

# Thermophysical Property Measurements in Microgravity: Chances and Challenges<sup>1</sup>

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The microgravity environment offers considerable advantages for the measurement of thermophysical properties, in particular, for high-temperature metallic melts. The absence of containers and of convection are the two major benefits. This paper reviews past microgravity experiments dealing with thermophysical property measurements and discusses the methods used. An outlook into the space station era is also given with special emphasis on chances and challenges.

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**KEY WORDS:** containerless processing; liquid metals; microgravity; thermophysical properties.

## 1. INTRODUCTION

A thermodynamic system tends to maintain spatial homogeneity. Therefore, gradients in intensive variables cause heat or mass flows which attempt to eliminate these gradients. In linear irreversible thermodynamics, the flows are linearly related to the gradients [1]:

$$\begin{pmatrix} \vec{j}_c \\ \vec{j}_Q \end{pmatrix} = - \begin{pmatrix} D & S \\ P & \lambda \end{pmatrix} \begin{pmatrix} \vec{\nabla}c \\ \vec{\nabla}T \end{pmatrix} \quad (1)$$

If  $S$  and  $P$  were zero, we would have two independent laws, namely Fick's law:  $j_c = -D \nabla c$ , defining the diffusion constant  $D$ , and Fourier's law:  $j_Q = -\lambda \nabla T$ , defining the thermal conductivity  $\lambda$ . The off-diagonal elements

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are usually very small. The existence of  $S \neq 0$  is called the Soret effect; it describes thermomigration, i.e., a mass flow caused by a temperature gradient, while  $P$  describes a heat flow caused by a concentration gradient.

The measurement of these and other thermophysical properties of the liquid phase, in particular, at high temperatures, is a very difficult task under terrestrial conditions. There are two major sources for these difficulties and the subsequent errors: chemical reactions with the container or substrate and the influence of convection.

In this review we shall discuss how the microgravity environment can be used for precise measurement of thermodynamic properties of metallic melts. To take full advantage of the microgravity environment, novel and unconventional techniques have to be developed. In some cases, these measurements are only possible in microgravity; in others, the microgravity environment improves the accuracy considerably [2]. The absence of convection is the essential benefit for measurements of transport properties, while containerless processing is the key technology for the study of high-temperature and undercooled melts.

Most past microgravity experiments suffered from the restricted access to the experiment and the limited time available for the execution of the experiment. The capability for real-time adjustment of the setup and for repetitive experiment series was very limited. With the advent of the International Space Station (ISS) and the development of new advanced facilities for thermophysical property measurements on-board the ISS, some of these problems may be alleviated.

## 2. THERMOPHYSICAL PROPERTY MEASUREMENTS

### 2.1. Container Methods

The methods described in this chapter require a container, generally a crucible. The main reason is that a temperature gradient has to be applied which, in the case of free surfaces, would lead to Marangoni convection [3], thereby destroying the convection-free environment provided by microgravity.

#### 2.1.1. Thermal Conductivity

The main difficulty in performing accurate measurements of thermal conductivity or thermal diffusivity of liquids lies in the separation of the conduction process from convective effects. Transient methods have been used quite successfully, based on the fact that the characteristic time for the acceleration of the fluid by buoyancy forces is much longer than the propagation time of the temperature change caused by a strongly localized

temperature gradient. Therefore, it is possible by a suitable choice of the experimental conditions and geometrical design of the measuring cells to perform, in principle, convection-free measurements even on ground. Microgravity experiments can be used to further reduce convection and to extend the convection-free time regime.

The first measurements of the thermal conductivity in microgravity have been performed by Hibiya et al. [4] on InSb. They applied the transient hot-wire (THW) method during a sounding rocket flight, and in a drop tower experiment. In this method the temperature increase in a wire, generated by a step function-like heat input at time zero, varies linearly with  $\ln t$ ,

$$\Delta T = \frac{q}{4\pi\lambda_{\text{ave}}} \ln t + C \quad (2)$$

where  $q$  is the heat input per unit length and  $\lambda_{\text{ave}}$  is the average thermal conductivity of the liquid and substrate. The constant slope of  $\Delta T$  versus  $\ln t$  permits the calculation of the thermal conductivity of the melt. If convection is present, the experimental temperature rise will not vary linearly with  $\ln t$ , and the calculated apparent thermal conductivity will increase with time. The onset of this deviation depends on both the material studied and the geometrical setup, as shown schematically in Fig. 1.

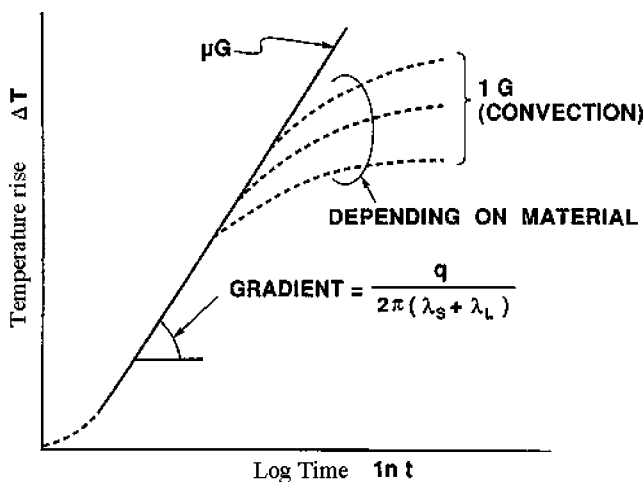


Fig. 1. Influence of convection on thermal conductivity measurements (Nakamura et al. [4]).

### 2.1.2. Diffusion

As in the case of thermal conductivity, diffusion measurements in microgravity exploit the absence of convection. Whenever there is convection, diffusion measurements are prone to large errors, because convection provides more effective mass transport than diffusion. In simple one-dimensional (1-d) geometries the apparent convective contribution to diffusion can be estimated from a fluid flow calculation [5]. Diffusion measurements are carried out in a container, usually a shear cell. Microgravity experiments on the self diffusion of tin isotopes and lead were carried out by Frohberg [6]. As an example, his data on lead are shown in Fig. 2. He found that the diffusion coefficients as measured in space are 20 to 40% lower than the values obtained from terrestrial experiments. The data are best fitted with a quadratic power law  $D = KT^2$ , which is in contradiction to the Arrhenius type behavior expected for an activated process. However, fluctuation theory [7] and mode coupling theory [8] both predict power law behavior, leading to the conclusion that diffusion may not be an activated process, but rather a collective phenomenon. Following Frohberg's pioneering work, measurements of the diffusion coefficient under microgravity have become quite popular, both in the U.S. [9] and in Japan [10].

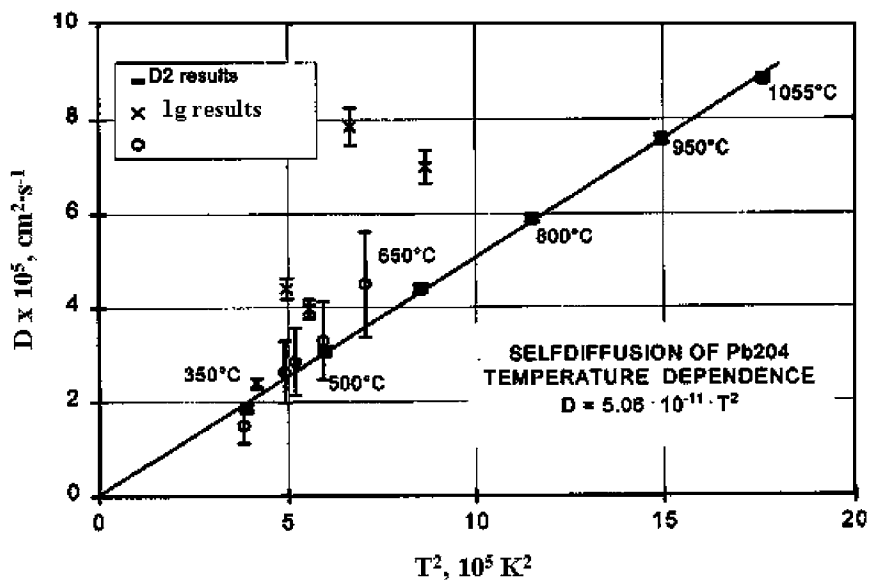


Fig. 2. Self-diffusion coefficient of  $^{204}\text{Pb}$ , measured during the Spacelab mission D2, and terrestrial data for comparison (Frohberg [6]).

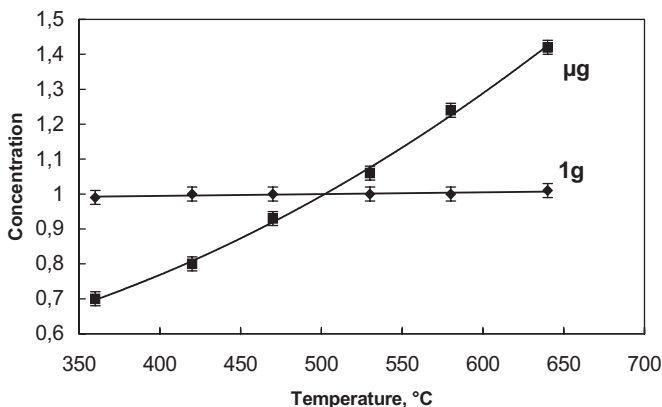


Fig. 3. Soret effect of tin in cobalt at 500°C (Malméjac and Froberg [11]).

Under microgravity, the Soret coefficient  $S$  was determined by Malméjac and coworkers [11] for cobalt in tin by using a shear-cell technique. Applying a temperature difference of  $200 \text{ K} \cdot \text{cm}^{-1}$ , they found a relative concentration difference of nearly 50%, whereas the same experiment performed on ground yielded a homogeneous sample, i.e., 0% concentration difference. This is shown in Fig. 3.

## 2.2. Containerless Methods

Thermophysical properties of high temperature, and highly reactive, melts can be conveniently measured by containerless methods, such as electromagnetic or electro-static levitation. These methods provide the purest environment possible. Since the surface of the liquid sample is not in contact with a wall, Marangoni convection will occur, if there is a temperature or concentration gradient along the surface. Therefore, containerless techniques are only applicable to those problems where convection plays no role. This is the case for all measurements to be discussed in the following.

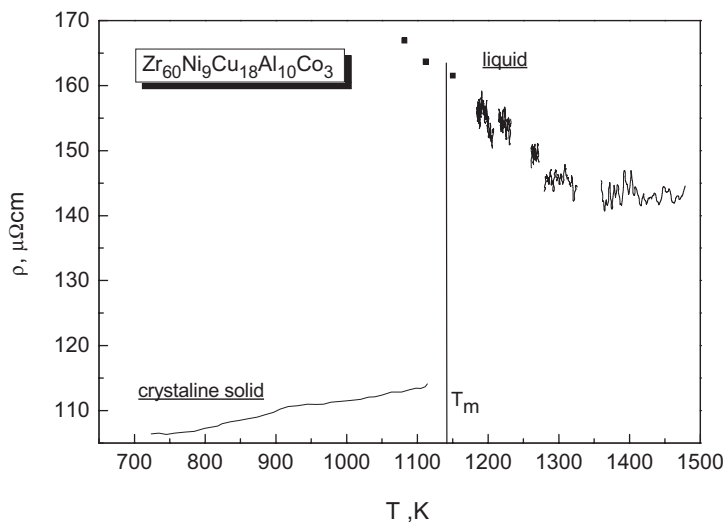
Both techniques, electrostatic as well as electromagnetic levitation, can be applied for thermophysical property measurements on ground and in space, the advantage of the microgravity environment being for both methods that the strong lifting forces required on earth can be reduced to small positioning forces, thereby minimizing any detrimental side effects of the electromagnetic or electrostatic levitation fields. Whereas electromagnetic and electrostatic levitation has been used quite successfully on ground [12, 13], only electromagnetic levitation has a microgravity record so far.

Thermophysical properties of liquid metals have been measured in microgravity during two Spacelab missions, using the electromagnetic levitation facility TEMPUS [14, 15].

### 2.2.1. Electrical Conductivity

It is possible to measure the electrical conductivity of levitated melts using a non-contact, inductive method. The impedance of a coil surrounding the sample is influenced by its electrical conductivity. For spherical samples and homogeneous magnetic fields, as realized in microgravity, this relation is rather simple [16]. Under terrestrial conditions, much mathematical and engineering effort must be spent to extract the required information from the measured impedance. The mathematical effort consists of taking into account the nonsphericity of the sample and the non-homogeneity of the electromagnetic field, whereas engineering effort is needed to measure very small voltage changes in a low current detector circuit in the presence of strong levitation fields. The solution to this latter problem is the use of a pulsed levitation field, allowing for impedance measurements while the field is switched off [17]. The complex impedance can then be determined by measuring both current  $I$  and voltage  $U$ , simultaneously.

Figure 4 shows the results of a microgravity experiment [15] on the metallic glass forming system  $\text{Zr}_{60}\text{Ni}_9\text{Cu}_{18}\text{Al}_{10}\text{Co}_3$ . No terrestrial data are



**Fig. 4.** Electrical resistivity of a Zr-Ni-Cu-Al-Co alloy as a function of temperature, not corrected for thermal expansion (Lohöfer and Egry [16]).

available for comparison. The microgravity data show a decrease in resistivity as a function of temperature in the liquid phase. This can be attributed to the break-up of clusters with increasing temperature. It should be noted, however, that this method yields the resistance, not the specific resistivity. This means that the temperature dependence of the electrical resistivity (or conductivity) can be obtained only if the thermal expansion is known and taken into account.

The electrical conductivity  $\sigma$  is of interest on its own, but it also can be used to obtain the thermal conductivity  $\lambda$  through the Wiedemann–Franz relation, which is known to hold well for liquid metals [18].

$$\lambda = L\sigma T \quad (3)$$

Here,  $L$  is a universal constant, the so called Lorenz number,  $L = 2.44 \text{ W} \cdot \Omega \cdot \text{K}^{-2}$ . Thus, electrical conductivity measurements provide an alternative way to determine the thermal conductivity with the extra advantage that the result is free of convective effects and therefore more accurate.

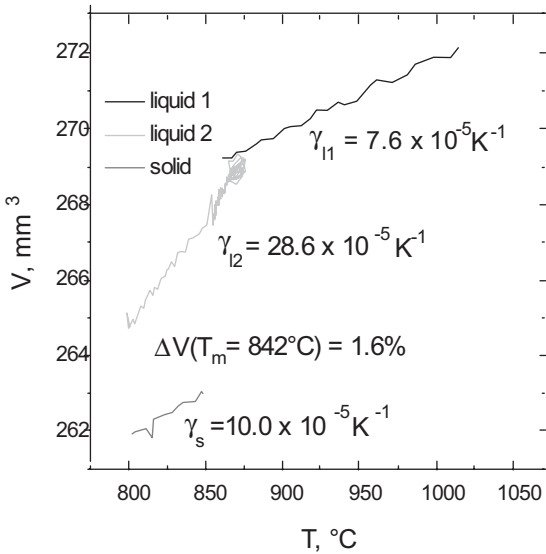
### 2.2.2. Density and Thermal Expansion

The density and thermal expansion of levitated drops are determined by recording the visible cross section of the sample. Assuming rotational symmetry, the volume is calculated. Since the mass of the sample is known and does not change, this yields the density. Typically, a resolution of  $\Delta V/V = 10^{-4}$  is required. This can be achieved using sub-pixel algorithms for edge detection, curve fitting of the shape, and statistical averaging [19, 20].

Although such measurements can be performed on ground [19], in microgravity, the precision of the data is improved and the accessible temperature range is extended. As an example, results of a microgravity experiment [20] on the thermal expansion of a glass-forming Zr-based alloy are shown in Fig. 5.

### 2.2.3. Specific Heat

A noncontact method developed by Fecht and Johnson [21] can be used to determine the specific heat in levitation experiments. It is a variant of modulation calorimetry, normally used in low-temperature physics. The heater power is modulated according to  $P_{\omega}(t) = \Delta P_{\omega} \cos(\omega t)$  resulting in a modulated temperature response  $\Delta T_{\omega}$  of the sample. If the internal relaxation, i.e., the time needed to achieve a stationary temperature distribution inside the sample, is faster than the external relaxation, i.e., the heat loss to



**Fig. 5.** Thermal expansion of  $Zr_{11}Cu_{47}Ti_{34}Ni_8$  in the solid and liquid states. The phase designated as liquid 2 might correspond to a solid-liquid two-phase region (Damaschke et al. [20]).

the environment, the modulation frequency  $\omega$  can be chosen such that a simple relation for the temperature variation is obtained:

$$c_p = \Delta P_\omega / (\omega \Delta T_\omega) \quad (4)$$

from which the specific heat,  $c_p$ , can be determined.

Due to the small positioning forces, a microgravity experiment can be performed in vacuo where the heat loss is due to radiation only and is therefore slow. A typical temperature signal recorded during the MSL-1 Spacelab mission [15], showing the temperature oscillations, including the undercooled regime, is shown in Fig. 6. From the figure  $\Delta T_\omega$  can be determined and plugged into Eq. (4) to obtain the specific heat.

On ground, the levitated sample must be cooled in a stream of inert gas, due to heat input caused by the large levitation fields necessary to overcome terrestrial gravity. This is a fast process, and Eq. (4) no longer holds. Therefore, this elegant method cannot be applied on ground.

#### 2.2.4. Viscosity and Surface Tension

Viscosity and surface tension are conveniently measured by the oscillating drop technique [22]. Liquid samples perform oscillations around



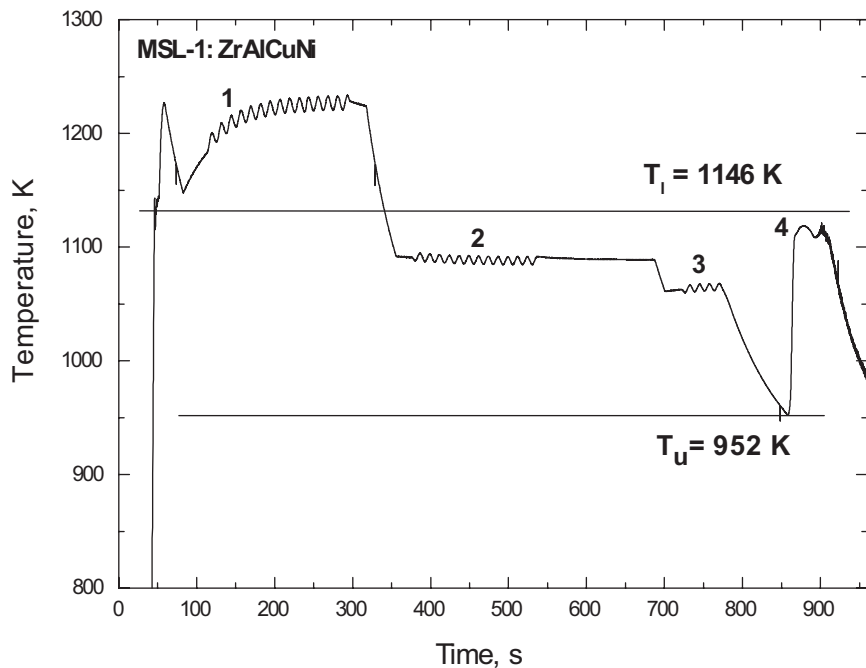


Fig. 6. Temperature response of a liquid Zr-Al-Cu-Ni alloy after power modulation at different temperatures. Regions 2 and 3 correspond to undercooled melts and point 4 shows the recalescence at solidification (Fecht et al. [15]).

their equilibrium shape. In microgravity, this is a sphere and in that case, simple formulae can be used to relate frequency  $\omega$  and damping  $\Gamma$  of the oscillations to surface tension  $\gamma$  and viscosity  $\eta$ , respectively. They can be expressed as

$$\omega^2 = \frac{32\pi}{3} \frac{\gamma}{M} \quad (5)$$

and

$$\Gamma = \frac{20\pi}{3} \frac{R_0\eta}{M} \quad (6)$$

where  $M$  is the mass of the droplet and  $R_0$  is its radius. The presence of the gravitational field distorts the sample shape and, consequently, the oscillation spectrum, which makes a quantitative analysis difficult. In Fig. 7, oscillation spectra of a gold-copper alloy are shown, recorded on ground

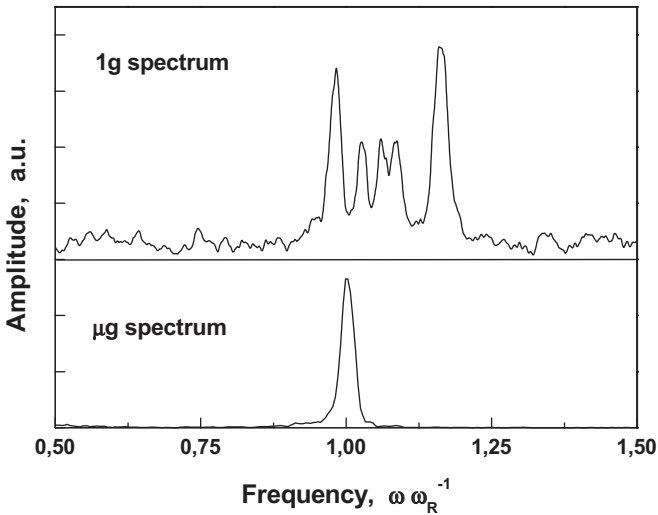


Fig. 7. Frequency spectrum of an oscillating AuCu drop under 1 g (top) and microgravity (bottom).

and in microgravity. As can be seen, both a splitting of the single frequency into five peaks and a shift to higher frequencies occur. The first effect can be attributed to the breaking of the spherical symmetry, while the latter is due to the magnetic pressure acting on the sample surface. By breaking the spherical symmetry, the degeneracy of the Rayleigh frequency, Eq. (5), is lifted. The magnetic pressure, present at the sample's surface, acts as an additional surface tension, because surface oscillations must perform work against this pressure. This leads to an apparent increase of the surface tension and must always be corrected in terrestrial measurements to avoid systematic errors in using the oscillating drop technique. Cummings and Blackburn [23] have developed such a theory, and Fig. 7 is the experimental verification of their prediction.

### 3. SPACE STATION ERA

#### 3.1. Hardware Design

In the past, thermophysical property measurements were only a minor portion of materials-science-related microgravity experiments. This will change in the future. The potential of microgravity for thermophysical property measurements and, *vice versa*, the potential of thermophysical property measurements for microgravity and, in particular, Space Station

utilization, is now fully recognized. One of the outstanding Microgravity Application Promotion programs of ESA, THERMOLAB, is entirely devoted to high precision thermophysical property data of industrially relevant alloys. Consequently, the hardware necessary to perform such experiments is presently being developed. On the one hand, this is Advanced TITUS [24], a German payload, which is based on a tubular isothermal furnace and which shall be capable of performing experiments on contained samples, like, e.g., diffusion experiments. It is planned to accommodate this facility in the Russian part of the Space Station. On the other hand, an electromagnetic levitator is under development as part of the Materials Science Laboratory. This facility, MSL-EML [25], is a joint European-German project, and is based on the TEMPUS Spacelab facility. It will be equipped with state-of-the-art noncontact diagnostic tools, like pyrometers, high-speed and high-resolution video cameras, and an infrared radiometer. MSL-EML can be used to perform the kind of experiments described above. In addition, owing to its modular concept, new experimental concepts can be implemented at a later stage. MSL-EML will be accommodated in the European COLUMBUS module and shall be operational, according to the present schedule, in 2007.

### 3.2. Chances and Challenges

With the advent of the Space Station one big bottle neck, namely operational time, in microgravity, will be removed. This is the chance for performing systematic investigations and parametric studies, like, e.g., the dependence of the surface tension on oxygen partial pressure. The ultimate goal is to use the Space Station routinely for benchmark experiments.

The challenge lies in the effective use of this precious resource. In the past, microgravity experiments had to work the very first time, which unfortunately, but not unexpectedly, was not always the case. It should now become possible to optimize an experiment iteratively, by modifying both the procedure and the hardware, just like in ordinary laboratory physics. Although experiment time should now be more easily available, other resources are not, like crew time, video downlink, and mass up- and down-load. The limited crew time and real-time video imply that experiments must either become autonomous or must be controlled from ground by telescience tools. In comparison to other types of experiments, like crystal growth, thermophysical property measurements mainly produce data and do not require download of microgravity processed samples. In addition, one single sample can be used for many different experiments. Therefore, synergetic effects between different investigator teams are to be expected and must be supported by the facility.

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