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A New High-Temperature Oscillating Cup Viscometer

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Abstract A new oscillating cup viscometer for temperatures up to 2,300°C has been constructed. A vacuum furnace with a graphite heater is used for heating the sample. The temperatures of the furnace and sample are measured by both a thermocouple and a pyrometer. The temperature is controlled with a stability better than 1 K. The oscillation of the cup is measured with a reflected laser beam using a position sensitive detector. The measured values of angle and time are then fitted to an analytical oscillation function. From the parameters of this function, the viscosity values are calculated using the Roscoe formalism. Measurements were carried out on pure metals at temperatures up to 1,700°C because of limitations of the thermocouple. The obtained viscosity values showed good agreement with literature data.

Keywords High temperature · Oscillating cup viscometer · Pure metals · Viscosity

1 Introduction

The viscosity of liquid metals and alloys is of importance for technical as well as for scientific applications. Several methods are available to measure the dynamic viscosity [1]. The method of the oscillating cup is the most common employed for metals and alloys at higher temperatures, because of a number of difficulties with other methods. A maximum temperature of 1,600°C has been reached using this method. For high-melting alloys of technical interest, for example, Ni- or-Fe based alloys, this maximum

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temperature is not sufficient. Based on experience with oscillating cup viscometers at moderate temperatures, a new high-temperature viscometer with a targeted maximum temperature of 2,300°C has been designed, built, and tested.

2 Method

The method of the oscillating cup is implemented in the new viscometer. The principle will be outlined as follows: A cylindrical cup containing a liquid is set in torsional oscillation about its vertical axis. The oscillation is then damped as a result of frictional energy absorption and dissipation within the liquid. The motion of the cup is described by the following second-order differential equation:

$$I\left(\frac{\mathrm{d}^2\varphi}{\mathrm{d}t^2}\right)+L\left(\frac{\mathrm{d}\varphi}{\mathrm{d}t}\right)+D\left(\varphi\right)=0, \tag{1}$$

where I is the moment of inertia of the oscillating system, φ is any small angle of torsional displacement, t is the absolute time, D is the force constant of the torsion wire, and L is a function of the density and viscosity of the liquid sample, the inner diameter of the cup, and the height of the liquid in the cup. L can be derived by solving the Navier–Stokes-equations for the motion of the liquid.

Several mathematical approaches are available to relate the measured parameters of the oscillation (logarithmic decrement and oscillation period) and the known properties of the sample to Eq. (1) [1]. The Roscoe analysis [2] is the most common. The viscosity η is given by the equation,

$$\eta=\left(\frac{I\delta}{\pi R^3 H Z}\right)^2\frac{1}{\pi\rho T} \tag{2}$$

with

$$Z=\left(1+\frac{R}{4H}\right)a_0-\left(\frac{3}{2}+\frac{4R}{\pi H}\right)\frac{1}{p}+\left(\frac{3}{8}+\frac{9R}{4H}\right)\frac{a_2}{2p^2}$$

$$p=\left(\frac{\pi\rho}{\eta T}\right)^{\frac{1}{2}}R$$

$$a_0=1-\frac{3}{2}\Delta-\frac{3}{8}\Delta^2$$

$$a_2=1+\frac{1}{2}\Delta+\frac{1}{8}\Delta^2$$

$$\Delta = \frac{\delta}{2\pi}$$

where δ is the logarithmic decrement, R is the inner radius of the cup, T is the oscillation period, ρ is the density of the sample, and H is the height of the sample in the cup.

The analysis of Beckwith and Newell [3,4] became more popular during the last decades. It was stated recently [5] that the numerical error of the Beckwith and Newell analysis is smaller than that of the Roscoe analysis. Since the difference of the calculated viscosities is smaller than 0.5%, this error can be neglected in comparison with the experimental error.

3 Experimental Setup

Two different designs of oscillating cup viscometers—with hanging [1,5] as well as with standing cup [6]—are known. Both setups have advantages and drawbacks that originate from the temperature dependence of the force constant of the torsional wire. The torsion wire has to be kept at a constant temperature during the measurement. The main benefit of the standing cup is good protection of the torsion wire against heat flow from the furnace, because the furnace is placed on top of the device above the important oscillating parts. From existing devices that use this setup, problems in construction and daily use, especially with initiating the oscillation, are known. The hanging cup setup is easy to construct and also simple to handle. But special care has to be taken to maintain the torsion wire at a constant low temperature. It was decided to use the hanging cup setup for the new viscometer.

The viscometer, which is shown schematically in Fig. 1, mainly consists of four parts—stand and support, vertical tube furnace, vacuum system, and oscillating system. The support is made from steel tubes and is standing on adjustable feet on a slab of concrete with a mass of 250 kg. The slab lies on a bed of sand to protect the system from external oscillations.

A resistance heater made of graphite is used to heat the sample. The inner diameter of the heater is 100 mm, and the height of the hot zone is 450 mm. Due to the special meandric design of the heater, the magnetic fields at the position of the sample are very small. Excellent uniformity of the thermal field at the position of the sample is also reached as verified by measurement and numerical simulation [7]. The vertical temperature gradient over the height of the sample is less than 2 K. The temperature of the furnace is measured by a Type B thermocouple and controlled by a PID-controller (Omron E5CK-T) with a stability better than ± 1 K. Additionally at temperatures higher than 850°C, the temperature of the sample is measured by a pyrometer (IMPAC IS300) looking at the bottom of the sample container. The maximum temperature of the furnace is 2,300°C. Graphite felt with an overall thickness of 10–15 cm is used as thermal insulation. The water-cooled outer wall of the furnace is made of steel and is part of the vacuum system.

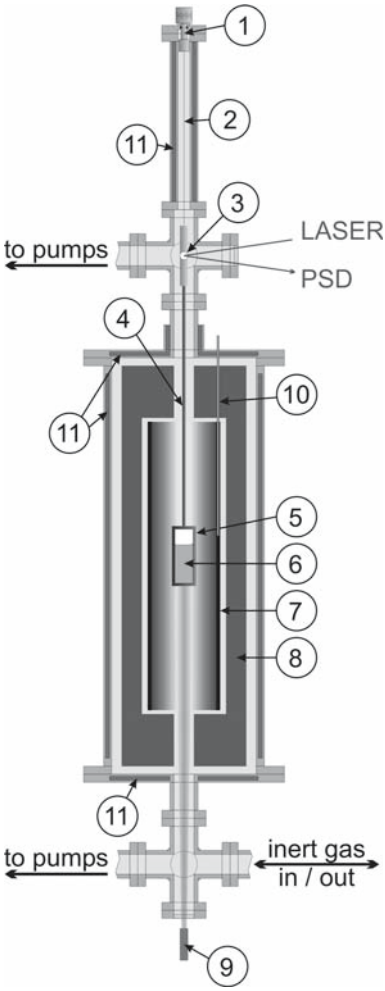

Fig. 1 Schematic diagram of the viscometer: (1) motion feedtrough, (2) torsion wire, (3) mirror, (4) tungsten rod, (5) sample container, (6) sample, (7) graphite heater, (8) thermal insulation, (9) pyrometer, (10) W/W Rh thermocouple, and (11) water cooling

The vacuum system consists of CF-flanged standard parts. Two turbomolecular pumps and one rotary vane pump are used. The pressure is measured by means of a capacitance gauge and a hot cathode gauge. Under normal conditions a pressure of 10^{-6} mbar can be reached after a pumping time of 12 h. During the measurement the vacuum chamber is filled with argon at constant pressure.

The oscillating system is the most important part of the device. Starting on top of the device, a rotation feedthrough is used for adjusting the zero position of the oscillation. A motor connected to the head of the feedtrough by an eccentric tappet is used for initiating the oscillations. At the vacuum side the axis of the feedthrough is connected to the upper fixing of the torsion wire. A steel wire with a diameter of 0.228 mm is used. For a fast and simple change of the torsion wire, a drill chuck is used. The temperature of the torsion wire is monitored by a Type K thermocouple at its lower end. The mirror holder for the oscillation detection is fixed to the lower

end of the torsion wire. An alumina tube makes the connection between the mirror holder and a tungsten rod of 4 mm diameter and 490 mm length. The tungsten rod is supporting the sample container made of graphite. The cup made from heat resistant material (alumina, graphite, boron nitride, etc.) and filled with the sample is placed inside the graphite container.

4 Data Acquisition and Processing

A red laser diode is used for monitoring the oscillations. The reflected laser beam is detected by a 60 mm long position sensitive detector (PSD). A voltage linearly proportional to the detected position is the output of the detector amplifier. The output voltage is measured by a data acquisition card (Adlink PCI-9112). From the voltage and the known distance between the mirror and the PSD, one can calculate the oscillation angle taking into account refraction effects at the planar viewport glass. The time is measured using the high-performance counter of the PC which gives an absolute time signal for each measurement of the angle. Usually the oscillation angle is measured with a frequency of 40 Hz over a period of 90 s yielding a signal as shown in Fig. 2. The equation of a damped oscillation,

$$\varphi(t) = \varphi_{\text{Offset}} + \varphi_0 \exp(-\lambda t) \sin\left(\sqrt{(\omega_0^2 - \lambda^2)}t + \alpha\right) \tag{3}$$

is then fitted to the obtained data, using the Marquardt–Levenberg algorithm [8]. In this equation φ is the oscillation angle, t is the time, φ_{Offset} is the offset from the middle of the PSD, φ_0 is the starting amplitude, λ is the damping factor, ω_0 is the oscillation frequency of the undamped oscillation, and α is the phase of the oscillation. Then the oscillation period T,

$$T = \frac{2\pi}{\omega_0} \tag{4}$$

and the logarithmic decrement δ,

$$\delta = \lambda T - \delta_0 \tag{5}$$

are calculated from the fitted parameters. The inherent decrement δ_0 has to be determined from a calibration measurement. The height of the liquid sample is calculated from the density, sample mass, and radius of the cup taking into account thermal expansion effects. Finally, the moment of inertia of the oscillation system is calculated with the expression,

$$I = \frac{DT^2}{4\pi^2}. \tag{6}$$

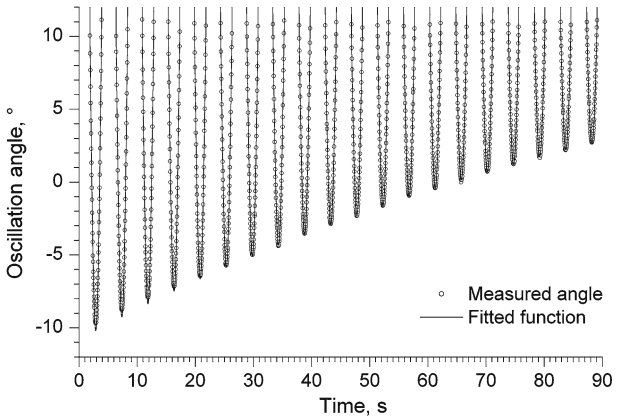

Fig. 2 Measured oscillation angle and fitted function using Eq. (8)

The force constant D of the torsion wire is determined in a series of calibration measurements. Now the viscosity can be determined in an iterative process using Eq. (2).

For controlling the functions of the device, and for measuring and performing the above described calculations, a software program was developed. It can be used for fully automated measurements.

5 Test Measurements

After setting up and calibrating the device, a number of measurements on pure metals has been performed. The experiments were carried out under an argon atmosphere at a pressure of 400 mbar. The samples were heated at a rate of $10\,\mathrm{K\cdot min^{-1}}$, held at maximum temperature for at least 90 min, and cooled at a rate of $1\,\mathrm{K\cdot min^{-1}}$. The viscosity was measured during the whole temperature cycle, but only the values measured during holding and cooling are used for further analysis. Due to the limitations of the Type B thermocouple and the cups utilized, experiments have been performed only up to 1,700°C. The results from this study and literature data for Sn, Ag, Cu, and Fe are shown in Figs. 3–6. The temperature-dependent densities of liquid metals have been taken from Ref. [9]. The Arrhenius-type law,

$$\eta = \eta_\infty \exp\left(\frac{E_\mathrm{A}}{RT}\right) \tag{7}$$

was fitted to the measured viscosities. The obtained values are shown in Table 1. For some of the experimental curves, one can see vertical accumulations of measured points. Here the temperature was maintained constant for 90 min. This has been done to check if there are any differences between the values measured at constant temperature and the values measured with continuously changing temperature. Comparing these values, one has to throw away the values measured during the first 10 min after

Table 1 Determined viscosity parameters using Eq. (7)

Element	Melting point T_m (°C)	η_0 (mPa·s)	E_A (kJ·mol^{-1})
Sn	232	0.358	6.34
Ag	962	0.438	22.1
Cu	1084	0.427	25.0
Fe	1538	0.205	45.5

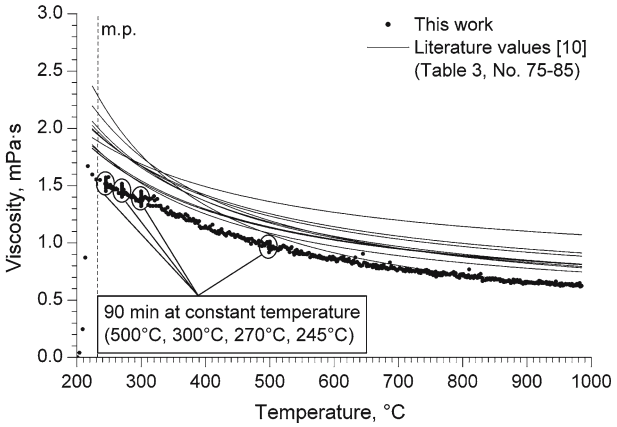

Fig. 3 Viscosity of Sn (purity 99.99%, $m = 32.6007$ g, measured in an alumina cup with $R = 8$ mm) in comparison with literature data

reaching a selected temperature because the thermal quasi-equilibrium is disturbed by overshooting of the furnace controller. The data measured during the next 80 min at a constant temperature are scattered less than ±5% around the average value.

The selection of a suitable material for the cup is a challenge. At temperatures higher than 1,500°C, most of the common materials are compatible with only a few metals without reactions. Cups made of alumina have lost their geometrical shape already beginning from 1,400°C. Alloys lead to additional problems if the different components can be measured only in different cups. Testing of cups suitable for temperature higher than 1,500°C is presently ongoing.

6 Experimental Errors

The uncertainty of the measured viscosities can be estimated using the expression,

$$\left(\frac{\Delta\eta}{\eta}\right) \approx 2.5\left(\frac{\Delta\delta}{\delta}\right) + 3.5\left(\frac{\Delta R}{R}\right) + 2\left(\frac{\Delta I}{I}\right) + \left(\frac{\Delta\rho}{\rho}\right) + \left(\frac{\Delta T}{T}\right) + 2.5\left(\frac{\Delta m}{m}\right). \tag{8}$$

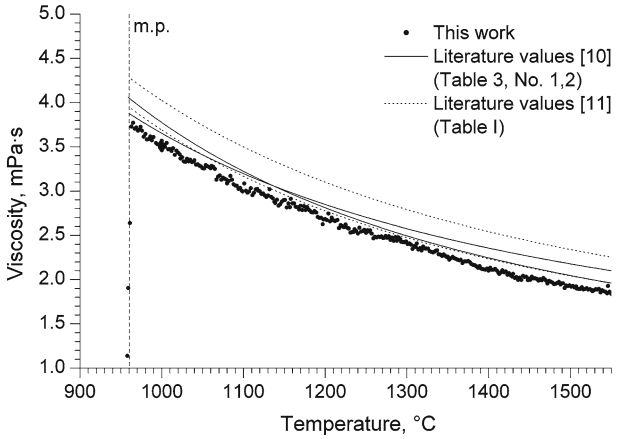

Fig. 4 Viscosity of Ag (purity 99.999%, $m = 54.2944\,\mathrm{g}$, measured in a BN cup with $R = 8\,\mathrm{mm}$) in comparison with literature data

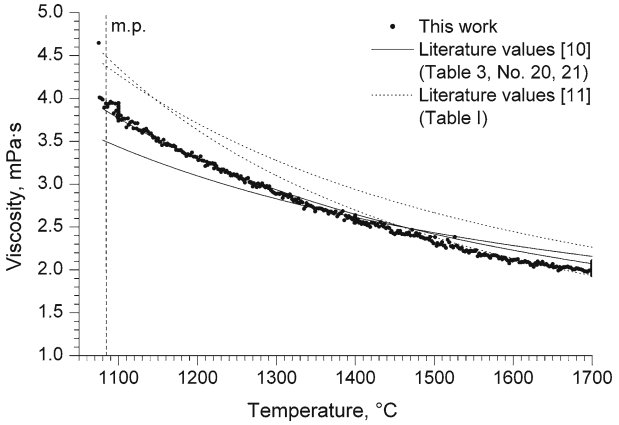

Fig. 5 Viscosity of Cu (purity 99.99%, $m = 41.1915\,\mathrm{g}$, measured in a graphite cup with $R = 8\,\mathrm{mm}$) in comparison with literature data

With the relative errors of I, T and m smaller than $5 \times 10^{-4}\%$, $\Delta\rho/\rho = 0.5\%$, $\Delta\delta/\delta = 0.4\%$, and $\Delta R/R = 1.3\%$, the estimated uncertainty of the measurement is less than 6% under optimal conditions.

The uncertainties of the viscosities shown in this article are less than 15%. The cups are identified to be the main reason for the increased uncertainty of the data presented. Most of the cups changed their inner diameter during the measurement because of reactions with the sample and/or loss of mechanical stability.

7 Conclusions and Outlook

A new viscometer has been constructed and successfully tested on pure molten metals at temperature up to 1,700°C. The main advantages of the new device are the

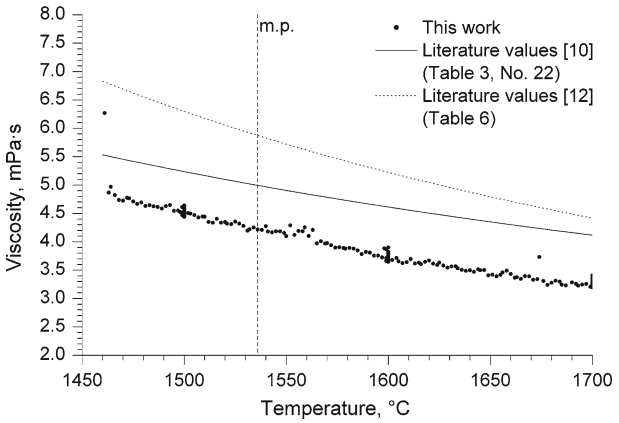

Fig. 6 Viscosity of Fe (purity 99.99%, $m = 17.1400$ g, measured in an alumina cup with $R = 8$ mm) in comparison with literature data

comparatively small volume of the sample, the good uniformity of the temperature field, and the fully automated measuring system. Efforts have to be made to find suitable cups for a wide spectrum of samples and temperature higher than 1,500°C and with a radius error smaller than 0.5%.

Experiments up to 1,900°C are now possible due to a change of the thermocouple (Type B to C). A change in the protective tube of the thermocouple from alumina to yttria will open the full measurement range up to 2,300°C in the near future.

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