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Electromagnetic Levitation Part II: Thermophysical Property Measurements in Terrestrial Conditions

Sayavur I. Bakhtiyarov[1] **and Dennis A. Siginer**[2]

1 Introduction

Undercooling is a natural extension in a levitation environment due to the elimination of container induced nucleation giving access to a metastable region of the phase diagram. It is important to solidification processes, and therefore, the knowledge of thermophysical properties in this region is crucial. The degree of undercooling determines the growth velocity, and is essential in selecting the metastable solid phase. The use of hydrogen gas reduces and removes heterogeneous nucleation catalysts (oxide and sulfide inclusions), and facilitates undercooling.

EML has the following advantages compared to other levitation methods:

- Contactless melting of the sample; elimination of container walls is important in studies of flow dynamics, glass formation, preparation of pure substances, undercooling and solidification.
- Molten sample can be protected from oxidation by a vacuum or an inert gas.
- It is feasible to solidify the sample in levitated position.
- Electromagnetic stirring occurs during the melting process.
- Equilibrium in gas-liquid metal systems is quickly achieved due to efficient stirring and the relatively large surface exposed to the gas phase.
- Alloying additions can be made to the levitated molten sample.

However, broadly speaking the relatively low electrical heating efficiency of induction coils and the limited amount of the specimen are discouraging factors in the use of EML on a large commercial scale. Another issue is the introduction of impurities which can occur through reaction with the gaseous environment during melting. Therefore using very pure gas in the process is a strict requirement. The greatest difficulty in EML is keeping the suspended droplet stable. There are two kinds of instabilities, global and local to the surface. Global instabilities cause the metal to move as a whole. They can be avoided by properly choosing the external current distribution that creates the supporting field. That can be realized by arranging axisymmetric coils with counter-windings at the top. However, Lorentz force along the axis of symmetry is zero, and surface tension is the only force to balance the hydrostatic pressure. As a result, the droplet has a conical shape with its apex pointing downwards. Surface instabilities do not allow levitating samples of large masses. Another disadvantage results from difficulties of temperature control and measurement. Only pyrometric methods can be used; either the emissivity of the material must be known or reliance has to be placed on measurements using multicolor pyrometers. In the past due to difficulties in stabilizing and controlling the liquid sample, a number of researchers considered the levitation melting industrially unsuitable as a production method and discontinued work on further developing it. However, the levitation melting method has been recognized as a useful small-scale laboratory melting technique. Liquid droplets with a mass of up to 50g

[1] Department of Mechanical Engineering, New Mexico Institute of Mining and Technology, Socorro, NM 87801-4796 USA
[2] Department of Mechanical Engineering, Wichita State University, Wichita, KS 67230-0133, USA

have been levitated by axisymmetric coil design. Non-axisymmetric coil design allows levitating much bigger samples, for example Sagardia and Segsworth (1977) levitated 1 kg of aluminum using multi-frequency non-axisymmetric coils.

2 Thermophysical Property Measurements

Accurate and reliable values for the thermophysical properties of molten metals become increasingly significant as new casting techniques are developed and progress is made in numerical modeling of these processes. At high temperatures chemical reactions can take place at the interface between liquid sample and crucible leading to the contamination of the test sample. EML offers the following advantages in the study of the thermophysical properties of liquid metals:

- Container effect is eliminated.
- High temperatures (up to 2400°C) can be achieved.
- Undercooled state can be maintained for an extended period of time.
- Non-contact diagnostic techniques (temperature measurements, imaging analysis, etc.) can be applied.

However, ground based EML also has some limitations in measuring the thermophysical properties of molten metals:

- Electromagnetic fields with high flux density deform the shape of the specimen.
- High frequency electromagnetic fields induce turbulent currents inside the sample.
- Convective cooling by high-purity inert gases is required.
- Convection is present within the molten metal.

2.1 Specific Heat

Specific heat is traditionally measured by adiabatic calorimetry techniques. The calorimeter is isolated from the environment and requires long relaxation times. However, this method cannot be used for non-contact measurements at high temperatures. Betz and Frohberg (1980) proposed a method to determine the enthalpy, the heat, and entropy of fusion for some refractory metals. In this method, the levitation fields are switched off when the desired temperature is reached. Then the sample is dropped into a copper block, and the temperature rise ΔT of the copper block is measured. Given the specific heat C_p of the copper block, the enthalpy loss ΔH of the liquid specimen and the specific heat of the sample can be determined,

$$\Delta H = C_p \Delta T, \quad c_p = \frac{\partial H}{\partial T}. \tag{1}$$

Betz and Frohberg (1980) also measured the heat of mixing by alloying two liquid metals during levitation and detecting the related change in temperature. The enthalpy of molybdenum was measured between 2282 and 3383 K by levitation calorimetry. Special attention was paid to the accuracy of optical temperature measurements ($\pm$ 1.5%) and to the evaluation of heat losses from the sample during the fall. But, comparison of experimental results with data from literature is not encouraging and shows rather large discrepancies. Ohsaka et al. (1992) measured the specific heat of undercooled liquid metals using the same technique. The sample temperature is monitored by an optical pyrometer and is varied by controlling the flow rate of the cooling gas. The measured enthalpy is translated into the specific heat. The specific heats of Al and a $Ti_{60}Cr_{40}$ alloy are determined in this work. Maximum undercooling levels reached are less than those expected under microgravity conditions suggesting that measurements can be extended to a deeper undercooling level if similar experiments were performed in a microgravity environment.

The AC temperature technique requires shorter relaxation times and it can be used for non-contact calorimetry. Fecht and Johnson (1991) applied a non-contact AC pulse heating method for specific heat measurements in molten metals and alloys in an electromagnetic levitation device. The method is a variant of non-contact modulation calorimetry. The power of the heater is modulated sinusoidally,

$$P_\omega(t) = P_{\omega 0}\cos^2\left(\frac{\omega t}{2}\right). \quad (2)$$

The power modulations result in a modulated temperature response (ΔT_ω) of the sample,

$$(\Delta T)_\omega = \frac{P_{\omega o}}{2\omega c_p}\left[1+(\omega\tau_1)^{-2}+(\omega\tau_2)^2\right]^{-\frac{1}{2}}, \quad (3)$$

in which τ_1 and τ_2 are external and internal relaxation times, respectively. The choice for the modulation frequency ω should satisfy the inequality,

$$\tau_2 \ll \frac{1}{\omega} \ll \tau_1. \quad (4)$$

Then, the specific heat c_p is defined by the following expression,

$$c_p = \frac{P_{\omega 0}}{2\omega(\Delta T_\omega)}. \quad (5)$$

The proposed method has been applied to estimate the specific heat of Zr-alloys. It is shown that for droplet diameters of the order of 5-10 mm, the heat capacity data can be obtained with an accuracy of 1%. Near the melting point the value of the specific heat for most pure metals is around 30 J K^{-1} mol^{-1}, close to the crystal specific heat value at the melting point,

$$c_p = c_v + c_d + c_{e.p.d.}, \quad (6)$$

where c_v, c_d, and $c_{e.p.d.}$ represent the vibrational, dilatational and the equilibrium point defects contributions, respectively. Above the melting point specific heat decreases with temperature, reaches its minimum value, and then increases up to 8-0.33 c_v. Temperature dependence of alloys is similar to that of pure metals, and Neumann-Kopp rule can be applied to estimate the additive values, Herlach et al. (1993).

2.2 Emissivity

The spectral normal emissivity ε as a function of wavelength λ and temperature T is determined from the following expression,

$$I_r(\lambda,T) = \varepsilon(\lambda,T)\, I_b(\lambda,T), \quad (7)$$

where I_r and I_b are the spectral radiance of the real and blackbody, respectively. Hansen et al. (1989) combined the electromagnetic levitation technique with ellipsometry to measure the emissivity in liquid metals. The spectral emissivity is calculated by Kirchhoff's law,

$$r(\lambda,T) + \varepsilon(\lambda,T) = 1, \qquad r(\lambda,T) = \frac{(n-1)^2+\kappa^2}{(n+1)^2+\kappa^2} \quad (8)$$

where $r(\lambda,T)$ is the spectral reflectivity, and n and κ are the real and imaginary parts of the complex spectral refractive index $\tilde{n} = n - i\kappa$. Using this method Hansen et al. (1989) measured the emissivity of liquid Co, Ag, Au, Ni, Pd, and Pt as a function of temperature and wavelength. Rhim and Paradis (2001) propose using a high-power laser beam to apply a torque to electromagnetically levitated samples in a vacuum. The torque can be used to control the rotation rate of the sample in measuring its properties. The magnitude of the torque which depends on the power of the laser beam, the fraction of incident photons absorbed, and the size of the moment arm can be estimated by applying the laser to a spherical sample of mass m and radius R. Assuming that absorption and emission of radiation by the sample is governed by the Stefan-Boltzman law, the torque is determined as

$$\tau = \frac{4\pi R^2\sigma\varepsilon a T^4}{c}, \quad (9)$$

where σ, a, c and T represent the Stefan-Boltzman constant, the moment arm, the speed of light and the steady-state temperature of the sample if the same laser beam is used to heat the sample. The angular acceleration of the sample is estimated by,

$$f = \frac{5\sigma\varepsilon a T^4}{mc}. \quad (10)$$

2.3 Thermal Expansion and Mass Density

Knowledge of the density of liquid metals is crucial in most theories and in particular for contraction simulation during solidification. There are several methods for measuring the density of the high-melting point liquid metals, balanced columns, pyknometer, immersed-sinker, maximum-bubble among others. However, the application of all these techniques is limited due to the reaction between liquid metal sample and the apparatus. Therefore, any EML technique is a good alternative for density measurements in molten metals. Assuming a spherical shape for the specimen to determine the volume V of the droplet of mass m and repeating the density measurements at different temperatures the coefficient of thermal expansion β can be obtained

$$\rho = \frac{m}{V}, \quad \beta = \frac{1}{V}\frac{\partial V}{\partial T}. \tag{11}$$

The determination of thermal expansion coefficient of these materials requires the measurement of the linear size of the specimen as a function of the temperature. There are two optical methods of length measurements, geometrical and interferometric. Some dilatometric techniques are described in review papers of Ruffino (1984, 1989, 1992) and Kirby (1992). *Geometrical dilatometers* use light rays, photometry, or optical imaging systems. The most precise geometrical dilatometer is the optical comparator, in which two microscopes compare the lengths of the specimen in a thermo-stated bath and standard conditions at room temperature. The method is well suited for thermal expansion coefficient measurements at low temperatures, up to 100^{o}C. At high temperatures there are problems, such as the distance between the hot specimen and the microscope lenses, precise definition of specimen temperature, and marks on the specimen. The most advanced geometrical dilatometer for measurements at high temperatures, up to 1600^{o}C, is described by Rothrock and Kirby (1967). *Interferometric dilatometers* utilize the interference phenomenon in light waves. The application of laser light sources in interferometric dilatometers increases the resolution and accuracy of the measurements. However, interferometric dilatometry requires the specimen end faces to be flat and polished which is difficult to maintain at high temperatures.

El-Mehairy and Ward (1963) developed a technique to determine the density of liquid metals from the profile of a levitated drop obtained by emitted light photography and calibration. The levitating system consists of a stabilizing ring used in conjunction with a three-turn levitated coil energized by a 10 kW *Toccotron*. The density of liquid copper was determined in the temperature range 1370^o - 2100^{o}K. Temperature dependence of the density is expressed by the following equation:

$$\rho(T) = 9.370 - 9.442 \times 10^{-4} T \pm 0.026, \tag{12}$$

where ρ is the density in g cm^{-3} and T is the temperature in ^{o}K. Molar volumes and thermal expansion coefficients were calculated from the data obtained. It is found that upon melting copper expands by 3.34%. A satisfactory agreement exists between the data and those previously obtained by the maximum-bubble and the immersed-sinker methods. Density measurements of liquid metals by the EML technique were reported by Shiraishi and Ward (1964). The method used is based on the determination of the volume of a liquid droplet of known weight ($\sim$ 1.5g) levitated in the field of a high-frequency coil. The method avoids the corrections for the temperature-dependent expansion of the apparatus. But, the deviation of the droplet geometry from the spherical shape could increase measurement errors. The coil-ring levitating system was powered by a 10 kW *Tocco* generator. The temperature was measured with an accuracy of $\pm$10^{o}C by a two-color pyrometer focused to the top of the droplet. A carbonyl nickel droplet sample of 99.9% purity was tested. During the experiments the specimen lost 10-20 mg. In this method one camera monitors the sample from the side and another from the top, so that one camera can always be used to determine the distance between the moving sample and the second camera. The known diameter of the solid sample is used to calibrate the camera image. Recorded frames are analyzed with an image-processing system and a computer to monitor the size of the spherical specimen. The density of the test sample is measured over a 300^{o}C range below the melting temperature to determine that density varies linearly with temperature over the whole temperature range studied. The following relationship summarizes the findings,

$$\rho(T) = 9.966 - 12.000 \times 10^{-4} T \pm 0.055, \quad (13)$$

where ρ represents the density in g cm^{-3}, and T the temperature in ^{o}K.

Racz and Egry (1995) describe the EML apparatus, method of image acquisition, digital image processing, and calculation methods to measure the density and thermal expansion of undercooled liquid metals as a function of temperature. Their analysis shows that the main sources of error are:

- temperature measurement
- image calibration
- image processing steps to determine droplet size
- Legendre polynomial fit.

Gorges et al. (1996) developed a method for measuring the density of levitated droplets using a CCD camera and image processing. Results for undercooled nickel samples are in good agreement with density measurements of previous researchers. An improved algorithm for edge detection in digital image processing is also described. Lohöfer et al. (2003) conducted density measurements using the experimental setup shown in Figure 1. A new optical measurement technique is combined with EML. The sample is levitated and melted in an argon atmosphere. As electromagnetic forces lift up the sample induced currents start heating it up. The sample is carefully cooled at controlled temperature by exposing it to flowing argon gas. The temperature is measured using an infrared pyrometer. When measuring the volume of the sample, it is important that the sample is fully visible from the side. Lohöfer et al. designed a coil which assured that no part of the edge of the sample was hidden by the windings. To measure the sample volume an expanded, parallel HeNe laser beam is used to illuminate the sample from behind, Figure 1. The laser light is then focused on the small aperture of a pinhole, which removed all non-parallel parts of the beam coming from interferences or the hot sample itself. It is assumed that a band-pass filter additionally blocks all light not originating from the laser. The shadow image of the sample is then captured by means of a digital CCD camera and fed into a computer to be analyzed in real time by an edge detection program. The optical setup is calibrated using well defined ball-bearing spheres. This optical setup has the advantage to prevent apparent size changes due to sample movements along the optical axes, and to guarantee a constant contrast for the edge detection algorithm independently of the brightness of the hot sample.

Wang et al. (2003) developed a containerless and contamination-free measurement method using EML techniques in order to determine accurately the thermal expansion coefficient and density of metals in solid and liquid states. A computer model for predicting the levitating force, absorption power and heating temperature is developed as well, and the predictions of the numerical simulations have been experimentally validated. Wang et al. also discuss a number of design issues with respect to a single levitation and heating coil. Several challenges related to this technique are discussed:

- Increasing the magnitude of the levitating force when the experiment is conducted in terrestrial conditions.
- Controlling the position of the levitated droplet and the temperature range by using a single induction coil to maximize the efficiency of the optical techniques.
- Detecting accurately the edge of the droplet in the imaging process.

After getting a stable and good position (the sample image is minimally blocked by the inductive coil) of the sample in the desirable temperature range, two "good" images, a side view and a top view, are then used to determine the volume of the sample. A new edge detection method has been developed, which reduces the error by about 30% as compared to a conventional edge detection method often used the maximum intensity gradient method. Optical measurement errors are corrected, and thermal expansion coefficient and density data for representative solid and liquid metals and alloys are given. Dupac

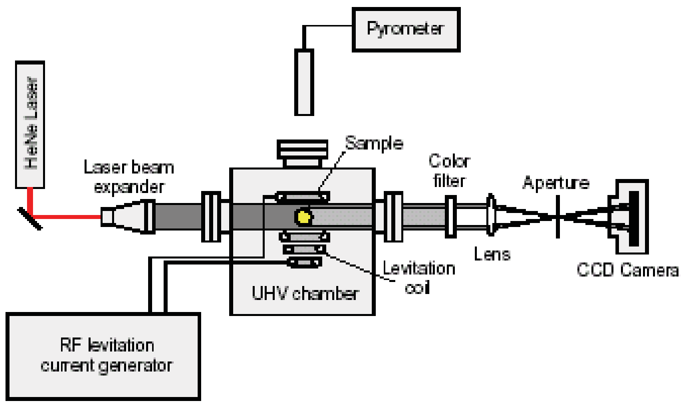

Figure 1: Experimental set-up for density measurements; the shadow of the liquid, levitated sample inside the levitation coil, illuminated from the back by an expanded HeNe laser beam, is recorded by a CCD camera, Lohöfer et al. (2003).

et al. (2003) developed a new image processing technique to predict the volume of the levitated aspherical droplet. The technique based on the cubic-spline convex-hull algorithm allows to restore the missing (blocked by the coil) portion of the frontal image, and to recover the transversal and frontal surface shapes from image shading. The normalized volume of the levitated aspherical droplet was determined by combining the recovered shapes. To validate the results, the algorithm performance has been tested on the images of the ductile iron specimens obtained experimentally.

2.4 Thermal Diffusivity

Thermal diffusivity is an important thermophysical property of materials which controls the temperature distribution in the body of the specimen during transient heat transfer processes as well as mass transfer through diffusion in heterogeneous boundary layers in many metallurgical processes like melting, solidification, crystal growth, welding and casting, among others. There are several methods to measure thermal diffusivity, such as transient hot-wire, flash heating, dynamic and stepwise heating methods. In the popular flash heating technique, first introduced by Parker et al. (1961), a small disk-shaped sample receives a high-energy irradiation burst on one surface. The increase of temperature on the opposite surface is recorded as a function of time. Comparison of the measured temperature with theoretical predictions allows the determination of the thermal diffusivity of the specimen. The technique has several advantages:

- Small amount of energy needs to be added to the specimen.
- The size of the specimen can be small
- The technique can be used by preheating or cooling the sample.
- Three thermal properties (thermal diffusivity, thermal conductivity and heat capacity) can be determined for the same sample.

However, the presence of convective flows and the container are the main drawbacks of this and other techniques aimed at measuring thermal diffusivity. To solve the problem of container contamination, a new measurement technique was proposed by Bayazitoglu et al. (1990). In this technique two experiments with different droplet sizes and with Biot numbers less than 0.1 and greater than (or equal to) 0.1 are conducted to bypass density. An inverse conduction problem is formulated for the large-droplet case, and Laplace transform methods are used to solve the problem. The method is demonstrated for three materials, Nickel, Niobium and Palladium. Bayazitoglu et al. use experimental surface temperature data instead of the inhomogeneous boundary condition, and they determine thermal diffusivity by minimizing a function that complies with the heat balance at the surface. The applicability of the proposed method is limited because of large specimen sizes required. Very large samples ($\sim$ 20 cm in diameter) must be levitated, which is practically impossible from an experimental point of view. The method proposed by Bayazitoglu et al. (1990) does not account for the two-dimensional temperature profile. To remedy this short coming Murphy and Bayazitoglu (1992) developed another two-step computational model of the transient heat conduction in the spherical specimen to account for the two-dimensional cooling profile. They consider a finite flash time which causes non-uniform temperature fluctuations in the absorption layer. Thermal diffusivity is estimated by minimizing the difference between the predicted and experimentally measured temperatures. The technique is demonstrated for Nickel, Iron and Copper using analytical data. However, parameters such as density, specific heat, incident heat flux, radiative emissivity must be known to estimate thermal diffusivity via this technique.

Shen and Khodadadi (1993) proposed an extended single-step containerless flash technique suitable to measure thermal diffusivity of levitated spherical specimens. Radiation and convection heat losses from the surface were taken into account to predict the temperature at any point analytically. Thermal diffusivity is determined from simultaneous temperature measurements at least at two different points on the surface. The method presents some advantages because it bypasses the need to know some parameters like incident energy flux intensity, absorption layer thickness and also heat loss. The diameter of the sample and the temperature rise history at two different points on the surface are the only two parameters needed to estimate thermal diffusivity. Shen et al. (1997) also proposed two extended containerless flash techniques to levitate spherical specimens. The process is modeled as an axisymmetric transient conduction heat transfer problem within the sphere. To validate the advantages of the techniques ground based experiments are conducted with high-temperature solid samples of pure Nickel and Inconel 718 superalloy near their melting temperatures. Comparison of the experimentally determined thermal diffusivity values to data available in the literature show that the proposed techniques are potentially promising.

2.5 Electrical Resistivity

Information on the electrical resistivity of molten metals and alloys is especially important in many metallurgical processes such as electroslug remelting, electromagnetic stirring in continuous casting, electrolysis, and induction melting in foundries. Due to the disordered arrangement of ions in the liquid state, molten metals and alloys exhibit higher ($\sim$ 1.5-2.3 times) electrical resistivity than those in solid state. However, relatively few studies have been reported on the electrical resistivity of molten metals and alloys, particularly at elevated temperatures, since measurements are extremely difficult. Methods to measure electrical resistivity can be categorized into three groups:

- direct resistance measurements using contact probes,
- contactless inductive measurements,
- non-contact containerless measurement techniques.

The technique of choice for solid materials is the direct resistance four-probe method based on Ohm's law. Although this direct method can be applied to low melting point, non-reactive liquid materials, reactions between the probes and the molten sample preclude using the four-probe method with high-melting point materials. Voltage drop along a sample in a capillary tube of known cross-section and length is measured at constant current density. The probe cell has to be calibrated using a liquid metal (usually mercury) of known resistivity. Selection of proper materials for the capillary cell and the electrodes remains the primary difficulty with this method. The technique is also limited to materials with very narrow freezing ranges.

Inductive techniques for measuring electrical resistivity are contactless and thus prevent chemical reactions between molten samples and contacting probes as in the direct method. There are two different types of contactless methods. The rotating field method is based on the phenomena that when a metal sample rotates in a magnetic field or when the magnetic field rotates around a stationary sample circulating eddy currents are induced in the sample generating an opposing torque proportional to the electrical conductivity of the sample. In liquid metals and alloys applied magnetic field also causes significant rotation of the liquid in the crucible decreasing the angular velocity between the field and the sample. Recently, Bakhtiyarov and Overfelt (1999a,b) developed a rotational contactless inductive measurement technique to measure both the viscosity and the electrical conductivity of liquid metals. Preliminary tests conducted with low melting point metals (lead and tin) and alloys (LMA-158 and Pb/Sn binary systems) agree well with data from the literature. But, it must be pointed out that this method leads to difficulties caused by convection.

In both direct resistance and contactless inductive techniques chemically reactive components contained in the crucible may easily contaminate the molten sample altering its electrical properties. The container may provide heterogeneous nucleants that generate early solid-phase nucleation. Electromagnetic levitation is very useful technique for the containerless measurement of electrical conductivity of liquid metals. Electrically conducting samples represent an additional, inductively coupled electric circuit that changes the impedance $Z(\sigma_e)$ of the coil, Lohöfer et al. (1991). It increases the resistance and decreases the inductance depending on the electrical conductivity of the sample. Since the molten sample in this method is isolated from container walls the sample will maintain its intrinsic properties allowing the study of the structures and properties of the material in deeply undercooled states. Impedance affects the alternating current in the coil causing changes in the power loss of the coil current at fixed voltage across the coil, in the phase difference between coil current and voltage and the resonance frequency of the alternating coil current, Herlach et al. (1993). Nyberg and Burgess (1962) developed electrodless methods for determining the electrical conductivity, the Hall mobility, and the magnetoresistance effect of semiconductors. Conductivity measurements were made by placing the sample into the core of a sinusoidally excited solenoid and by relating the impedance changes reflected into the solenoid to the conductivity of the sample. Tomlinson and Lichter (1969) and Lee and Lichter (1972) applied the inductive coupling method to measure the change of the coil resistance after having inserted a cylindrical sample into a solenoid. This electrodless technique is used to measure the electrical resistivity of liquid Cd-Bi, Cd-Sn, Cd-Pb, In-Bi, and Sn-Bi alloys. Positive temperature coefficients of electrical resistivity were obtained in all samples except $Cd_{10}Bi_{90}$ alloy. Zhuravlev et al. (1982) measured the inductance change using an Owen bridge circuit. The electrical conductivity of liquid Fe, Co, and Ni is measured by this method. The data agrees well with those obtained by the rotational magnetic field method. The uncertainty of the method is $\pm$ 4%. However, the method has some shortcomings like the small quality factor of the measuring coil, significant variation of the coil impedance itself with temperature, thermal deformation of the coil, and recrystallization of the winding wire among others.

The change in both the resistance and the inductance was simultaneously measured by Delley et al. (1980). In this work the electrical resistivity of the specimen is determined from the impedance change of the coil when the specimen is inserted. The system automatically adjusts the measuring frequency for a constant loss angle of the impedance change corresponding to a constant skin depth in the sample. The sample electrical resistivity is directly proportional to the measuring frequency, and the empty coil impedance is automatically compensated for. Electrical resistivity measurements of liquid tin, copper, manganese, nickel and iron are reported. Data from other sources agrees well with the results of the study.

Garnier and Moreau (1983) investigate the stability of the interface between a liquid metal and an insulating atmosphere, in which an inductor generates a uniform alternating magnetic field. In particular the influence of the electrical conductivity of the liquid sample was studied. Linear stability analysis shows that the influence of the alternating magnetic field on the perturbed interface is neutral for wave-vectors perpendicular to the magnetic field. The stabilizing effect is largest when the angle between the wave-vector and the magnetic field is zero. It increases with increasing wave-number, is maximum for an infinitely conducting medium decreasing with the electrical conductivity. Kraftmakher (1991) proposed a method for contactless electrical conductivity measurements based on the determination of the phase angle of the effective magnetic susceptibility of cylindrical samples in an axial AC magnetic field. The output voltage of a differential transformer is compensated by a voltage with a phase shift which depends on the sample size, so that compensation becomes possible only at the magnetic-field frequency numerically equal to the electrical conductivity of the specimen. Lohöfer (1994) analytically calculated the magnetization and the impedance of an electrically conducting sphere inductively coupled with an external, sinusoidally alternating current density distribution. The general results are applied to the special case, where the external current density distribution is concentrated on thin, toroidal rings surrounding the spherical specimen. It is shown that impedance measurements can be used for the contactless determination of the electrical conductivity. When the skin depth is much less than the sample radius ($\delta \ll$R), the changes in resistance and inductance are determined as

$$\frac{\Delta R}{\omega L_0} \sim \frac{\delta}{2R}, \quad \frac{\Delta L}{L_0} \sim \left(\frac{\delta}{2R} - \frac{1}{3}\right). \tag{14}$$

L_0 represents the inductance of the solenoid without the sample. These changes are related to the phase shift between current and voltage, which can be measured experimentally. The relation between the phase shift and the skin depth allows an estimate of the electrical conductivity.

Enderby et al. (1997) describe the combined application of aerodynamic levitation and electrodless conductivity techniques to determine the high-temperature electrical conductivities of liquid metals. The hybrid method allows the determination of the electrical conductivity at temperatures as high as 2500^{o}C. Rhim and Ishikawa (1998) report a non-contact technique to measure the electrical resistivity of liquid metals based on a conducting drop that is levitated by a high-temperature electrostatic levitator in a high vacuum. The relative changes in torque are measured as a function of temperature when a rotating magnetic field is applied to the sample. The technique was tested for pure aluminum at solid and liquid states. Richardsen and Lohöfer (1999) describe a facility for non-destructive measurement of the electrical conductivity of liquid metals above and below the melting temperature. A new technique combines the containerless positioning method of electromagnetic levitation with the contactless method of inductive conductivity measurements. Earlier, this technique was applied only to low-melting point liquid metals contained in an ampoule, which gives the sample a well-defined cylindrical shape. In the modified method advocated by Richardsen and Lohöfer the sample is freely suspended within the measuring field, and it does not have a predefined (nearly spherical) shape. Lohöfer et al. (2003) conducted electrical resistivity measurements using the experimental set-up in Figure 2, which shows the arrangement of the measuring transformer between the levitation coils. The alternating current in the primary coil of the transformer generates a high frequency magnetic field that induces a voltage in the secondary coil, which depends on the electrical resistivity of the sample, its radius, and the deviation of its shape from spherical symmetry. Through the measurement of the absolute values of the current in the primary coil and the voltage in the secondary coil as well as the phase difference between both, the (complex) impedance is determined. Next electrical resistivity of the liquid droplet is calculated from the theoretical relationship between impedance and electrical resistivity, which is well known except for calibration constants that depend on the radius and

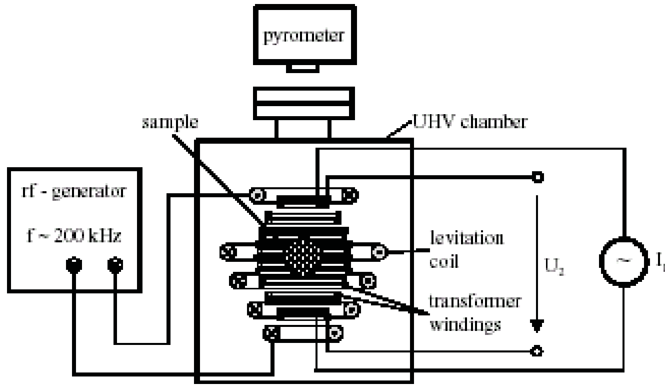

Figure 2: Electrical resistivity measurements; primary and secondary measurement coils are integrated with the surrounding levitation coil in an UHV chamber, Lohöfer et al. (2003).

the shape factor. To determine these constants, measurements were performed at different current frequencies in the range between 10 kHz and 1 MHz. Further, a containerless calibration experiment with a spherical sample of well-defined resistivity and radius was carried out. The sample was positioned by the levitation field in the center of the measurement transformer. To prevent inductive interactions between the high frequency magnetic levitation field and the measuring coils, the measurement itself was performed only in short time intervals of about 1 ms duration during which the levitation field is completely switched off.

2.6 Thermal Conductivity

Accurate measurement of thermal conductivity of liquid metals and alloys is usually more difficult than the measurement of electrical conductivity and thermal diffusivity. The difficulties concern accurate heat flow measurements. Thermal conductivity is directly related to the change in the atomic vibrational frequency. A number of non-metallic substances abide by

$$\frac{\lambda}{\sqrt{M}} = 2.4 \times 10^{-3}, \tag{15}$$

where λ and M are the thermal conductivity and M the molecular weight, respectively. Since free electrons are responsible for the electrical and thermal conductivities of conductors in both solid and liquid states many researchers use the Wiedemann-Franz-Lorenz law to relate thermal conductivity to electrical resistivity,

$$\frac{\lambda \rho_e}{T} = \frac{\pi \kappa^2}{3e^2} = L_0, \tag{16}$$

where κ and e represent the Boltzman constant and the charge of the electron, respectively. The constant

$$L_0 = \frac{\pi^2 \kappa^2}{3e^2} = 2.45 \times 10^{-8} W \Omega \mathrm{K}^{-2},$$

is the Lorenz number. The validity of this relationship was confirmed experimentally with high accuracy by many researchers.

Haller et al. (1977) developed a new technique to determine the Lorenz ratio directly for temperatures up to 500°C. The Kohlrausch method is applied to a spherical specimen with a constriction. The measurements on liquid tin are in a good agreement with the Wiedemann-Franz law. Fecht and Johnson (1991) applied the AC temperature technique to the problem of non-contact calorimetry. The proposed method allows an accurate determination of the thermal conductivity of metastable undercooled liquid metal droplets of spherical shape using an electromagnetic levitator. It is shown that the external (τ_1) and internal (τ_2) relaxation time constants are the most critical parameters in achieving high accuracy in the measurements. An accuracy of better than 1% can be achieved if $\omega\tau_1$ >10 and $\omega\tau_2$ <0.1.

2.7 Surface Tension

Surface tension is determined by the microscopic structure of the liquid near the surface. At a liquid-vapor interface density undergoes a steep change from a high value in the liquid state to a very low value in the gas phase. Therefore, surface atoms experience an attraction toward the liquid phase, which is the cause of the surface tension. Due to its energetic and entropic origin, it is necessary to calculate the free energy of the system to determine the surface tension. Thus surface tension is determined as the additional free energy required to generate a unit surface area separating the liquid from its vapor phase.

There are many techniques for surface tension measurements: sessile drop, pendant drop, maximum bubble pressure, maximum pressure in a drop, detachment or maximum pull, capillary-rise, drop weight, and oscillating drop methods. The sessile-drop technique has been widely used due to its many advantages. The method utilizes a molten drop resting on a horizontal ceramic substrate, and allows measurements over a wide range of temperatures. However, the surface tension data may be affected by contaminants. The surface tension of a liquid droplet can be measured by exciting surface oscillations. The frequency of the oscillations is related to the surface tension. The radius of the viscous sphere undergoes oscillations of the form, Egry and Szekely (1991)

$$R \sim R_0 \cos(2\pi \nu_l t)\, e^{-\Gamma_l t} P_l(\cos\vartheta), \qquad (17)$$

where l denotes the normal modes, P_l represents the Legendre polynomials, and ν_l and Γ_l are the frequency and damping of the oscillations. The oscillating drop technique which uses electromagnetic levitation prevents surface contamination and provides deep undercooling of the molten metal. The sessile-drop and the oscillating drop techniques have been used for surface tension measurements on superalloy 718 Overfelt et al. (2000). Due to chemical reactions between the specimen and the substrate the temperature dependence of the surface tension obtained by the sessile-drop method is higher than expected. A significant sample "shaking" was observed during the heat-up in sessile-drop experiments. Keene (1993) presents a literature survey of the experimentally determined values for the surface tension of molten pure metals which shows significant discrepancies between published values of surface tension and its temperature coefficient. The discrepancies are attributed to:

- experimental errors during measurements due to both the characteristics of the apparatus and the competence of the operator,
- incorrect density data used for surface tension calculations,
- effect of impurities namely surfactants,
- surface tension data at various temperatures may not be enough to obtain reliable values for the slope of the $\gamma(T)$ curve.

Surface tension is the only restoring force for surface oscillations, and consequently it determines the oscillation frequency. In the EML technique surface tension can be calculated from the frequency of the surface oscillation of the non-rotating spherical sample utilizing the following equation, Lamb (1945),

$$\nu^2 = \frac{l(l-1)(l+2)\gamma}{3\pi m} = \frac{l(l-1)(l+2)\gamma}{4\pi^2 \rho R^3}, \qquad (18)$$

where γ, ρ, m, R, ν and l represent the surface tension, the density, the sample mass, the unperturbed radius of the droplet, the frequency of the surface oscillation, and an integer denoting the normal modes, respectively. In (18) it is assumed that the amplitude of deviations from the spherical shape is small, the liquid viscosity is low, the damping effects on the natural oscillations of the liquid droplet are negligible, and the liquid is incompressible. The lowest two modes are excluded by the requirements of conservation of mass (l = 0) and fixing the center of mass of the drop at the origin of the coordinate system (l = 1). The Rayleigh frequency is the frequency of the fundamental mode, which corresponds to l = 2 in (18),

$$\gamma_R = \frac{3}{8}\pi m \nu_R^2. \qquad (19)$$

For non-spherical rotating droplets the fundamental mode is shifted and split into five peaks, whereas for non-rotating droplets only three peaks exist, Busse (1984). Experimental observations show that two pairs of oscillation frequencies are related, Natarajan and Brown (1986)

$$\nu_8 = \pm 2\nu_5 \quad \nu_{16} = \pm 2\nu_{10}. \tag{20}$$

This means that quadratic nonlinearities in equation (18) will create internal resonances between these two pairs of modes. A resonance between two modes causes secular terms to appear in (18) for higher-order corrections. Natarajan and Brown (1986) derive the interaction equations that describe the quadratic internal resonance between two oscillating modes of an inviscid droplet. Specific oscillation modes are coupled by quadratic nonlinearities caused by inertia, capillarity, and drop deformation. The equations describing the interactions of these modes were derived from the variational principle for the appropriate Lagrangian. It is shown that axisymmetric drop motions due to the internal resonance are unstable to non-axisymmetric perturbations.

The oscillations can be identified as changes in the shape of the droplet. Frequency of the oscillations can be defined from the Fourier transformation of the time signals. The position of the peaks is related to the frequency. To obtain surface tension values from measured frequency spectra, the correct labels (l,m) need to be assigned to each peak in the spectrum. As is clear from (18) the mass of the sample is required to estimate the surface tension, but that can be easily measured with high accuracy. Consequently this technique has higher accuracy than the methods which require knowledge of the density. However, (18) is valid only for free linear oscillations of inviscid, non-gravitating spherical drops.

Prosperetti (1980) formulated the problem as an initial-value problem, and determined that the motion consists of modulated damped oscillations with the damping parameters and frequency approaching only asymptotically the results of the normal-mode analysis. An estimate of the order of magnitude of the convective term in the momentum equation shows that it is negligible compared with the inertial term. Marston (1980) considered the forced oscillation case. Using spherical harmonic expansions for the radial and tangential stresses droplet deformations opposed by surface tension and driven by radiation stresses at the interface were calculated. The decay time of free oscillations is also computed, where a new term is found which is small but significant for liquid surrounded droplets. Reid (1960) introducing the viscosity of the liquid droplet obtained the relationship between the oscillation frequencies of inviscid $(\sigma_{l;0})$ and viscous $(\sigma_{l;\nu})$ liquid droplets of radius R and kinematic viscosity ν,

$$\sigma_{l;0} = \frac{\alpha^2 \nu}{R^2}, \quad \frac{\sigma_{l;\nu}}{\sigma_{l;0}} = \frac{q^2}{\alpha^2}. \tag{21}$$

And calculated for the principal mode l = 2, α^2 = 3.69, $\frac{q^2}{\alpha^2} = 0.968$. Myshkis et al. (1987) solve the problem of free oscillations of liquid droplets by introducing a self-gravity factor. Asymptotic formula for free oscillating and self-gravitating liquid globe with low and high viscosity are derived. Pozrikidis (2001) developed a numerical method for simulating the surface tension induced three-dimensional oscillations of inviscid liquid droplets. The numerical procedure is based on a generalized vortex formulation, which employs the double-layer representation for the harmonic potential. Results of simulations are presented to illustrate the performance of the numerical method for axisymmetric and non-axisymmetric oscillations.

Temperature dependence of surface tension is related to surface entropy and surface excesses. Thus, the changes in the structure of the liquid specimen with temperature are reflected in the temperature coefficients of the surface tension. The temperature coefficient of surface tension of a liquid can be thermodynamically expressed by the following equation, McLean (1957)

$$-\frac{d\gamma}{dT} = S_0^s + \sum_i \Gamma_i \frac{d\mu_i}{dT} \quad , \tag{22}$$

where S_0^s is the surface entropy, Γ_i is the surface excess concentration of the i^{th} species in the liquid, and μ_i is the corresponding chemical potential. The variation of surface tension with temperature for most liquid metals can be expressed by a linear relationship,

$$\gamma_T = \gamma_m - \frac{d\gamma}{dT}(T - T_m), \tag{23}$$

where γ_T and γ_m are the surface tension values at temperature T and at the melting point T_m, respectively.

The first technique to determine the surface tension of molten metals through the measurement of the natural oscillation frequency of levitated droplets (∼0.5g) was developed by Lu and his coworkers, Fraser et al. (1971), Murarka et al. (1971, 1975). A high-speed video camera has been used to record the surface oscillations. A systematic analysis of the effect of impurity level, drop size, viscosity, inertia, electromagnetic field, electrostatic charge, and amplitude of vibration on surface tension measurements is conducted using the electromagnetic levitation method. The surface tension of pure liquid iron and nickel was measured in a 6% H_2-He gas mixture in the temperature ranges 1550-1780°C and 1475-1650°C, respectively. A linear relationship was found between surface tension and temperature for both test samples. Surface tension of pure iron does not strongly depend on temperature in the range from its melting point to 1700°C. Supercooled iron at 1480°C has a higher surface tension than its corresponding liquid at temperatures above its melting point. A good agreement is found between the surface tension values obtained for iron and nickel at 1550°C using the electromagnetic levitation technique and the sessile drop method favored by previous researchers. However, some surface tension values were ∼10% higher than those measured by static methods, and the temperature dependence of surface tension was found to be positive. The effect of trace amounts of oxygen on the surface tension of liquid iron is studied by Murarka et al. (1975). The oscillating drop technique was used to measure the surface tension of liquid iron-oxygen alloys in the temperature range 1560°-1645°C. It is shown that surface tension does not change significantly with temperature, and the surface activity of oxygen in iron is 2.1×10^6 dynes/cm.

To evaluate both the applicability of the levitation technique and the validity of Rayleigh's equation for determining the surface tension of liquid metals, Soda et al. (1977) developed a new melting technique. The surface tension of liquid copper is evaluated in an atmosphere of hydrogen at 1000°-1330°C, including an 80°C incursion into the supercooled temperature range. The surface tension of copper is found to vary linearly with temperature as,

$$\gamma = 1.390 - 0.00043\,(T - 1356), \tag{24}$$

and the applicability of Rayleigh's equation in calculating the surface tension of the levitated metallic droplets is demonstrated. It is shown that oscillation frequency, and hence the simulated surface tension value strongly depends on oscillation amplitude, and that reliable surface tension values can be achieved at low amplitude oscillations. The surface tension of liquid iron and iron-oxygen alloys has been measured by Kasama et al. (1983) via EML. They found that the surface tension of liquid iron varies linearly with temperature over the range 1781°K to 2015°K. These results are 50 mN m^{-1} higher than those obtained by the sessile drop method. The discrepancy is attributed to sample contamination in the sessile drop technique. It is shown that the levitation technique eliminates container contamination and produces an extraordinarily clean droplet surface. Surface tension data were used to analyze the oxygen adsorption layer.

Nogi et al. (1986) measured the surface tension of liquid Fe, Co, Ni, Cu, Ag, Zn, Pb, Cd, and Sn by the sessile droplet method and/or the levitation droplet method over wide range of temperatures. The values of surface tension measured by the levitated droplet method are higher than those obtained by the sessile droplet method. The discrepancy is explained by decreased droplet contamination in the containerless levitation method. Negative temperature coefficients of surface tension were obtained for all test samples. The positive values of the temperature coefficients of surface tension for liquid Zn and Cd found in literature are attributed to impurity effects. Surface tension measurements for Zn in a purified hydrogen atmosphere and in hydrogen saturated with water vapor at 273°K lead to the conclusion that surface tension decreases linearly with increasing temperature in the former and that it increases with increasing temperature near the melting point in the latter. Schade et al. (1986) use the electromagnetic levitation technique to measure the surface tension of undercooled pure liquid metals (Fe, Ni, Cr and Co) and binary alloys (Fe-Si and Co-Si). The natural frequency of oscillations is determined using a Fourier wave analyzer, and surface tension is then calculated from Rayleigh's equation. The technique allows reaching quite large undercoolings for test samples ($\sim 0.3T_m$). Experimental observations show that surface tension of all test specimens varies linearly with temperature.

Two independent laboratories conducted measurements of the surface tension of some metals by the levitating drop technique to establish confidence levels, Keene et al. (1986). Results agree reasonably well for pure iron, cobalt, gold and copper. However, significant differences were observed in the data for stainless steel, which is attributed to the different hydrogen concentrations in the respective environmental gases. These experiments and others similar in nature by Schade et al. (1986) and Nogi et al. (1986) use photodiodes to record the incoming light intensity as an integrated signal; therefore to define the actual type of oscillations becomes an impossible task.

Equation (8) in Part I of this series of papers Bakhtiyarov and Siginer (2008) is based on an unconstrained droplet of spherical geometry, and surface restoring forces depend only on surface tension. In terrestrial conditions oscillation spectra may deviate (splitting and/or shifting) from those predicted by Rayleigh's equation due to factors such as droplet rotation, asphericity of the droplet, effect of the electromagnetic field on the surface restoring forces, stirring, etc. Therefore corrections are needed to surface tension values calculated via Rayleigh's equation. There are two approaches to make corrections:

- application of the Lagrange equations based on kinetic and potential energy considerations
- derivation of the equations of motion from consideration of applied forces

Gagnoud and Garnier (1985) analyzed the interaction between the equilibrium shape of the molten droplet and the magnetic field distribution. Two models are used and coupled to simulate the surface tension, frequency and inducting current intensity, the electromagnetic model and the free boundary model. To ensure an electromagnetic skin depth smaller than the size of the levitated droplet a high frequency is assumed. Therefore the magnetostatic problem was solved, and the influence of the internal fluid motion on the free surface shape was ignored. It is shown that the intensity of inducting current significantly affects the shape and the position of the molten sample. However, surface tension does not significantly affect the equilibrium of a levitated charge.

The inclusion of asphericity in the analysis is a very inexpensive alternative to maintaining the sphericity of the droplet by means of complex instrumentation, Suryanarayana and Bayazitoglu (1991a). The effect of the asphericity and additional restoring forces introduced by the levitating field on the vibrations of a non-rotating inviscid liquid droplet using the linear perturbation theory is studied by Cummings and Blackburn (1991). The following equation, which takes into account both magnetic and gravitational forces, is derived,

$$\begin{aligned} \nu_R &= \frac{1}{5}\sum_{m=-2}^{2}\nu_{2,m}^2 - \nu_t^2\left[1.9+1.2\left(\frac{z_0}{R}\right)^2\right], \\ z_0 &= \frac{g}{8\pi^2\overline{\nu}_t^2}, \\ \nu_t^2 &= \frac{1}{3}\sum_{m=-1}^{1}\nu_{l,m}^2. \end{aligned} \tag{25}$$

$\nu_{2,m}$ and $\nu_{1,m}$ are the frequencies of surface oscillation for the l = 2 mode and the frequencies of translational oscillation of the droplet's center of mass, respectively, and R is the radius of the droplet. The simulations show that the expected single frequency is split into three or five unequally spaced bands. Frequencies are higher than those of a spherical droplet, and the surface tension is higher than its normal value. A frequency sum rule to predict Rayleigh frequency from the observed frequencies is derived. Unfortunately, the parameters that describe the asphericity of the sample depend on gravity and the surface tension of the sample and are actually unknown.

Equation (25) has been used by Przyborowski et al. (1995) to measure the surface tension of molten silicon by an oscillating drop method using the EML technique over a wide range of temperatures (1100°-1500°C) including the undercooling condition. Corrected surface tension of the specimens is determined with an accuracy of 3-4%. Similar measurements carried out in microgravity would lead to a significant improvement in accuracy. Equation (25) derived by Cummings and Blackburn (1991) is also used by Eckler et al. (1991) to evaluate the oscillation spectra in their experimental studies. They perform the experiments in an electromagnetic levitation facility, which allows containerless measurements of surface tension of electrically conducting bulk samples in a vacuum or inert gas atmosphere. Nickel-iron alloys of various concentrations were used as the test samples. The approximate sample diameter and mass were 6 mm and 1 g, respectively. The experiments with pure nickel demonstrate the linear temperature dependence of the surface tension of pure nickel with decreasing temperature. Iron additives to nickel reduce the surface tension. The fundamental l = 2 mode was split into three peaks due to the asphericity of the droplet.

Another theory for the dynamics of aspherical droplets subjected to external forces is developed by Suryanarayana and Bayazitoglu (1991a,b) who show that for an arbitrary shape deformation the frequency spectrum splits into $2l-1$ peaks for a mode l oscillation. The splitting of the frequency spectrum for mode 2, 3 and 4 oscillations is calculated, and the frequency split is expressed in terms of external forces. It is shown that the effects of asphericity sufficiently explain the splitting of the frequency spectrum observed in the experiments. The results were applied to some previous experimental results related to surface tension measurements, and it is shown that the accuracy of surface tension measurements can be improved if corrections are made for the asphericity of the droplet. However, difficulties persist in their analysis in interpreting the electromagnetic spectra more accurately as compared to acoustic levitation. Later, Bayazitoglu et al. (1996) calculated the magnetic pressure distribution on the surface of the droplet as a function of the parameters that govern the external magnetic field. They assume that the pressure distribution on the surface of the spherical droplet can be used even when the droplet becomes aspherical because the static deformation is smaller than the droplet radius. They determine that for small droplet deformations the order of the peaks in the frequency spectrum is a function of the position of the droplet in the magnetic field, and the variation of the magnetic pressure distribution is not important in determining the surface tension.

A new method of oscillation detection based on the inspection of certain geometrical parameters by digital image processing is presented by Sauerland et al. (1992). The method has been used to measure the surface tension of pure nickel in the temperature range 1300-1620°C and in He-H_2 atmosphere to reduce oxidation. Equation (22) is used to calculate the surface tension. It is shown that the experimental data can be described by the linear relationship (23). The data are slightly higher than those in the literature, but the temperature dependence is the same. This is attributed to the elimination of systematic errors in the assignment of the modes to the frequency peaks in the spectra and by the lower oxygen content of their samples.

An improved method for measuring the surface tension of molten metals has been proposed by Egry (1991). The spectrum of surface oscillations of an electromagnetically levitated liquid droplet is evaluated by digital image processing. Preliminary results for FeNi samples are presented. In a follow-up work Egry et al. (1992) conducted terrestrial measurements in a conventional electromagnetic facility, where a generator operating at 330 kHz was connected to a conical coil providing levitation and heating. A two-color ratio pyrometer was used to measure temperature. The facility was capable of processing samples up to

1 g mass with oscillations in the 40 Hz range. A modified standard video camera with 150 Hz sampling rate has been used to obtain images of the oscillating droplet. The frequency spectrum for *m*= 0.71 g 90%Fe-10%Ni alloy sample at 1520ºC had two low-lying peaks and triply split *l*=2 mode. The individual *m*-labels could not be clearly identified, and therefore the surface tension was estimated by Rayleigh's equation (19). The computed surface tension value (1.76 ± 0.06 N m^{-1}) is in the range reported by Keene (1988). Egry et al. (1994) also measured the surface tension of noble metals (copper, silver and gold) by the EML technique. Noble metals were chosen due to their high resistance to surface oxidation. It is shown that surface tension changes linearly with temperature in the undercooled regime. Experimental data is in good agreement with the results obtained with conventional techniques leading to the conclusion of negligible contamination of the noble metal samples in ceramic crucibles and that the levitated-drop technique is capable of obtaining accurate surface-tension values for liquid metals.

Lohöfer et al. (2003) conducted surface tension measurements using the experimental setup shown in Figure 3. The levitated Cu-Ni sample melts in a quartz-glass tube filled with a mixture of helium and hydrogen. The magnetic levitation coils are located outside the glass tube. The temperature is measured by a pyrometer and controlled by the gas flow. The turbulent fluid flow of the Cu-Ni melt, excited by the strong rf magnetic levitation field, caused oscillations of the droplet surface recorded by a camera. As a containerless technique EML eliminates perturbations and distortions from outside sources and allows the observation of the unperturbed oscillations of the liquid droplet. Surface oscillation frequencies of the sample around its equilibrium shape can be computed by the 'oscillating drop method' via Fourier analysis using the recorded image.

2.8 Viscosity

Viscosity is one of the most important transport properties of molten metals. It is related to the internal friction within the liquid and provides some information about the structure of the material. Viscosity which relates the shear stress τ to the shear rate $\dot{\gamma}$

$$\tau = \eta \dot{\gamma}, \tag{26}$$

enters the definition of some of the most crucial hydrodynamic criteria such as the Reynolds number, the Rayleigh number, the Hartmann number, and the Marangoni number. The viscosity of the material in the glass forming process changes by 14 orders of magnitude in a temperature range of approximately 500 °K between melting and glass temperature, Egry and Szekely (1991). An early review of experimental methods for liquid metal viscometry was done by Beyer and Ring (1972). There are a number of theories, such as Arrhenius, Vogel-Fulcher, power law, Vogel-Fulcher-Tammann for the temperature dependence of the viscosity. However, the temperature dependence of the viscosity of undercooled liquid metals is not well understood. The relationship (27) between viscosity and surface tension which is based on the analogy with the Wiedemann-Franz-Lorenz law and the Stokes-Einstein relation is widely used. κ_B and m are the Boltzmann constant and the atomic mass, respectively.

$$\frac{\gamma}{\eta} = \frac{15}{16}\sqrt{\frac{\kappa_B T}{m}} \tag{27}$$

Since viscosity measurements are not affected by convection as diffusion measurements, the diffusivity D may be estimated from the viscosity data using Stokes-Einstein theory,

$$D = \frac{\kappa_B T}{6\pi R \eta} \quad . \tag{28}$$

Viscosity can be measured by exciting and detecting surface oscillations of a levitated specimen using EML techniques. The damping of the oscillations (Γ_l) is related to the viscosity (η) of the liquid specimen as given by Lamb (1945),

$$\Gamma_l = \frac{4\pi (l-1)(2l+1)\eta R}{3m}. \tag{29}$$

The frequency does not change with viscosity in the weak damping limit according to Reid (1960).

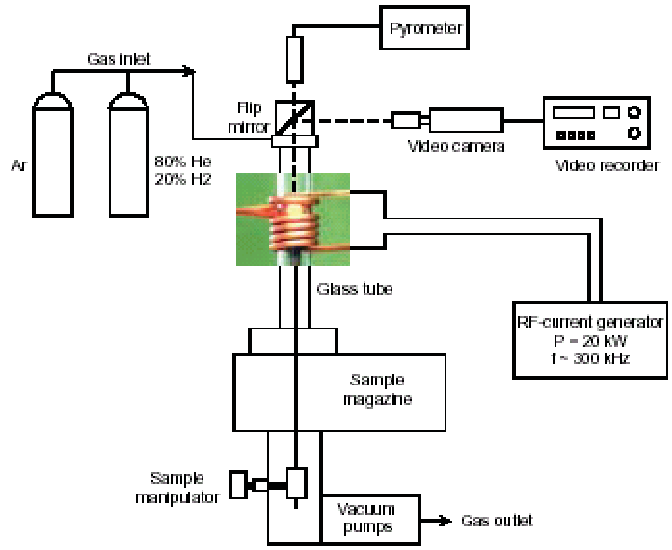

Figure 3: Experimental set-up for surface tension measurements; the sample is levitated in a quartz tube. The temperature and surface oscillations of the droplet are alternatively observed from the top over a mirror by a pyrometer or a video camera, Lohöfer et al. (2003)

Under 1 g conditions the electromagnetically induced flow in the droplet may be turbulent, and the prevailing magnetic field can generate an additional damping, which can lead to systematic errors, Herlach et al. (1993). Damping of the oscillations can be determined from the half width of the peaks in the Fourier transformation of the time signals. The damping effects are closely related to a decay factor (τ) defined as the time that must elapse for the amplitude of the vibration to decay to e^{-1} of its original value,

$$\tau = \frac{R^2\rho}{\eta\,(l-1)\,(2l+1)}. \tag{30}$$

Chandrasekhar (1961) derived the following transcendental equation for the viscous damping of a free oscillating droplet,

$$\left[\frac{l\gamma}{R^3\rho}\,(l+2)\,(l-1)+f^2\right]- \left\{\frac{2\left(l^2-1\right)}{\frac{R^2}{c^2}-\frac{2R}{cQ\left(l,\frac{R}{c}\right)}}+\frac{2l\,(l-1)}{\frac{R^2}{c^2}}\left[1-\frac{(l+1)\,Q\left(l,\frac{R}{c}\right)}{\frac{R}{2c}-Q\left(l,\frac{R}{c}\right)}\right]\right\} \cdot f^2=0, \tag{31}$$

$$Q\left(l,\frac{R}{c}\right)=\frac{J_{l+\frac{3}{2}}\left(\frac{R}{c}\right)}{J_{l+\frac{1}{2}}\left(\frac{R}{c}\right)},$$

$$c=\left(\frac{\mu}{f\rho}\right)^{\frac{1}{2}},$$

$$f=i\omega+\Gamma,$$

where J_i represent the Bessel functions of order i. Simulations were performed for low and for high Reynolds numbers. The intermediate regimes were investigated by Suryanarayana and Bayazitoglu (1991b). They developed a method to determine both surface tension and viscosity from a single experiment in which the damping rate and frequency of oscillations are measured. For high Reynolds numbers equation (25) flows out equation (31). Bratz and Egry (1995) investigated the influence of gravity and Lorentz force on the damping of surface waves on electromagnetically levitated liquid-metal droplets. The case of high Reynolds numbers and a linear magnetic field was simulated. They found a correction of the damping due to the static deformation. The correction disappears for fixed l-values if the average over different m-values is considered. However, the influence of external forces on the frequencies does not disappear on average.

2.9 Determination of Gas Content in Metals

Mechanical properties of metals, such as tensile strength, yield stress, elongation, age hardening, and drawing properties are strongly dependent on gas content. The gas content in metals is conventionally determined by the inert gas carrier fusion/thermoconductometric method which is based on melting the specimen in a graphite crucible in an inert gas medium, and detecting the released gas (hydrogen, nitrogen, carbon, oxygen, etc.) quantitatively with a thermal conductivity detector. The melted specimen reacts with the crucible causing the latter to release contaminants not easy to eliminate from the crucible. The EML technique sidesteps this difficulty as metals melt without crucibles.

A new technique for quantitative determination of hydrogen in steels using electromagnetic levitation was developed by Nishifuji et al. (1996). Melting is induced in steel specimens levitated in a nitrogen gas flow. Hydrogen is completely extracted within 1 min after melting. The released hydrogen gas is detected and measured quantitatively with a thermal conductivity detector. A cylindrical sample of 6 mm diameter, 6 mm length and 1.5 g mass was melted using an electromagnetic levitator with the levitation coil of 1.5 kW power and 200 kHz frequency. The calibration of the technique showed an excellent linearity in the range of 0.4-6.7 μg/g, with a relative standard deviation of 13% at the 1 μg/g level. A levitation melting/thermoconductometric technique also has been applied to measure the nitrogen gas level in steel, Nishifuji et al. (1998). About 5 min was required to extract nitrogen from the steel specimen. The calibration curve for nitrogen was linear up to 156 μm/g, with a relative standard deviation of 6.5% at the 20 μg/g level. The technique was applied to determine nitrogen content in a certified reference material. The result (17.5 $\pm$ 1.1 μg/g) is in good agreement with the certified value (17.0 μg/g) obtained by alkalimetric method after distillation. They conclude that the levitation melting/thermoconductometric technique could be applied to determine other gaseous elements, such as oxygen and carbon in all kinds of conductive metal.

3 Conclusions

The advantages of the EML as a technique in the study of the thermophysical properties of liquid metals are many, elimination of container effect, attainment of high temperatures, possibility to maintain undercooled state for an extended period of time and application of non-contact diagnostic techniques among others. However, the EML as a technique to measure thermophysical properties of molten metals in terrestrial conditions is plagued with substantial problems such as instabilities (global and local to the surface) of the suspended droplet, shape deformations of the specimen, requirement of high purity inert gas for convective cooling, and convection inside the sample to name some. Researchers try to avoid these difficulties by properly arranging the coils with counter-windings and by choosing the external current distribution that creates the supporting field. Using the EML technique, a certain success was achieved in measuring some of the properties of molten metals such as surface tension, viscosity, thermal diffusivity, density and thermal expansion.

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