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Surface Tension Measurements of Metallic Melts Under Microgravity[1]

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The surface tension of liquid metals and alloys was measured for the first time in microgravity using the oscillating drop technique. Data for pure gold, a congruently melting gold–copper alloy, and an eutectic zirconium–nickel alloy are presented. We find excellent agreement with available results obtained on Earth by the same technique, but only if the latter are corrected to account for gravity effects. This not only shows the necessity for the correction of the surface tension data derived from Earth-bound oscillating drop experiments, but also proves its correctness.

KEY WORDS: liquid alloys; liquid metals; microgravity; oscillating drop technique; surface tension.

1. INTRODUCTION

The surface tension of liquid metals and alloys is essential from the scientific point of view of validating theories of the liquid state, as well as for practical applications in processing by chemical and metallurgical industries [1]. The knowledge of the surface tension itself is important, e.g., for casting, molding, refining, sintering, and crystal growth operations.

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In addition, the temperature dependence of surface tension leads to Marangoni convection, which makes it important for processes such as welding and crystal growth [2].

The electromagnetic levitation technique is an established method for containerless processing of metallic melts. It avoids sample contamination by the use of high-purity processing gas and the use of contact-free measuring techniques, such as pyrometric temperature measurement. The oscillating drop method uses the oscillations of a levitated drop about its equilibrium shape [3]. The restoring force for these surface oscillations is the surface tension, which can be related to the oscillation frequency [4].

There has been some skepticism about the results obtained by the oscillating drop technique because it tends to yield higher values for the surface tension than those from conventional, noncontactless methods. Although some authors have attributed this to the better purity of the sample's surface, it may also be a systematic error due to the presence of the electromagnetic levitation field, which produces a magnetic pressure at the surface and leads to an apparent increase in the surface tension. Taking this effect into account by a correction formula derived by Cummings and Blackburn [5], it was possible to obtain surface tension values for several materials which are in good agreement with those derived by conventional techniques [6]. This demonstrates the validity of the oscillating drop method and introduces it as the preferred method for determining the surface tension of reactive metals.

Surface tension measurements with the oscillating drop method can be improved by performing the experiments in a microgravity environment, which reduces the residual accelerations on the levitated sample by an order of 10^{-4} from its value in Earth's gravity, since much weaker electromagnetic fields are necessary for positioning the sample.

The experiments were conducted during the Spacelab IML-2 mission in 1994 using the electromagnetic containerless processing facility TEMPUS. This facility was designed to provide independent control of heating and positioning by use of two independent coils [7]. Switching off the heating coil will have two effects: first, the power input into the sample can be greatly reduced, allowing cooling of the sample without a cooling gas; and second, forces on the sample's surface can be drastically reduced.

2. EXPERIMENTS

Using the oscillating drop technique, we measured the frequency of oscillations of levitated droplets about their equilibrium shapes. Rayleigh calculated the frequencies of low-amplitude oscillations of an inviscid spherical liquid droplet of mass M due to the surface tension γ [8]. The relationship between the frequency of the oscillation mode n and the surface tension is given by

$$\nu_n^2 = \frac{1}{3\pi} n(n-1)(n+2) \frac{\gamma}{M} \tag{1}$$

The fundamental frequency ($n = 2$) is called the Rayleigh frequency and is expressed as

$$\nu_R^2 = \frac{8}{3\pi} \frac{\gamma}{M} \tag{2}$$

Under terrestrial levitation conditions, the sample is distorted due to the magnetic field, which has two effects on the sample. First, it deforms the sample and leads to a splitting of the Rayleigh frequency into five frequencies, and second, the field exerts a magnetic pressure on the sample, which leads to a shift of all these frequencies. These magnetic field effects have

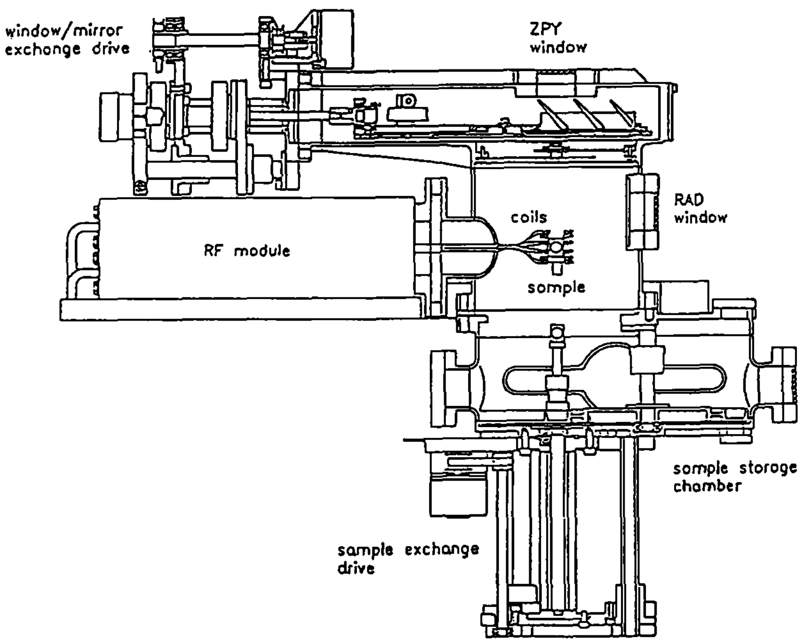

Fig. 1. Schematic diagram of the TEMPUS experimental arrangement showing the major subsystems.

been calculated by Cummings and Blackburn [5]. They derived the following (approximate) correction formula:

$$\omega_R^2 = \frac{1}{5} \sum_{m=+2}^{m=-2} \omega_{2,m}^2 - 1.9\omega_\tau^2 - 0.3\omega_\tau^{-2} \left(\frac{g}{R}\right) \tag{3}$$

where R is the radius of the sample and ω_τ is the mean translational frequency of the droplet's center of mass.

To determine the five frequencies for the $n = 2$ modes, a method of oscillation detection based on inspection of certain geometrical parameters by digital image processing was developed [9].

It was thought that, due to the much weaker magnetic field strength necessary to position a sample in microgravity, the droplet would remain spherical, eliminating the frequency splitting, and that the negligible magnetic pressure on the samples surface will cause no frequency shift. For that, application of the Rayleigh formula, Eq. (2), should be possible.

In the microgravity experiments the solid sample was introduced from below into the two-coil system. After it was held in a stable position by the positioning coil, the sample was melted and overheated by the heating coil. When the heating coil was switched off, the temperature of the sample decreased due to radiation losses. To excite the surface oscillations, a short current pulse of about 0.1 s was applied to the heating coil, which squeezed the sample. The pulsing was repeated every 5 s until the droplet solidified to make surface tension measurements over a wide temperature range. After solidification, the sample was melted again and the procedure was repeated for as many cycles as possible. The oscillations were observed by a high-speed video camera with a sampling rate of 120 Hz from the top and from the side and the films were analyzed off-line after the mission. With our image processing system, it was possible to obtain a time signal showing the variation of the visible area of the sample's surface (Fig. 2). A frequency spectrum was obtained using the 5-s time interval between the heating pulses and performing a 512-point Fourier tansform. It can be seen from Fig. 3 that in microgravity experiments the splitting of the $n = 2$ mode did not occur. There is only one oscillation peak visible, and for evaluation of the surface tension this frequency is set into the Rayleigh formula, Eq. (2).

Using the method described above, we measured the surface tension of liquid pure gold, the congruently melting alloy Au–44 at% Cu, and the eutectic alloy Zr–36 at% Ni. Temperature measurements were performed from the top of the sample using a two-color broadband pyrometer which was equipped with an InAs detector and had a sampling rate of 100 Hz.

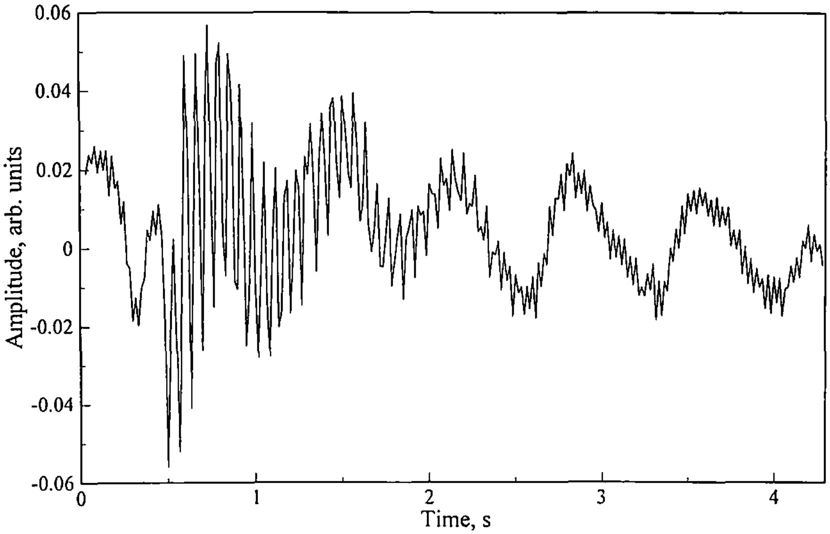

Fig. 2. Time signal showing variation of the visible area of the sample's surface after stimulation of the oscillation.

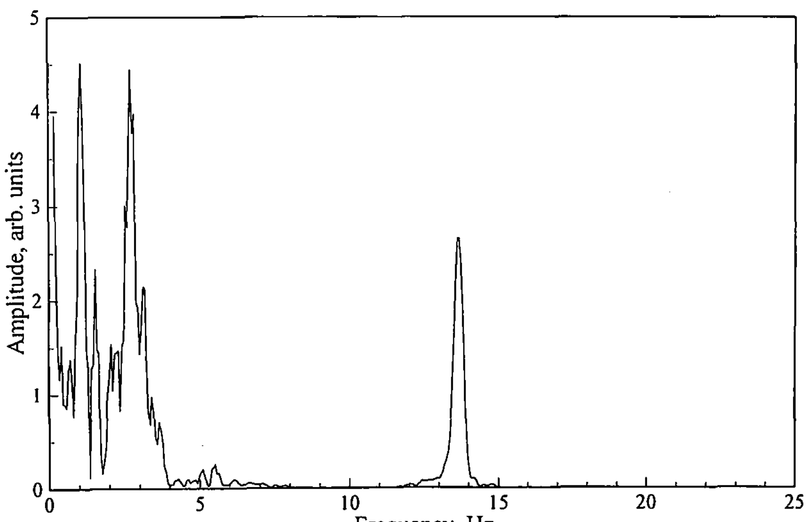

Fig. 3. Frequency spectrum of the surface oscillations of liquid gold. The peaks in the lower-frequency region show the translational oscillations and the peak at 14 Hz is the surface oscillation peak used to evaluate the surface tension.

3. RESULTS

3.1. Gold

The surface tension of a liquid gold sample of mass 5.21 g was measured in the temperature range 1225–1330°C. The filled symbols in Fig. 4 were obtained from the microgravity experiments and evaluated according to Eq. (2). For comparison, data obtained by Sauerland [9] in Earth-bound experiments on a gold sample of mass 0.72 g are given. The open squares indicate the uncorrected 1-g (Earth-bound) data points evaluated according to Eq. (2), whereas the open circles stand for the corresponding corrected values evaluated according to the Cummings formula, Eq. (3). A least-squares fit to the microgravity and the corrected 1-g data points leads to the surface tension–temperature relationship

$$\gamma(T) = 1.149 - 0.14 \times 10^{-3}(T - 1064) \tag{4}$$

where $\gamma(T)$ is in $N \cdot m^{-1}$ and T is in °C.

From Fig. 4 it can be seen that the agreement of the microgravity results with the corrected 1-g results is very good, whereas the neglect of the magnetic pressure in Earth-bound experiments leads to distinctly higher values.

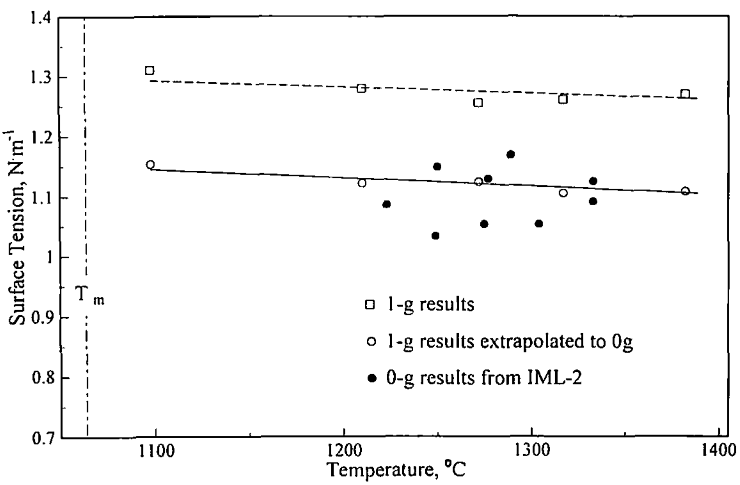

Fig. 4. Surface tension data for liquid gold. The filled symbols show the microgravity data, the open squares indicate the results of Earth-bound experiments evaluated according to Rayleigh, and the open circles indicate the corresponding values according to Cummings and Blackburn [5].

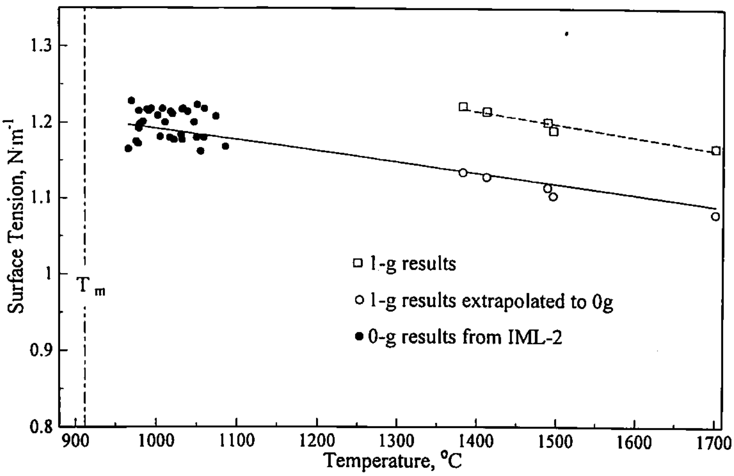

Fig. 5. Surface tension data for liquid $Au_{56}Cu_{44}$. The filled symbols show the microgravity data, the open squares indicate the results of Earth-bound experiments evaluated according to Rayleigh, and the open circles indicate the corresponding values according to Cummings and Blackburn [5].

The value of surface tension at the melting point of 1.149 $N \cdot m^{-1}$ obtained in this study compares remarkably well with the mean value for the surface tension of liquid gold, 1.145 $N \cdot m^{-1}$, published by Keene in his review article [10].

3.2. Au–44 at% Cu Alloy

The surface tension of liquid $Au_{56}Cu_{44}$ was measured in the temperature range 970–1080°C using a sample of mass 4.21 g. The filled symbols in Fig. 5 indicate the microgravity results according to Eq. (2). The open squares show some data points of an Earth-bound experiment with a sample mass of 0.35 g evaluated according to the Rayleigh formula, Eq. (2), and the open circles indicate the corresponding values with consideration of the magnetic pressure. Due to differences between the facilities, the experiments were carried out at two different temperature ranges and a least-squares fit to the microgravity and the corrected 1-g data points leads to the following temperature dependence of the surface tension (in $N \cdot m^{-1}$):

$$\gamma(T) = 1.205 - 0.15 \times 10^{-3}(T - 910) \tag{5}$$

The experimental data of Gallois and Lupis [11] obtained with the sessile drop technique on this alloy suggest a value of 1.12 $N \cdot m^{-1}$ at a temperature of 1108°C, which is in good agreement with the result of this study.

3.3. Zr–36 at% Ni Alloy

The surface tension of liquid $Zr_{64}Ni_{36}$ was measured in the temperature range 980–1150°C using a sample of mass 1.94 g. Figure 6 shows the data points and a least-squares fit to these points gives the following temperature dependence of the surface tension (in $N \cdot m^{-1}$):

$$\gamma(T) = 1.545 + 0.08 \times 10^{-3}(T - 1010) \qquad (6)$$

Our microgravity experiment has been the first experiment on this eutectic alloy, for which there are no data available on the surface tension. For the pure components, Keene [10] cites surface tension values of 1.796 $N \cdot m^{-1}$ for nickel and 1.430 $N \cdot m^{-1}$ for zirconium at their melting points. Based on the lower surface tension, zirconium should be surface active in nickel. Since the solubility of oxygen in zirconium is high, surface contamination should not seriously affect the surface tension data. The small positive temperature coefficient found in our experiments may also be explained through the strong interaction between nickel and zirconium.

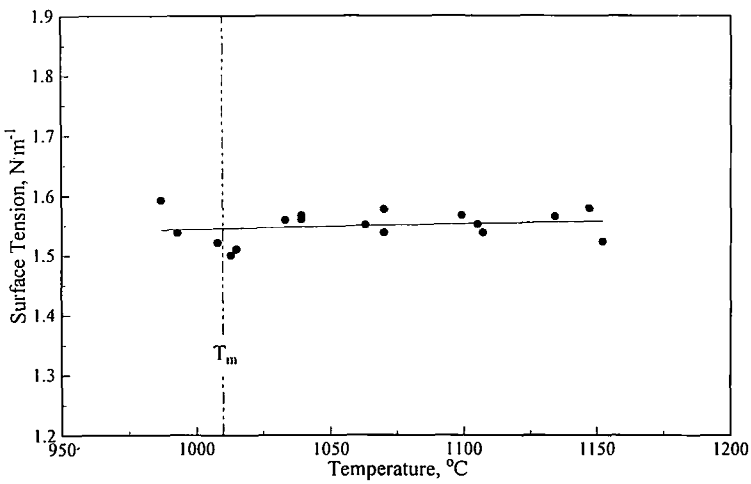

Fig. 6. Surface tension data for liquid $Zr_{64}Ni_{36}$. The filled symbols show the microgravity data.

4. CONCLUSIONS

For the first time, surface tension measurements with the oscillating drop technique have been performed in microgravity. As sample materials a pure metal, a congruently melting alloy, and an eutectic alloy were used. The accuracy of these results was shown by comparison with prior experimental results. We have demonstrated the advantages and unique possibilities offered by microgravity experimentation.

It was shown that in microgravity, in contrast to Earth-bound conditions, the magnetic pressure of the levitation field on the sample is very small and can be neglected, whereas for levitation experiments under 1-g the correction by Cummings and Blackburn [5] has to be used. The agreement of our microgravity results with previous data obtained by conventional methods and the corrected data of earthbound levitation experiments shows that the correction formula accurately accounts for the effects of the magnetic field and the gravity.

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