times \mathbf{H} &= \mathbf{J} \\ \nabla \cdot \mathbf{B} &= 0 \end{aligned} \qquad \text{(eq. 3)}$$

$$\begin{aligned} \mathbf{J} &= \sigma \mathbf{E} \\ \mathbf{B} &= \mu_0 \mathbf{H} \end{aligned} \qquad \text{(eq. 4)}$$

Then, once the current density and magnetic field are known, the electromagnetic force field F (Lorentz force per unit volume) can be calculated (eq. 5). Similarly, the distribution of Joule heating per unit volume P can be calculated for the known distribution of current density (eq. 6).

$$\mathbf{F} = \frac{1}{\tau}\int_0^{\tau} \mathbf{J} \times \mathbf{B}\, dt \qquad \text{(eq. 5)}$$

$$\mathbf{P} = \frac{1}{\tau}\int_0^{\tau} \mathbf{J} \cdot \mathbf{E}\, dt \qquad \text{(eq. 6)}$$

The equilibrium shape of the droplet can be estimated by a balance of static pressure and magnetic pressure [5,8]. However, more sophisticated model of the shape of a deformed droplet must take into account the internal fluid flow [9]. This flow is governed by the Navier-Stokes equations (eq. 7,8), with terms added to account for the distributed magnetic force field. Here u is the velocity, ρ the density, and μ the viscosity of the fluid, while F_{em} is the electromagnetic body force.

$$\nabla \cdot \mathbf{u} = 0 \qquad \text{(eq. 7)}$$

$$\rho \frac{\partial \mathbf{u}}{\partial t} = \rho \mathbf{u} \cdot \nabla \mathbf{u} = \mu \nabla^2 \mathbf{u} - \nabla P + \mathbf{F}_{em} \qquad \text{(eq. 8)}$$

The magnetic force field was determined by the method described above, and in [4,7]. Then the FIDAP commercial finite element fluid dynamics package was used to solve the fluid flow field determined by the magnetic force field, the materials properties, and the appropriate free-surface boundary conditions. The equilibrium location of the free surface was determined by the requirement of zero fluid velocity perpendicular to the surface and no traction on the liquid free surface. The position of the free surface was calculated for a range of different operating parameters for the materials to be used in the flight experiments.

These calculations determined that the droplet would not be significantly deformed by the positioning coils' field alone, but that the dipole heating field would provide sufficient deformation to allow the planned measurements to be performed. Based on these calculations, appropriate parameters were chosen to excite free surface oscillations with a short duration pulse (100 msec) in the heating field.

Also, the velocity field provided by the solution of the Navier-Stokes equations was analyzed for stability. Preliminary calculations suggested that the parameters planned would provide laminar flow in the droplet, allowing viscosity as well as surface tension measurements.

II. Flight Research

The purpose of this research was realized when the TEMPUS facility flew in July 1994 aboard the Space Shuttle Columbia as a part of the Second International Microgravity Laboratory (IML-2). The research program consisted of stably positioning and melting a sample, and then using pulses of the heating field to deform the sample and induce oscillations of the free surface. Both top and side views of the oscillations were recorded from the TEMPUS on-board video cameras at a high frame rate (240 Hz). Then the video data were processed using digital image analysis to determine the deformations of the droplet as a function of time.

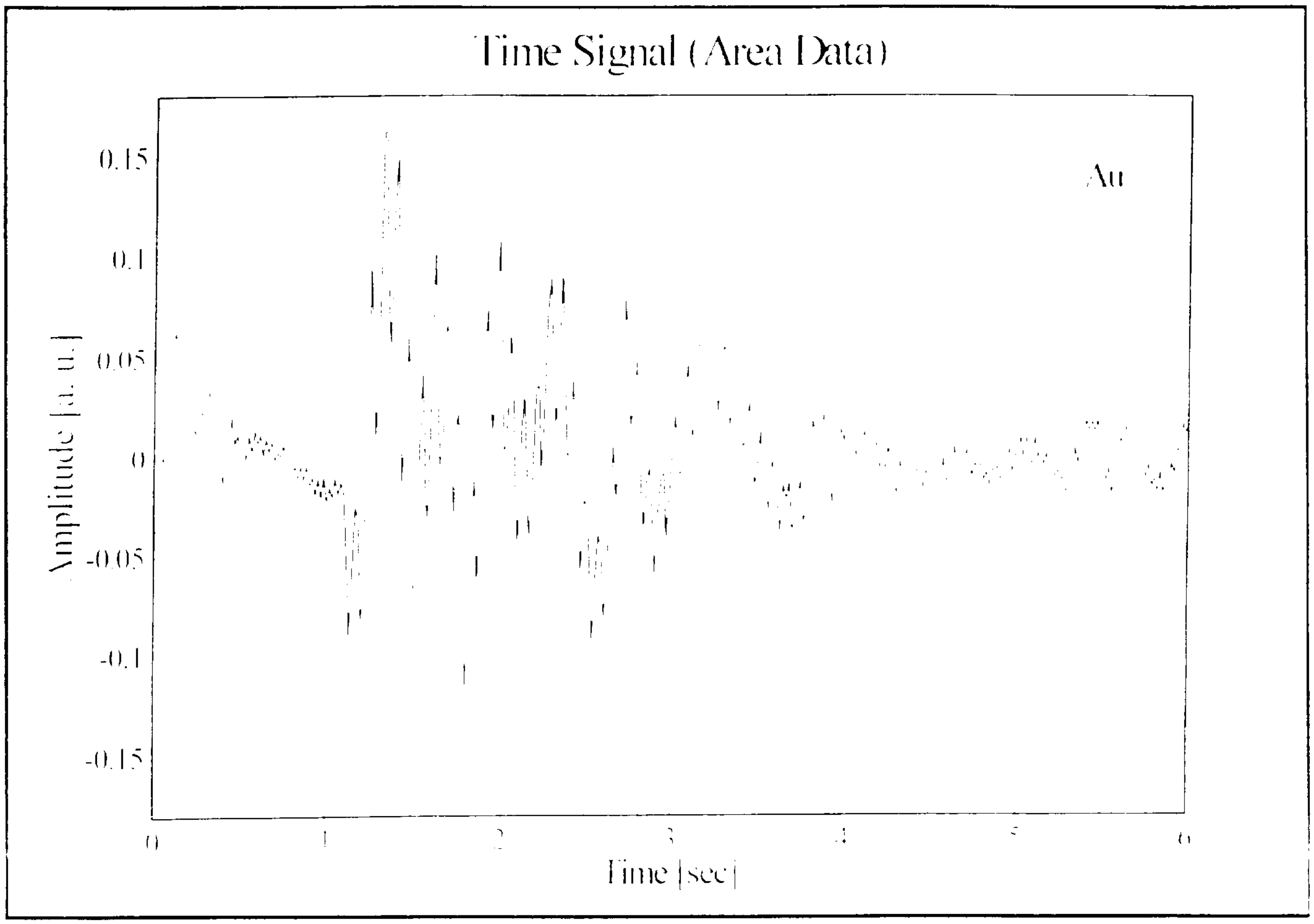

Figure 2: Raw oscillation data from Au sample in IML-2. From Egry, et al.

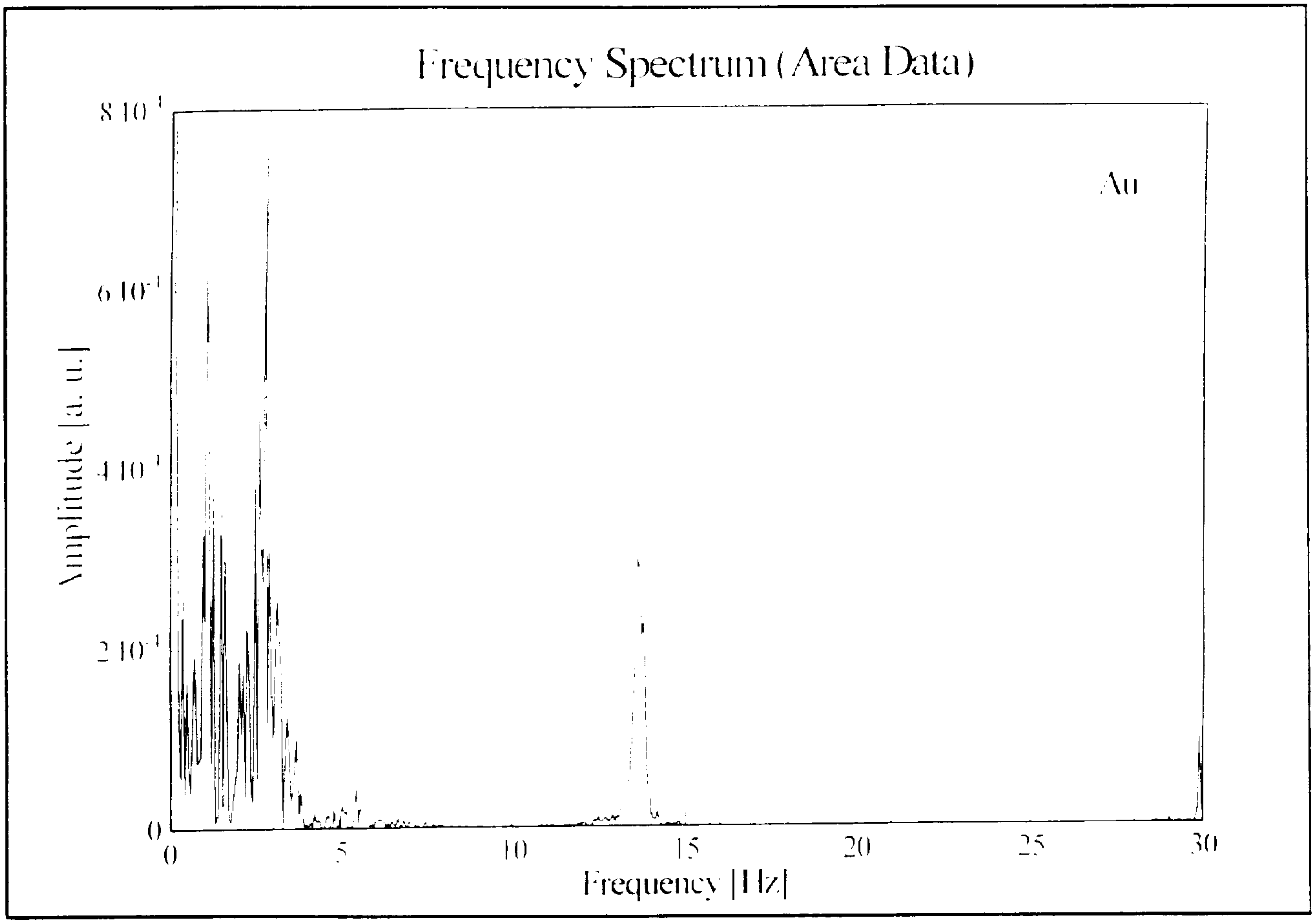

Figure 3: Fourier spectrum of Au sample in IML-2. From Egry, et al.

After the deformation signal was extracted from the video record, a Fourier transform of the signal was taken to determine the frequency of oscillations. Then the Fourier spectrum was filtered to eliminate errors due to translational and rotational motion of the sample and back-transformed. The resulting data were fit to an exponential envelope by statistical methods.

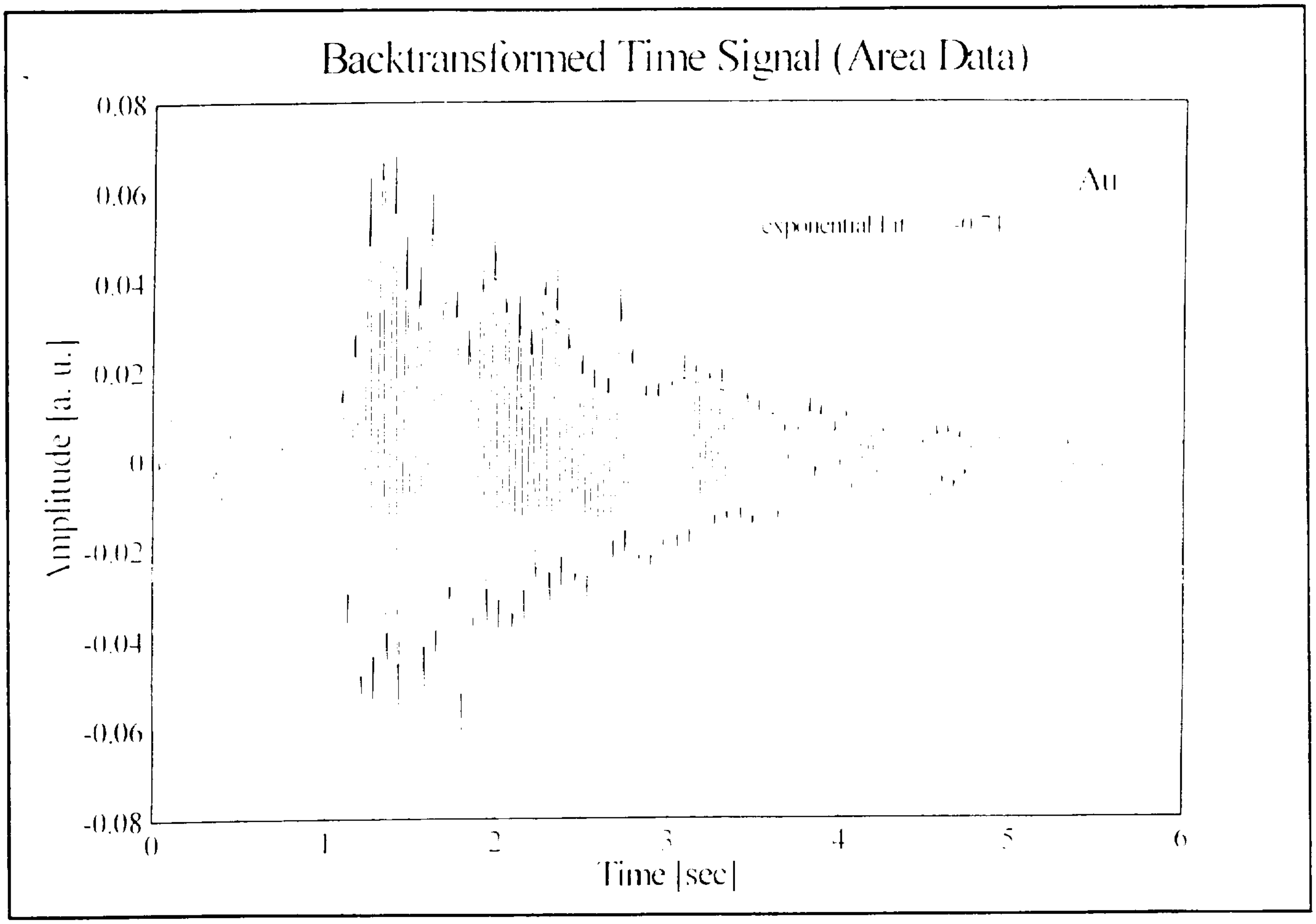

Figure 4: Filtered oscillation data for Au in IML-2 TEMPUS experiment. The constant γ is the reciprocal of the time constant τ. From Egry, et al.

Because of problems with the TEMPUS facility, discussed further in section IV., the 10mm Cu sample was lost to impact with the sample cage before providing oscillation measurements. The Au and AuCu samples provided good surface tension data, providing experimental evidence in support of the correction developed by Cummings and Blackburn [17]. These results can only be obtained in microgravity, where the correction due to the fields supporting the sample's weight is negligible.

The surface tension data for Au and AuCu are presented in figs 5 and 6.

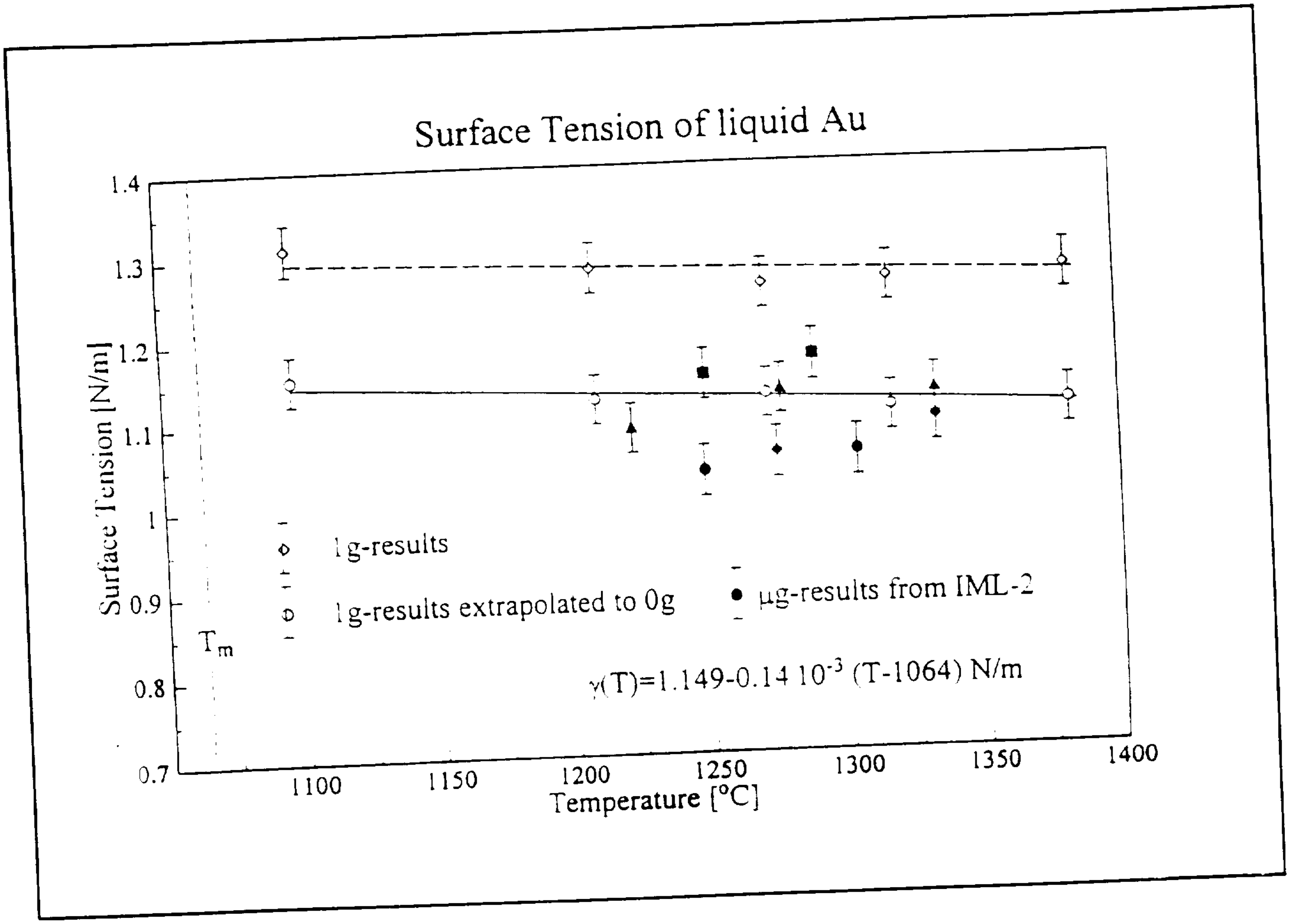

Figure 5: Surface tension data for Au in IML-2 TEMPUS experiment. From [14]

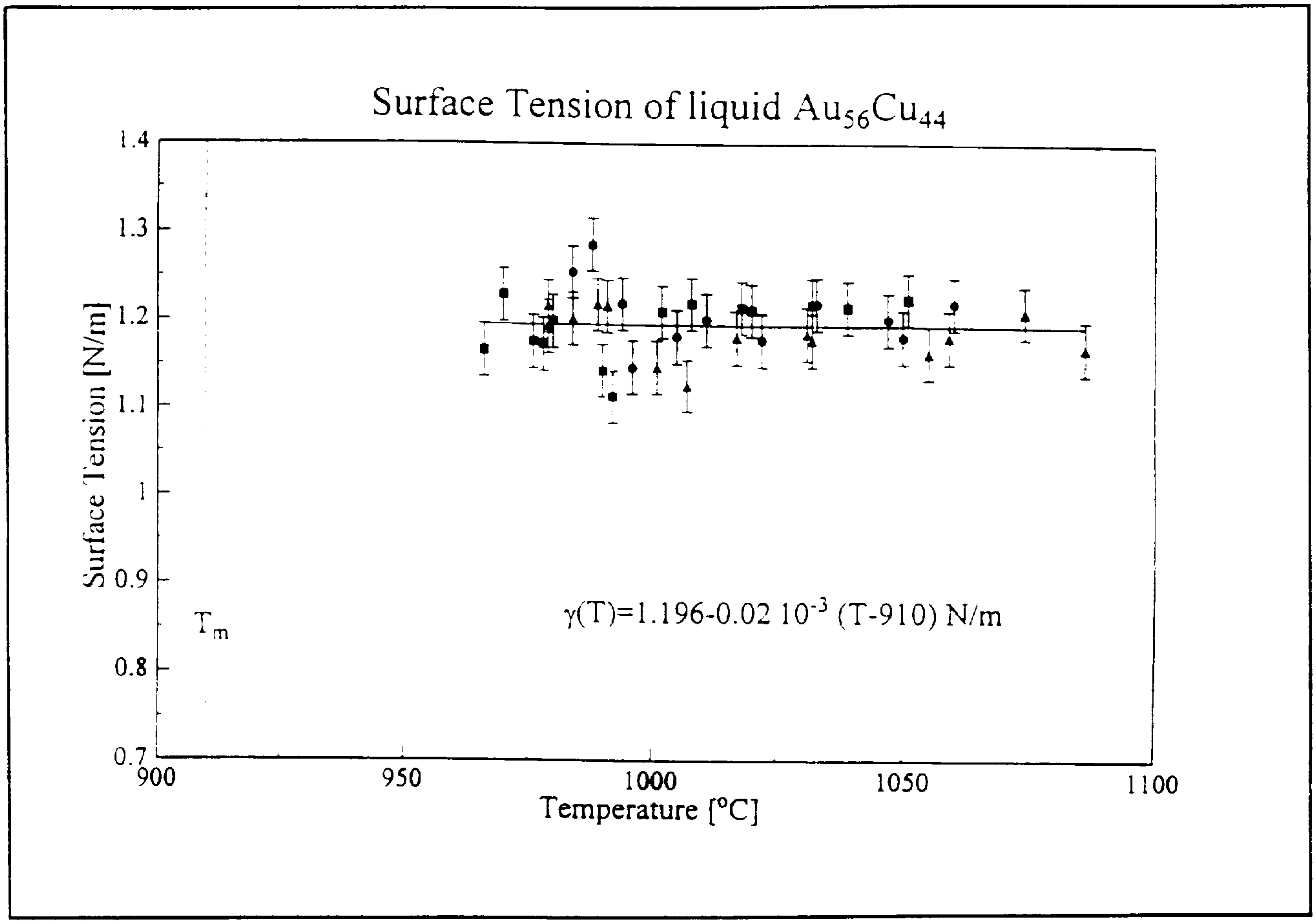

Figure 6: Surface tension data for AuCu in IML-2 TEMPUS experiment. From [14]

The method for measuring viscosity by this oscillating drop technique was successfully demonstrated. The data collection and analysis techniques provided very clear information about the damping of the surface oscillations. However, because of the loss of samples in earlier Functional Objective steps (FO's), it was decided by the TEMPUS team to run only at elevated positioner settings. For reasons discussed in the following section, we believe that the elevated positioning forces caused an increase in fluid flow velocity sufficient to induce turbulent or transitional flow in the sample, and therefore prevent the measurement of a meaningful viscosity. The values measured were too great by an order of magnitude.

IV. Discussion

The surface tension results for Au and AuCu are consistent with the data from the literature, obtained by other methods. The oscillating drop technique has been demonstrated to be accurate and may be used for materials and conditions to which other techniques are not applicable, such as refractory and undercooled metals. Only a containerless process can be used to measure properties of undercooled metals.

The measurement of viscosity by the oscillating droplet technique relies on a number of factors. First, the oscillations must be small enough to allow the linear analysis presented by Lamb [16] to apply. Second, any secondary flows, such as that caused by the magnetic force field, must be laminar, or turbulent damping of the droplet's oscillations will dominate the normal laminar damping of the droplet.

As explained in the introduction, it was to escape these turbulent magnetically driven flows that the viscosity measurements were performed in microgravity. However, because the IML-2 TEMPUS coil design provided much poorer positioning stability than originally believed, the decision was made by the TEMPUS team to use greater positioning forces than originally planned in order to extend the processing life of the remaining samples and of the facility itself. While this decision did allow much of the science of the mission to be salvaged, postflight analysis of the experiment data have determined that the measured value of the viscosity is too great, as would be expected if there were transitional or mildly turbulent flow. This theory will be addressed in the experimental program planned for the reflight of TEMPUS aboard MSL-1.

The key issue then is the exact location of the turbulent transition. This point must be determined as a function of the materials properties and process parameters, i.e. positioner and heater currents. A better understanding of the phenomenon of transition from laminar flow will allow determination of the acceptable limits of operation of the facility for the viscosity experiments. The measurement of this transition is being undertaken as a part of the ground-based preparation for the TEMPUS reflight.

V. Conclusion

In conclusion, we performed very successful surface tension measurements on pure gold and a gold-copper alloy. These measurements are very exciting and important, first because they demonstrate the usefulness and accuracy of this technique that is uniquely applicable to materials systems such as highly reactive metals and undercooled melts for which it was previously not possible to make such thermophysical property measurements. Secondly, the experiments provide experimental verification for the theoretical correction developed by Cummings and Blackburn to account for the magnetic forces necessary to support a droplet in ground-based experiments. Such verification requires the special properties of the microgravity environment, so that the smaller magnetic forces provide a negligible effect on the surface tension measurements.

Unforeseen difficulties with the TEMPUS hardware required operation in a regime that did not allow the measurement of the viscosity of these sample systems. However, the data collection and analysis techniques were successfully demonstrated. The theories that were developed regarding the difficulties with the viscosity measurements are being addressed and the measurements themselves will be repeated under better conditions as a part of the reflight of TEMPUS in spring 1997.

VI. Recommendations

After the mission was completed, almost six months were required before we received copies of the videotapes recorded onboard the Shuttle. This left us with only six months of funding remaining to perform a year's work analyzing the data. We would recommend that a procedure be developed which would allow quicker dissemination of recorded data to the PI's, and also that more time be granted for the analysis of that data.

VII. Publications and Awards

A list of awards received and publications made as a result of this research follows.

AWARDS

J. Szekely: TMS Educator Award, 1991
Alexander von Humboldt-Stiftung Prize, 1991
Fellow, TMS, 1993

E. Schwartz, Ph. D., Dept. of Materials Science and Engineering, Massachusetts Institute of Technology, 1995

Robert Hyers, NASA Graduate Student Researchers Program Fellowship, 1993-1996.

PUBLICATIONS

[1] I. Egry and J. Szekely, "The Measurement of Thermophysical Properties in Microgravity Using Electromagnetic Levitation," Adv. Space Res., Vol. 11, No. 7 pp (7)263 - 6, 1991.

[2] J.H. Zong, J. Szekely, and E. Schwartz, "An Improved Computational Technique for Calculating Electromagnetic Forces and Power Absorption's Generated in Spherical and Deformed Body in Levitation Melting Devices," IEEE Trans., Vol. 28, No. 3, pp. 1833-42, May 1992.

[3] E. Schwartz, J. Szekely, O.J. Ilegbusi, J.H. Zong, and I. Egry, "The Computation of the Electromagnetic Force Fields and Transport Phenomena in Levitated Metallic Droplets in the Microgravity Environment," TMS Int'l Conference on MHD in Process Metallurgy, San Diego, pp. 81-7, March 1992.

[4] J.H. Zong, B.Q. Li, and J. Szekely, "The Electrodynamic and Hydrodynamic Phenomena in Magnetically Levitated Molten Droplets -- I. Steady State Behavior," Acta Astronautica, Vol. 26, No. 6, pp. 435-49, 1992.

[5] E. Schwartz, S. Sauerland, J. Szekely, and I. Egry, "On the shape of Liquid Metal Droplets in Electromagnetic Levitation Experiments,"

Proceedings on Containerless Processing, TMS Conference, Denver, pp. 57-64, 1993.

[6] J.H. Zong, J. Szekely, and G. Lohofer, "Calculations and Experiments Concerning Lifting Force and Power in TEMPUS," Acta Astronautica, Vol. 29, No. 5, pp. 371-8, 1993.

[7] J.H. Zong, B.Q. Li, and J. Szekely, "The Electrodynamic and Hydrodynamic Phenomena in Magnetically Levitated Molten Droplets -- II. Transient Behavior and Heat Transfer Considerations," Acta Astronautica, Vol. 29, No. 4, pp. 305-11, 1993.

[8] E. Schwartz and J. Szekely, "The Shape of Liquid Metal Droplets in Electromagnetic Levitation Experiments: The Conclusion of an Ongoing Study," Proc. on Experimental Methods for Microgravity Materials Science, TMS Annual Meeting, San Francisco, February 1994.

[9] E. Schwartz and J. Szekely, "The Shape of Liquid Metal Droplets in Electromagnetic Levitation Experiments Considering Internal Fluid Flow," Proc. on Experimental Methods for Microgravity Materials Science, pp. 73-9, TMS Annual Meeting, San Francisco, February 1994.

[10] J. Szekely and E. Schwartz, "Some Perspectives on Electromagnetic Levitation in Space Experimentation," Electromagnetic Processing of Materials, ISIJ Symposium Proc., pp. 9-14, Nagoya, October 1994.

[11] E. Schwartz and J. Szekely, "Mathematical Modeling: An Essential Component of the Design of Space Experiments," TMS Int'l. Symposium on Materials Processing in the Computer Age II, pp. 147-61, Las Vegas, 1995.

[12] J. Szekely, E. Schwartz, and R. Hyers, "Electromagnetic Levitation -- A Useful Tool in Microgravity Research," J. of Materials, pp. 50-53, May 1995.

[13] TEAM TEMPUS (I. Egry, ..., J. Szekely, *et al.*), "Containerless Processing in Space: Recent Results", submitted to Journal of Materials.

[14] E. Schwartz, "Measurement of the Surface Tension of Electromagnetically Levitated Droplets in Microgravity", Ph. D. Thesis, Massachusetts Institute of Technology, 1995.

VIII. Other References

[15] J.W.S. Rayleigh, "On the Capillary Phenomena of Jets", Proceedings of the Royal Society of London, 29(1879), 71-97.

[16] H. Lamb, "On the oscillations of a Viscous Liquid Globe", Proceedings of the London Math. Society, 13(1) (1881), 51-66.

[17] D.L. Cummings and D.A. Blackburn, "Oscillations of magnetically levitated aspherical droplets", Journal of Fluid Mechanics, 224 (1991) 395-416.

Appendix: Select Publications