
Measurement of viscosities of metals and alloys with an oscillating viscometer

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Abstract. An oscillating viscometer with the following features has been successfully constructed: improved detection of the logarithmic decay curve of oscillation through the introduction of a system with a 40-diode array; use of the Roscoe equation to derive viscosities from the decay curve; the introduction of a high-temperature two-zone furnace to provide a uniform temperature zone for temperatures up to 1650 °C.

Viscosity measurements have been carried out on mercury, tin, aluminium, copper, nickel, and iron and viscosities were in good agreement with the recommended values except in the case of iron. However, the results of the present study are in excellent agreement with viscosities of iron reported recently by Sato et al. Viscosity measurements are reported for nickel-based superalloys, for steels, and for the aluminium alloys LM25 (or A356). An analysis of potential uncertainties in the measurements has been carried out.

1 Introduction

There is increasing use of mathematical models to simulate a variety of processes involving liquid metals such as ‘cast to shape’ primary and secondary metal production, powder production by spray forming, and welding. Depending on what aspect of the process is being modelled, there is the need for viscosity data of commercial alloys but historically there are wide discrepancies in the viscosity data reported for metallic elements and alloys (Iida and Guthrie 1988). This work was prompted by the need to improve the reliability of the method for the measurement of viscosity of commercial alloys at high temperatures (up to 1600 °C).

After an initial review of the literature, the decision was taken to construct a state-of-the-art oscillating viscometer. It was considered that the mechanical designs were well established but improvements could be made in the measurement of the logarithmic decrement and the time period of the oscillations by the use of multiple diode arrays to reconstruct the decaying waveform. In recent years oscillating viscometers with maximum temperature capabilities ranging from 1200 °C to 1500 °C for measuring the viscosities of materials with higher melting points have been constructed (Overfelt et al 1996; Sato et al 1997; Nunes et al 1998) but all use two or three diodes to determine the logarithmic decrement. The objectives of this study were (i) to improve the apparatus as outlined above and (ii) to increase the temperature range to 1600 °C.

We describe the equipment, the measurement of viscosities, and the results obtained for selected metallic elements and alloys and give a brief description of source of errors.

2 Experimental

2.1 Materials

The purities of the elements used in this study were mercury (99.95%), tin (99.5%), aluminium (99.99%), copper (99.95%), nickel (99.98%), and iron (99.8%).

The compositions of alloys used in this study (mass percent) provided by the manufacturers and are listed in table 1.

Table 1. Compositions of alloys used in this study (mass percent) provided by the manufacturers.

Element	Composition/wt%			
	LM25	IN718	eutectoid steel	low carbon steel
Aluminium	balance	0.5	0.001	0.04
Carbon		0.08	0.74	0.05
Chromium		19	0.018	
Cobalt		1		
Copper	0.07	0.3		0.03
Iron	0.37	16.7	98.4	99.6
Magnesium	0.46			
Manganese	0.16	0.35	0.52	0.21
Molybdenum		3.1		
Nickel		52.5	0.02	0.02
Niobium		5.2		
Nitrogen			0.0054	0.0056
Phosphorus			0.009	0.013
Silicon	7.3	0.35	0.24	0.01
Sulphur			0.013	0.016
Titanium		0.9		

2.2 Experimental

Figure 1 shows the overall design of the oscillating viscometer (Brooks et al 1997) which is based on an earlier design (Ejima et al 1985, 1990). The major differences were in the use of (i) a crucible assembly with a low moment of inertia in order to maximise the viscosity range of the measurements, and (ii) an assembly of 40 diodes to determine the damping characteristics.

The sample is contained within an alumina crucible (105 mm long \times 14 mm inside diameter) with a molybdenum lid. The crucible is screwed onto the end of a molybdenum suspension rod. The suspension rod is supported by a 0.2 mm Pt–8%W torsion wire which is 450 mm long and which was annealed at red heat to remove kinks. A rotary solenoid is connected to the wire to initiate oscillations. The suspension wire is contained within a water jacket maintained at 30 °C. Above the suspension rod and below the wire is mounted a flat mirror (10 mm by 25 mm) and a window within the jacket wall allows a 1 mW laser to be shone directly at the mirror.

The reflected light from the mirror is detected by an array of 40 light-sensitive diodes arranged in an arc ($\sim 60^\circ$) of a circle approximately 350 mm from the mirror. The array needs to be carefully aligned to ensure that the laser falls on the central diode when the mirror is in its rest position. The output voltages from all but the central diode are combined and measured using an analog-to-digital (A/D) card and computer. This output and that from the central diode are separately logged as a function of time. A wave form is deduced from the position of the diodes and the output as the reflected light passed over them. By the use of a downhill simplex algorithm the logarithmic decrement of the decaying sine wave is obtained over a period of usually 200 s. The time period of the system is obtained by amplifying the output of the central diode and timing the pulses. The half-time periods from the left-hand and right-hand parts of the oscillations are compared in order to check that the system is properly aligned.

A two-zone furnace with MoSi₂ heating elements has an extensive uniform hot zone for temperatures up to 1650 °C. The two zones are controlled by a master and slave control system for very close temperature control. A closed-ended alumina work tube, 80 mm OD \times 70 mm ID, is used to enclose the crucible and a graphite radiation/heat sink situated above the crucible. The presence of the graphite helps to reduce the oxygen potential in the flowing argon atmosphere. The furnace was commissioned and tests were

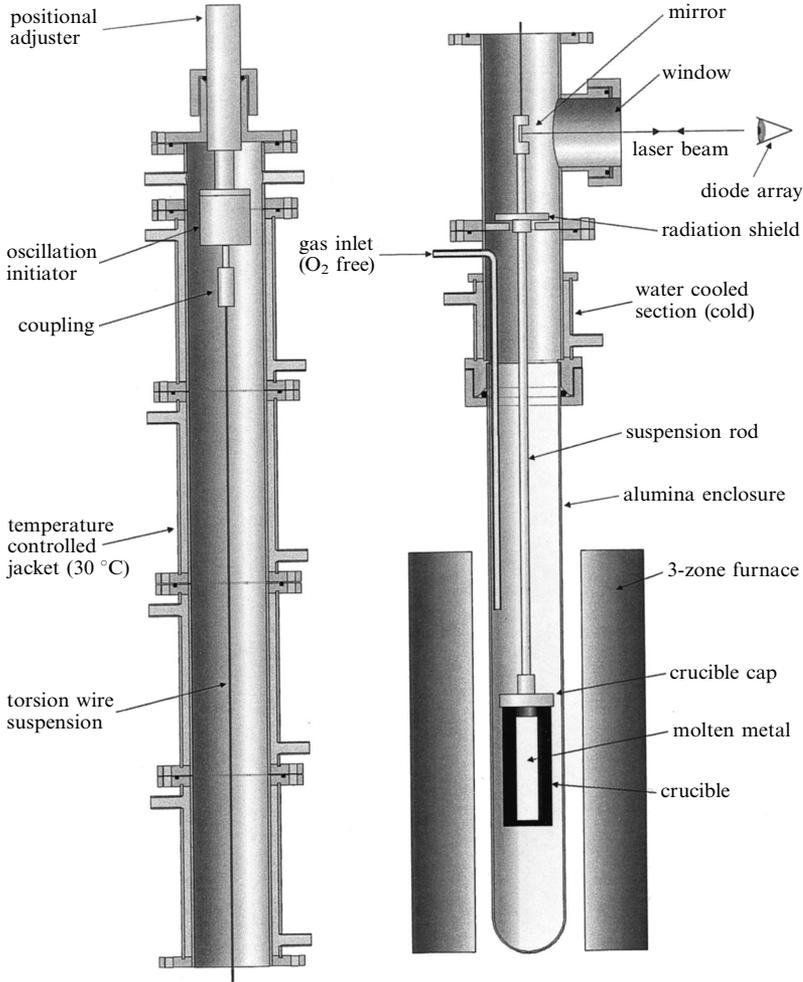


Figure 1. Schematic diagram of the oscillating viscometer.

run to identify the length of the uniform temperature zone for any specific temperature and to identify the corrections to compensate for the differences between the measurement temperature and the melt temperature.

The measurements were usually carried out in a series of steps in which the temperature was allowed to equilibrate. In a few experiments measurements were performed in the dynamic mode where viscosities were recorded continuously as the furnace control temperature was ramped at $10\text{ }^{\circ}\text{C min}^{-1}$.

2.3 Theoretical treatment of results

The damped oscillation of the crucible can be described by a decaying sine wave:

$$\theta(t) = A_0 \exp\left(-\frac{t\delta}{\tau} + \phi\right), \quad (1)$$

where $\theta(t)$ is the rotation angle of the crucible at time t , A_0 is the initial amplitude, τ is the time period, δ is the logarithmic decrement, and ϕ is the phase.

The oscillating viscometer program gathers diode data at a rate of 2 kHz. It searches for peaks in voltage and records their time of occurrence and duration. The time differences between reference peaks provide information about the time period of the oscillation.

By counting the number of diodes, and noting the peaks that occurred in each half swing, it is possible to deduce which diode produced the relevant peak and hence reconstruct a quantised oscillation curve for the crucible.

The counting of diode and reference peaks is shown in figure 2, where the combined voltage from all the diodes is shown as a function of time. The data have been partitioned for each successive swing. Interference from the mains electricity supplies can be seen as a slight ripple on the data. It can be seen from figure 2 that diode 6 is 'disappearing' as the oscillation amplitude decays. The specimen was water at ambient temperature and the measured time period was 4.684 s, the initial amplitude was 19.4 diodes, and the logarithmic decrement was 0.00784.

Once the oscillation curve has been reconstructed it is necessary to fit it to a decaying sine wave, equation (1). The unknowns are A_0 , the initial amplitude, and δ , the logarithmic decay. These can be calculated by minimising a 'goodness of fit' parameter, g_{fit} , that describes the average error in the diode. The diodes are evenly spaced apart; $\theta \approx 1.5^\circ$. The goodness of fit is calculated by summing the squares of the differences between the measured diode position and the calculated one that occurred at time t_n .

A downhill simplex method is used to adjust the values of amplitude, A_0 , and logarithmic decrement, δ , until g_{fit} is minimised. Unfortunately, the minimisation surface tends to be a long thin valley (see figure 3) and slight errors in the positions of the active elements within the diodes can randomly affect the position of the minimum value. This probably causes most of the scatter in the viscometer data but could be reduced either by moving the diodes further from the main row or by using a fibre optic array to channel light onto a single diode.

For the IN718 measurements it would appear that even with this problem, the scatter on repeat measurements is significantly less for the 40-diode system than that obtained by Overfelt et al (1996) using a 3-diode system.

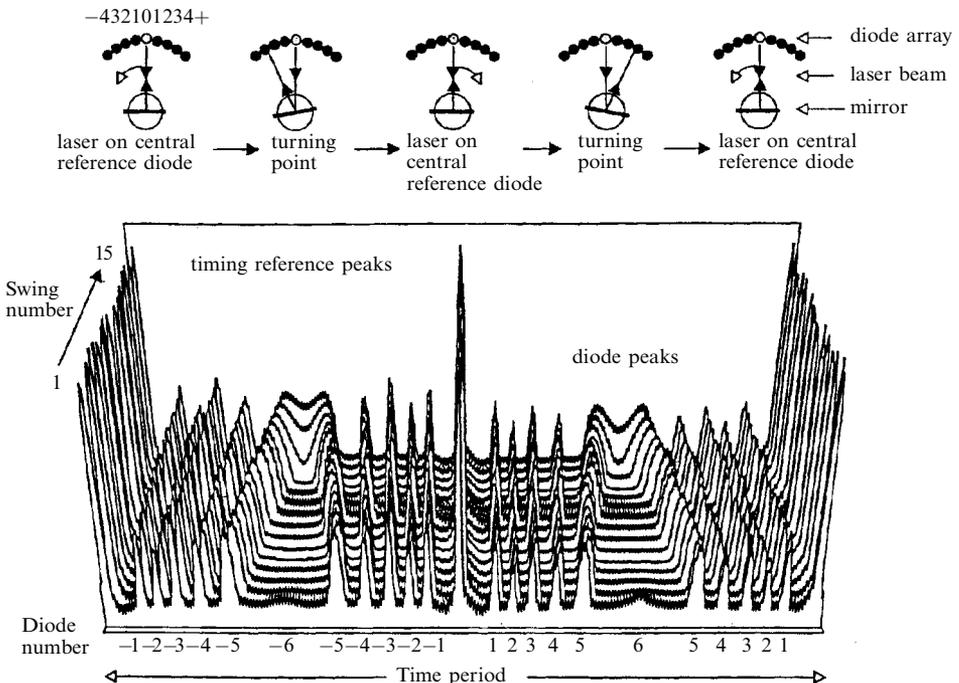


Figure 2. Diode voltage as a function of time for successive swings.

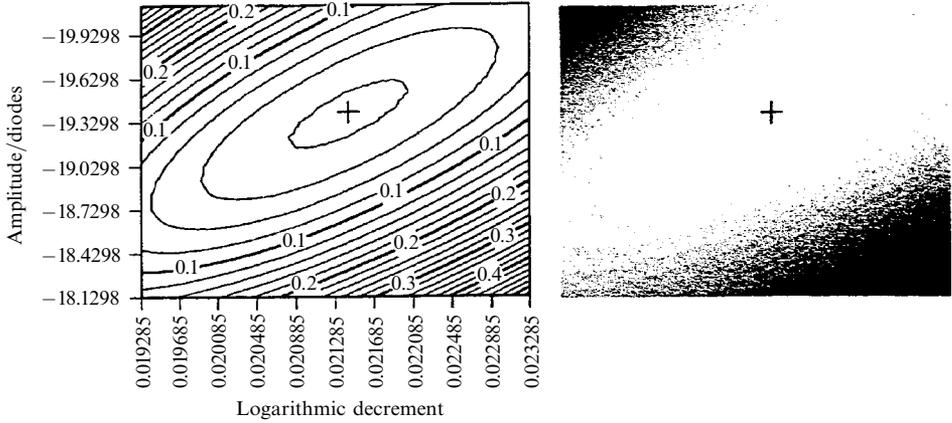


Figure 3. Example of minimisation surface. The true minimum is indicated with a cross.

Once the logarithmic decrement, δ , has been calculated it can be inserted into the Roscoe equation (Roscoe and Bainbridge 1958). The equation is considered to be more rigorous than the equations used by other investigators. The equation is as follows:

$$\eta = \left(\frac{I_e \delta}{\pi R^3 h Z} \right)^2 \frac{1}{\pi \rho \tau}, \quad (2)$$

where η is the viscosity, ρ is the density, R is the radius of the liquid sample, h is the height of the liquid sample, τ is the time period, δ is the logarithmic decrement, I_e is the moment of inertia for an empty system, and

$$Z = \left(1 + \frac{R}{4h} \right) a_0 - \left(\frac{3}{2} + \frac{4R}{\pi h} \right) \frac{1}{p} + \left(\frac{3}{8} + \frac{9R}{4h} \right) \frac{a_2}{2p^2} - \left(\frac{63}{128} - \frac{45R}{64h} \right) \frac{a_4}{4p^4} + \dots,$$

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

$$a_0 = 1 - \frac{1}{2} \Delta - \frac{3}{8} \Delta^2 - \frac{1}{16} \Delta^3 + \dots,$$

$$a_2 = 1 + \frac{1}{2} \Delta + \frac{1}{8} \Delta^2 - \frac{1}{16} \Delta^3 + \dots,$$

$$a_4 = 1 + \frac{1}{2} \Delta - \frac{3}{8} \Delta^2 - \frac{5}{16} \Delta^3 + \dots,$$

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

The height of the liquid is calculated from the crucible radius and the liquid density and does not take meniscus effects into account. However, for tall columns of liquids, where (height/radius) > 5 , this does not appear to introduce a noticeable error.

The moment of inertia of the empty crucible was determined by adding a series of disks of known moments of inertia to the empty crucible and measuring the change in the time period of the swings (Wittenberg 1971). The moment of inertia was then calculated by extrapolating back to zero time period.

The radius, inertia, and logarithmic decrement of the empty crucible are all corrected for their relevant temperature dependences. The radius was corrected by calculations based upon the expansion coefficients of the crucible material and the moment of inertia variation with temperature was determined from the variation in oscillation period as a function of temperature (Wittenberg et al 1968).

A screen snapshot from an experiment is shown in figure 4. The screen is split into three main parts: (i) at the top, the signal from the diodes is displayed as a function of time; (ii) both the times at which peaks appeared and their widths are shown as a function of time for successive swings; (iii) at the bottom, the reconstituted oscillation curve (which is split into two halves) is shown.

Figure 5 shows example oscillation curves for an aluminium alloy LM25 at various temperatures. At 610 °C the material is semi-molten. It should be noted for the solid and semi-liquid specimen the results show considerable variation and a marked apparent decrease in viscosity which are absent for the liquid alloy. Differences in time period and logarithmic decrement, δ , are due to expansion of the crucible and to changes in viscous drag from the atmosphere. When the specimen is fully molten, eg at 700 °C, a rapid decay of the oscillation curve can be seen. This is due to damping from the liquid movement.

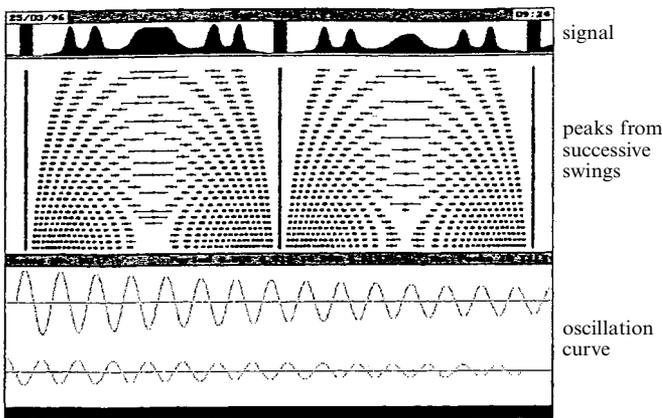


Figure 4. Screen snapshot showing data capture and oscillation curve.

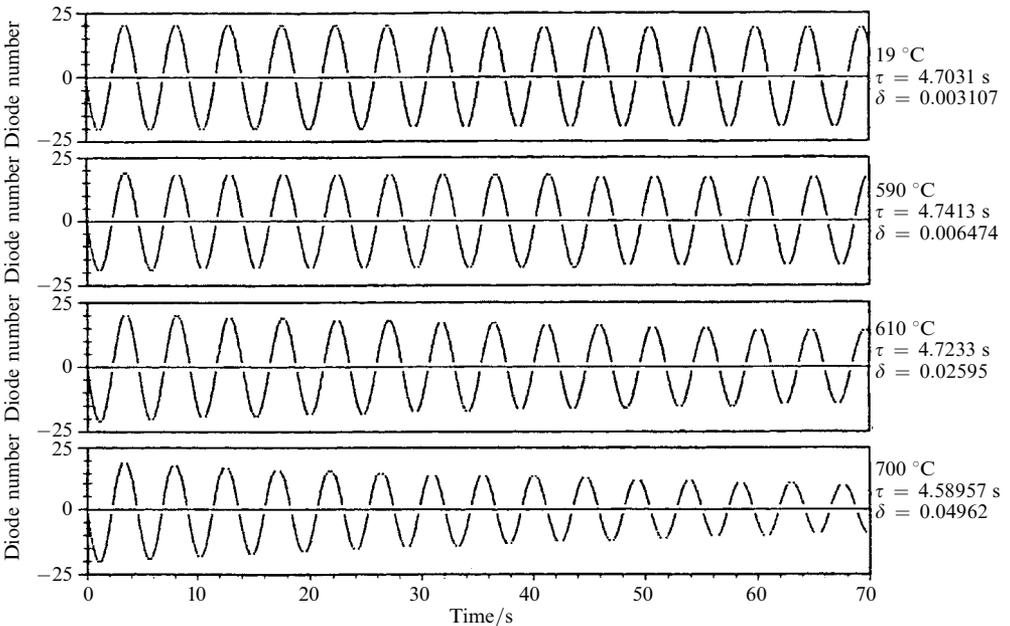


Figure 5. Example of the oscillation curves for the aluminium alloy LM25. 610 °C is the measured liquidus temperature for this sample.

3 Results

3.1 Elements

Viscosity measurements have been obtained for a variety of liquid elements, mercury, tin, aluminium, copper, nickel, and iron. The values obtained for iron and nickel are given in figures 6 and 7, respectively.

The viscosity measurements for all the elements were in good agreement with the recommended values⁽¹⁾ (Iida and Shiraishi 1988) except in the case of iron. There are a range of reported values for aluminium (Iida and Guthrie 1988) probably due to the effect of the oxide film on the surface. The present values lie at the lower limit of the

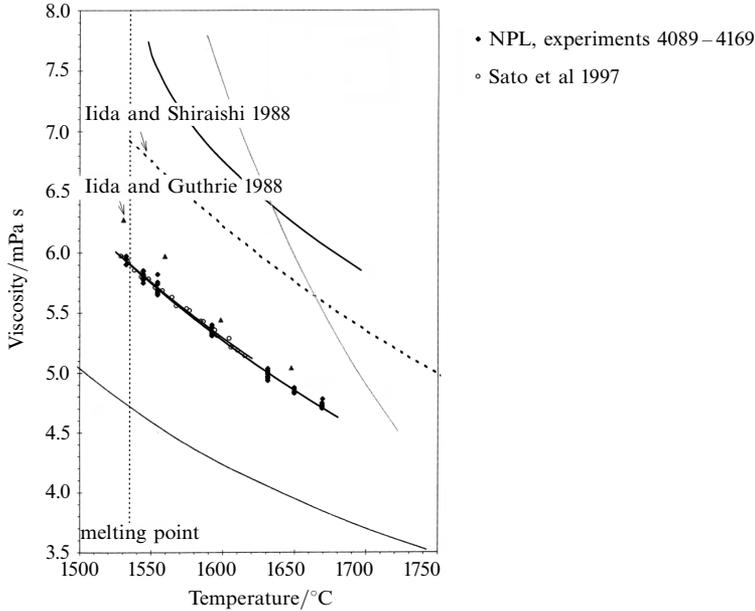


Figure 6. The viscosity versus temperature for iron reported by Saito et al (1997) and this study compared to the range of values in the literature and the recommended values (Iida and Guthrie 1988; Iida and Shiraishi 1988).

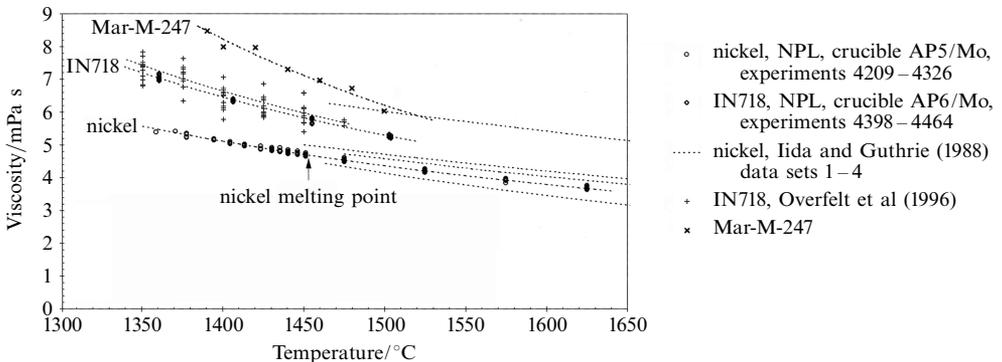


Figure 7. A comparison of the viscosity versus temperature for nickel; two sets of data for IN718, and Mar-M-247 (Overfelt et al 1996).

⁽¹⁾ Recommended values for elements with low melting temperatures (eg mercury, tin, aluminium, and copper) tend to be based on capillary measurements, but this method is prone to problems at high temperatures and values tend to be based on oscillation viscometry measurements.

reported range but close to the recommended values (Iida and Shiraishi 1988). For the copper the measured values were in excellent agreement (within 1%) with the recommended values (Iida and Shiraishi 1988).

Figure 6 shows the measured viscosity of nickel and demonstrates the substantial supercooling of the melt. This effect was also noted when specimens taken from this nickel sample were cooled during DSC experiments. Other results for nickel quoted are in reasonable agreement although again the measured values are at the lower limit of the reported range. Figure 7 shows the range of viscosity values for iron (Iida and Guthrie 1988) reported by various workers together with those recently measured by Sato et al (1997) and ourselves using oscillating viscometers. It is believed that the results of Sato et al are an average of several measurements at each temperature whereas we have plotted individual results to indicate the scatter in our measurements. There is excellent agreement between the sets of results from the two laboratories but neither set of values agrees well with the recommended values for iron (Iida and Guthrie 1988). Some of the variation of the reported results can be ascribed to the use of equations other than the Roscoe equation to derive the viscosities.

3.2 Commercial alloy

Values for nickel-based superalloys, steels, and an aluminium alloy (LM25) are given in figures 7, 8, and 9, respectively.

3.2.1 *IN718*. Figure 7 shows the individual measurements for this alloy compared to those obtained in another determination (Overfelt et al 1996). The scatter in viscosity values at each temperature is smaller in this study and is ascribed to the improved

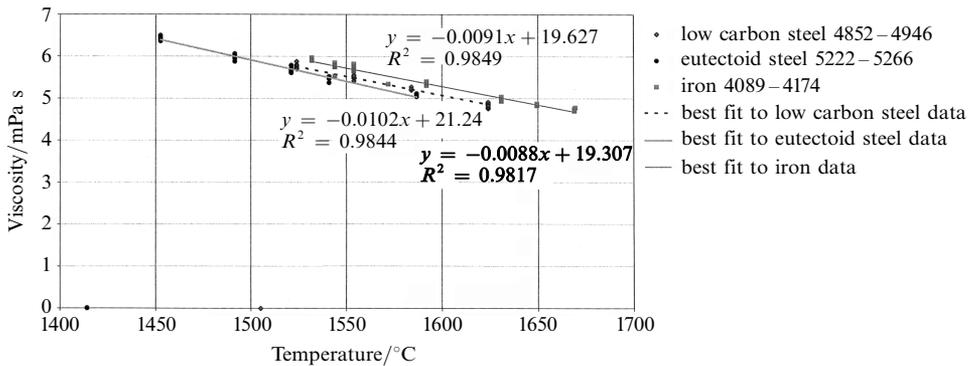


Figure 8. A comparison of the viscosities versus temperature for iron, low carbon steel, and an eutectoid steel.

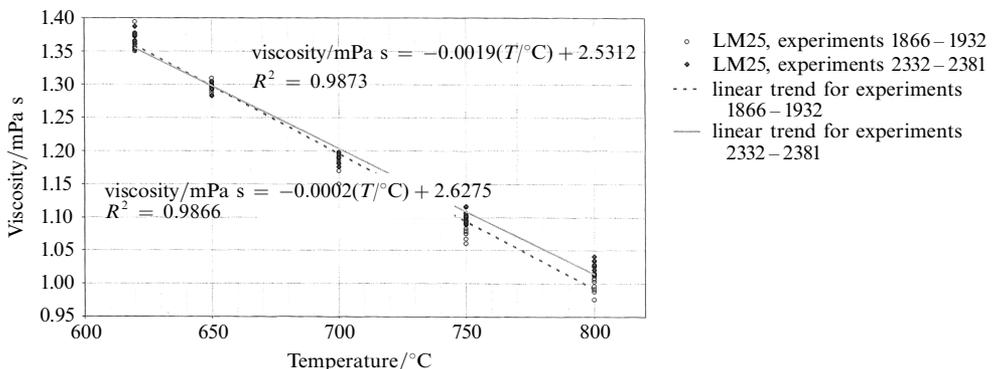


Figure 9. Viscosity values versus temperature for two experiments on the aluminium alloy LM25.

resolution of the present diode array compared with the 3-diode system. The overall values are very similar. For comparison the result by Banerjee and Overfelt (1999) for the alloy Mar-M-247 is also included showing a further increase in the viscosity compared to nickel.

3.2.2 Steels. Figure 7 shows the viscosity for the low carbon and eutectoid steels versus temperature together with the iron data. It is clear that there is little difference in the viscosity of the various types of iron. It is difficult to establish (within the experimental error) whether alloying results in any change in viscosity at a given temperature.

4 Errors

Figure 8 shows two sets of data for the viscosity as a function of temperature for the aluminium alloy, LM25, measured at different times. The largest scatter in one experiment at a given temperature was $\pm 2.5\%$ (a measure of repeatability) and the maximum total spread combining all values at one temperature was $\pm 4\%$ (a measure of the reproducibility). These larger scatter bands were observed at higher temperatures and may reflect material instabilities at these temperatures. Table 2 shows the average results at a given temperature together with the standard deviation.

Table 2. Average viscosity values and the standard deviation of the alloy LM25 for two experiments.

Temperature/ $^{\circ}\text{C}$	$\eta/\text{mPa s}$	Standard deviation
Temperature average, experiments 2332–2381		
620	1.368	0.0040
650	1.296	0.0020
700	1.186	0.0033
750	1.100	0.0026
800	1.027	0.0030
Temperature average, experiments 1866–1932		
620	1.368	0.0043
650	1.298	0.0030
700	1.181	0.0050
750	1.090	0.0056
800	1.000	0.0040

Table 3. Viscosity parameters and their errors. Typical values used for copper.

Viscosity parameter	Value	Estimated error	Error/%	Sensitivity
Radius/m	0.0075	± 0.0001	1.3	-3.08
Inertia/ kg m^2	1.5159×10^{-5}	$\pm 2 \times 10^{-7}$	1.3	2.43
Empty δ	0.004242	± 0.0001	2.4	-0.10
Density/ kg m^{-3}	8890	± 20	0.23	1.08
$\frac{dR}{dT}/\text{m K}^{-1}$	3×10^{-8}	$\pm 5 \times 10^{-9}$	17	-0.01
$\frac{dI}{dT}/\text{kg m}^2 \text{K}^{-1}$	2.5×10^{-10}	$\pm 2 \times 10^{-11}$	8	0.05
$\frac{d\delta}{dT}$	6.7×10^{-6}	$\pm 2 \times 10^{-7}$	3	-0.18
δ	0.12369	± 0.0001	0.1	2.78
Period/s	4.1052	± 0.001	0.025	-1.00
Mass/kg	0.1069689	± 0.000001	0.001	-2.45
Temperature/ $^{\circ}\text{C}$	1200	± 5	0.42	-0.26

It is difficult to estimate the uncertainties in the measurement because the viscosity is derived from the Roscoe equations numerically. Some attempt was made by investigating the effect of altering each input parameter in turn and determining the sensitivity of the viscosity value to this change. The estimated errors in each parameter are given in table 3 with representative values for copper. The sensitivity is a measure of the rate of change in viscosity for small changes in a parameter. For example a change of 5% in the radius results in a change of about -15% in viscosity.

The errors are combined by multiplying the square of the percentage error in a parameter by the absolute value of its sensitivity, summing over all the parameters, and taking the square root of the result. This gives an estimated error of 4.3% in viscosity. Including a coverage factor of two gives an estimated uncertainty of about 9%, providing a level of confidence of approximately 95%.

5 Conclusions

An oscillating viscometer has been constructed with an improved method for measuring the logarithmic decrement with a multidiode system. It is capable of measurements to 1650 °C.

The viscosities of a number of elements were measured and the results are in good agreement with the recommended values (Iida and Shiraishi 1988) except in the case of iron. Large undercoolings were noted during the experiments with nickel.

Comparison of recent literature data for iron (Sato et al 1997) and IN718 (Overfelt et al 1996) with oscillating viscometers shows good agreement with values measured in the present study. The scatter is reduced by means of the multidiode array.

The two steels appear to have viscosities similar to iron and it is difficult to establish whether the small differences are significant.

In terms of a simple error budget, the uncertainty (multiplied by a coverage factor of 2, providing a level of confidence of approximately 95%), is 9% in viscosity for typical experimental conditions used for copper.

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References

- Banerjee P, Overfelt R A, 1999 *Int. J. Thermophys.* **20** 1791–1800
- Brooks R F, Day A P, Mills K C, Quedstedt P N, 1997 *Int. J. Thermophys.* **18** 471–480
- Ejima T, Sato Y, Takeuchi E, 1985, in *Proceedings of the Sixth Japan Symposium on Thermophysical Properties* Ed. S Ohtani (Sendai: Japanese Thermophysical Society) pp 69–72
- Ejima T, Sato Y, Yamamura T, 1990, in *Proceedings of the Eleventh Japan Symposium on Thermophysical Properties* (Kyoto: Japanese Thermophysical Society) pp 291–294
- Iida T, Guthrie R J L, 1988 *The Physical Properties of Liquid Metals* (Oxford: Clarendon Press)
- Iida T, Shiraishi Y, 1988, in *Handbook of Physical Properties at High Temperatures* Eds Y Kawai, Y Shiraishi, ISIJ Special Issue 41 (Tokyo: ISIJ) chapter 4
- Nunes V M B, Santos F J V, Nieto de Castro C A, 1998 *Int. J. Thermophys.* **19** 427–435
- Overfelt R A, Matlock C A, Wells M E, 1996 *Metall. Trans. B, Process Metall.* **27** 698–701
- Roscoe R, Bainbridge W, 1958 *Proc. Phys. Soc. London* **72** 585–595
- Sato Y, Moriguchi S, Yamamura T, 1997, in *Proceedings of the Eighteenth Japan Symposium on Thermophysical Properties* Ed. M Niwa (Nara: Japanese Thermophysical Society) pp 149–152
- Wittenberg L, 1971, in *Physicochemical Measurements in Metals Research* volume IV, Ed. R A Rapp (New York: Interscience) pp 193–218
- Wittenberg L J, Ofte D, Curtiss C F, 1968 *J. Chem. Phys.* **48** 3253–3260