Containerless experimental techniques to obtain thermophysical properties of liquid metals and alloys at high temperatures and density data for liquid aluminium

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Abstract
High temperature experimentation with solid and especially liquid specimens faces numerous difficulties. Traditional static steady state techniques for the measurement of thermophysical properties are generally limited to temperatures of about 2300 K. This limitation is a result of chemical interaction of the specimens with the containers, the loss of mechanical strength, problems with heat transfer, evaporation and electrical insulation while the sample and its environment are kept for times up to hours at high temperatures.
Containerless investigation methods have been developed to avoid these difficulties and to permit the extension of the measurements to higher temperatures. The methods used by our workgroup are on the one hand ohmic pulse heating and on the other hand an electromagnetic levitation technique both can be called containerless techniques.
Pulse heating delivers thermophysical properties of electrically conducting materials far into the liquid phase. The measurements allow the calculation of specific heat capacity and the temperature dependencies of electrical resistivity, enthalpy and density of the samples at the end of the solid phase and in the liquid phase. Measurements of normal spectral emissivity at 684.5 nm increase the accuracy of the pyrometric temperature measurements. Thermal conductivity and thermal diffusivity as a function of temperature are estimated from resistivity data using the Wiedemann-Franz-law.
Electromagnetic levitation, as the second experimental approach used, delivers data for surface tension (which is not available by means of pulse heating technique) and for density of liquid metals as a function of temperature.
Properties of matter at high temperatures are useful for high-temperature technologies such as aerospace, nuclear energy and the establishment of temperature reference points, including applications which are subjected to high temperature - high pressure conditions, as well as input data for modeling, which got very popular recently in steel working industry to simulate casting and welding processes and in jewelry industry to reduce reject due to defects.

Key words: Liquid metal/alloy, thermophysical properties, containerless techniques, density liquid aluminium

Introduction

For about 70 % of all industrially formed metal parts the production starts with the liquid metal/alloy. To reduce rejection parts due to casting defects computer-based simulations allow modeling of casting, heat transport, solidification shrinkage, residual stress, heat treatment, welding, forging, rolling and cutting or even predictions of microstructures. But the quality of the simulation might be restricted when wrong thermophysical parameters are used. Thus experimental obtained thermophysical properties of pure metals and also of binary and ternary alloy systems as are of great importance as input parameter for various simulation tools.
The most relevant properties for casting simulations are heat of fusion and heat capacity, and for the liquid phase electrical resistivity, density, thermal conductivity and thermal diffusivity, thermal expansion, hemispherical emittance, viscosity and surface tension.

Pulse heating

A dynamic calorimetric pulse experiment utilizes resistive self-heating of an electric conductor - typically wire shaped samples (with diameters ranging from some hundred micrometres up to a few millimetres), rectangular shaped samples (if the material cannot be drawn into wires and test samples have to be cut), foils, or tubes - by passing a large current pulse over the sample. As a result of the materials' resistivity, the test specimen can be heated from room temperature up to melting, further through the liquid phase and finally up to the boiling point in a split second. The following parts are common for all pulse heating experiments: energy storage (typically battery banks or capacitor banks) with charging unit, main switching unit (i.e. high-voltage mercury vapour ignition tubes), experimental chamber with windows for optical diagnostics and the ability to maintain a controlled ambient atmosphere. Pulse heating experiments are commonly performed under inert ambient atmosphere, e.g., nitrogen or argon at ambient pressure or in vacuum. Quite often data recording equipment is placed in a shielded room (see Figure 1).

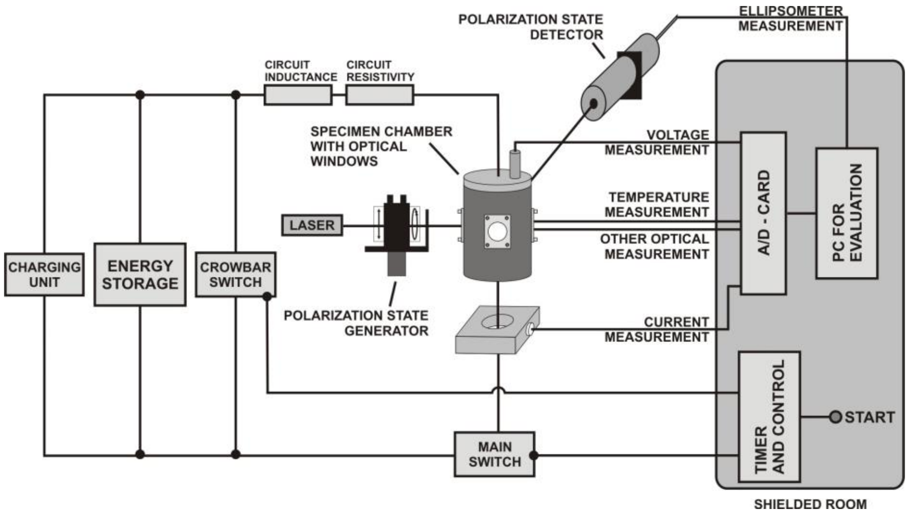

Figure 1: Schematic plot of components in a typical pulse heating experiment at TU-Graz.

The quantities typically recorded and/or accessible (as a function of experimental duration, *t*) during such an experiment in the solid and in the liquid phase are the current trough the sample, the voltage drop across the sample, the temperature using surface radiation for optical thermometry and further optical quantities such as thermal expansion of sample, sound velocity measurements, polarization information for optical properties measurements.
Based on the initially measured quantities, thermophysical properties such as the change of enthalpy, resistivity, density, thermal conductivity, and thermal diffusivity as a function of temperature can be deduced assuming that the mass, *m*, or the combination of density and volume of the sample at room temperature are known. Only viscosity and surface tension cannot be obtained by pulse heating.

Our working group has rendered outstanding services in the field of dynamic pulse calorimetry within the last twenty years [1 – 11]. A summary of thermophysical data for 22 Elements has been published recently [12].

Levitation techniques
Levitation is the process where a sample is suspended against gravity, in a stable position, by an external force without physical contact [13]. All that is required on earth for levitation is a force vertically upwards equal to the object's weight. This can be achieved by many different means, for example, electrical, magnetical [14], acoustic [15], optical, electrostatic [16] and aerodynamic forces [17, 18]. A different approach is levitation in microgravity such as parabolic flights or in space [19, 20]. A summary and description of different levitation techniques is given in [21] and reviews can be found in [22-24].
For metallic melts electromagnetic levitation provides an elegant method of noncontact containerless measurement, eliminating most interactions between the sample and its environment [25]. This technique has been used in the past mainly for the study of highly reactive melts at high-temperatures. The sample, assuming a simple spherical shape, is contained in a clean environment and can be studied over a large temperature range [25]. Temperatures again have to be measured with a pyrometer. An electromagnetic levitation apparatus employs inhomogeneous radio-frequency electromagnetic fields to heat and position the samples. Such a field has two effects on a conducting, diamagnetic body. First, it induces eddy currents within the material, which, due to ohmic losses, eventually heat up the sample by inductive heating, and second, it exerts a Lorentz force on the body, pushing it towards regions of lower field strength. The latter effect can be used to compensate the gravitational force [26].
The significant difference between dynamic pulse heating (for this technique it is easy to measure enthalpy and electrical resistivity as a function of temperature) and levitation is the fact, that the actual energy input during a levitation experiment cannot be determined. But on the other hand, levitation is not limited to a subsecond timescale and is required to measure high temperature thermophysical properties such as viscosity, surface tension and density of liquid samples, which cannot be obtained by means of pulse heating. Due to the influence of gravitational forces reliable viscosity measurements only can be performed by levitation in microgravity. These measurements are possible by digital image processing and frequency analysis of induced surface waves of a levitated liquid sample.
A schematic display of the experimental technique and some main items of instrumentation for an electromagnetic levitation system as established at TU Graz are given in Figure 2. The details of the components vary for different measurements and for different techniques. The sample is levitated in a special designed high frequency coil, which is supplied by a RF – generator (100 kHz to 1 MHz). The levitation coil is usually water-cooled. The levitated sample is placed in a vacuum chamber with quartz windows for various types of optical diagnostics such as a fast CCD - camera to monitor the sample shape and thus obtain density or surface tension results, a pyrometer for temperature measurements, and a probe laser, which sometimes also can be used for heating. The ambient cooling-gas also has to be supplied to the experimental chamber.

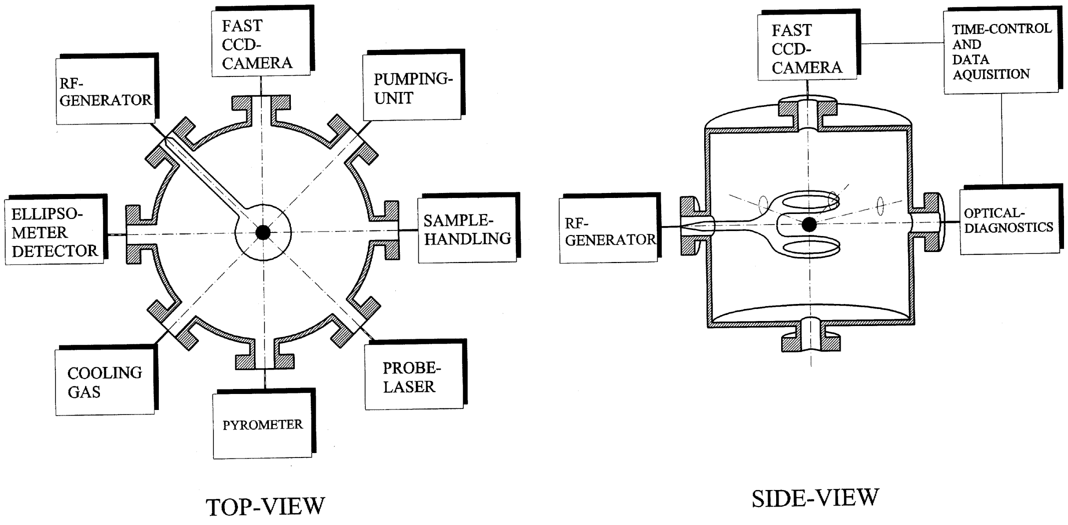

Figure 2: Schematic plot of components of an electromagnetic levitation experiment.

The underlying concept of the oscillating drop technique is that the surface oscillations of the liquid drop around its equilibrium shape can be related to surface tension as the restoring force. Translation frequencies and oscillation frequencies allow to calculate surface tension. With the electromagnetic levitation device at TU Graz surface tension and density of liquid copper and nickel [27 - 29] have been determined recently. Recent results for density versus temperature for liquid aluminium obtained by the levitation technique are presented in Figure 3. The linear fit (full line) to our data (full triangles) is $D_{liquid}(T) = 2569 - 0.255 \cdot T$ in the temperature range 1000 K < T < 1370 K.

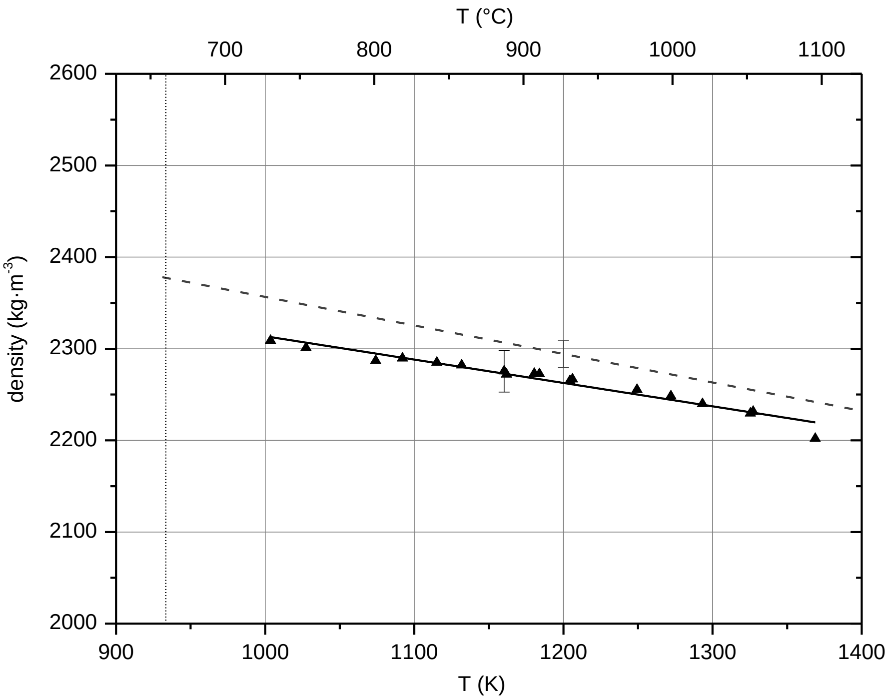

Figure 3: Density versus temperature for liquid aluminium obtained by the levitation technique. Full line with triangles this work, dashed line recommended values from Assael [30].

Uncertainty of liquid aluminium density

The uncertainty of our density results obtained with the levitation technique (k = 2) is 1 % and indicated in Figure 3 with an uncertainty bar, the uncertainty for the data given by Assael (k = 2) is stated in [30] with 0.65 %.

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