

VISCOSITIES OF LIQUID METALS AND COMMERCIAL ALLOYS

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ABSTRACT

This report summarises the results obtained with the oscillating viscometer in the period 1.4.96 to 31.3.99. The temperature range of the viscometer has been extended to 1650 °C by introducing, commissioning and calibrating a new furnace. Results have been obtained for pure metals (Hg, Sn, Al, Cu, Fe and Ni) and alloys (Cu/Sn, Ni-IN 718, Co-X45, and two steels). Recommended relationships are given for the temperature dependencies of the various samples and the data are compared with literature values.

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Approved on behalf of Managing Director, NPL, by Dr C Lea,
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SYMBOLS, ABBREVIATIONS, UNITS

T Temperature, °C, K

T_{liq} Liquidus temperature, °C, K

η Viscosity, mPa.s

ρ Density, kg m⁻³

1 INTRODUCTION

This report details the results obtained with the oscillation viscometer in the period 1.4.96 to 31.3.99. The objectives of the report were

- (i) to provide reliable viscosity data for commercial alloys
- (ii) to compare data obtained in this programme with
 - (a) the values reported in the general literature to show the wide range of uncertainty in reported data and
 - (b) recently-obtained results in the literature to show the much diminished uncertainty of viscosities obtained in modern oscillating viscometers.

2 IMPROVEMENTS TO THE VISCOMETER

In order to extend the temperature range of the oscillating drop viscometer a new furnace with MoSi₂ heating elements capable of reaching 1650 °C was installed. The furnace had two sets of heaters to provide a longer uniform-temperature zone. The furnace was commissioned and tests were run to (i) identify the length of the uniform temperature zone for any specific temperature and (ii) determine the magnitude of corrections to compensate for the differences between the measurement temperature and the melt temperature.

3 EXPERIMENTAL

3.1 MATERIALS

The metals used in this investigation are given below in Table 1 where they are arranged in order of increasing melting point of the elements followed by increasing liquidus temperature (T_{liq}) for the alloy.

Table 1 Details of metals and alloys used in this study

Metal (Identifier)	Supplier	Purity, Comments
Hg	-	-
Sn (ABL)	NPL Metals Store	Chempure
Al (CGY)	NPL Metals Store	Specpure
Cu (CGQ)	NPL Metals Store	99.95%
Ni (ATF)	Goodfellow Metals	99.98%
Fe (ACF)	Goodfellow Metals	Vacuum melted, 99.88%
Cu/Sn (ATN)	Prepared at NPL	C 0.011%; S 0.005%, Si 0.005
Ni - IN 718 (EMT)	Howmet	Ni 52.5%; Cr 16.7%; Fe 6.7%
Co-X45 (EMY)	Dentend Precision Castings	Co 52.6%; Cr 25.9%; Ni 10.5%
Fe-eutectoid steel (PAF)	British Steel	C 0.74%; Mn 0.52%; Si 0.22%
Steel (ATO)	British Steel	C 0.05; Mn 0.21%

3.2 EXPERIMENTAL

Figure 1 shows the overall design of the oscillating viscometer [1]. The sample is contained within an alumina crucible (105 mm long x 14 mm inside diameter) with an alumina lid. The crucible is screwed on to 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. The wire was annealed at red heat to remove kinks. A rotary solenoid is connected to the wire to set the system oscillating. 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 (~60°) 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 A/D card and computer. This output and that from the central diode are separately logged as a function of time. A waveform is deduced from the position of the diodes and the output as the reflected light passed over them. By use of a downhill simplex algorithm the logarithmic decrement of the decaying sine wave is obtained over a period of usually 200 seconds. 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- and right-hand parts of the oscillations are compared to check that the system is properly aligned.

A two zone furnace with an extensive uniform hot zone for temperatures up to 1650 °C is used to heat the specimen. 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 x 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 measurements were usually carried out in a series of steps in which the temperature was allowed to equilibrate. In a few experiments (eg Cu/Sn) measurements were performed in the dynamic mode where viscosities were recorded continuously as the sample cooled.

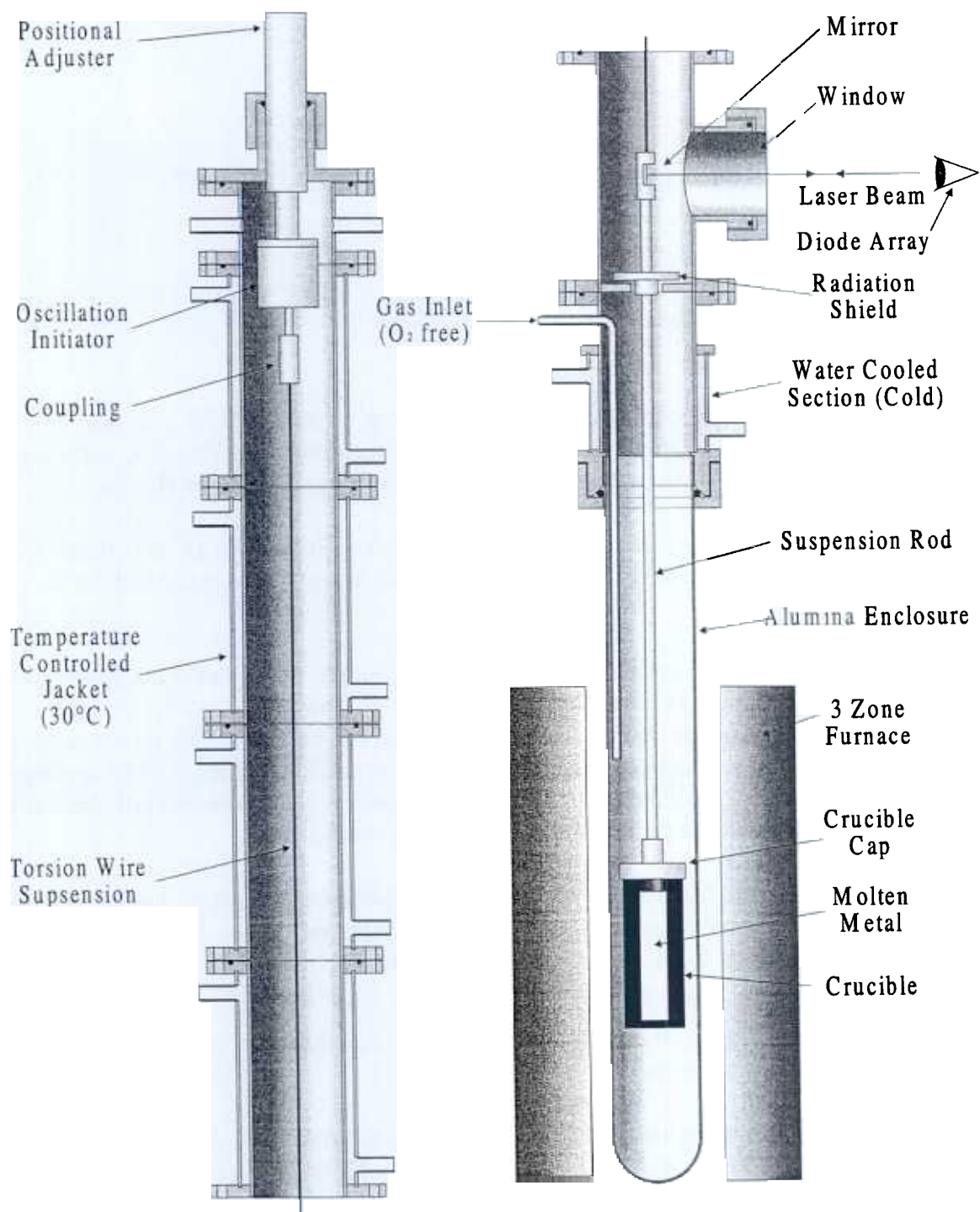


Figure 1 Schematic diagram of the oscillating viscometer.

3.3 THEORETICAL TREATMENT OF RESULTS

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

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

where

$\theta(t)$ is the rotation angle of the crucible at time t
 t is time
 A_o is the initial amplitude
 τ is the time period
 δ is the logarithmic decrement
 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, 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 can be seen as a slight ripple on the data. It can be seen 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 seconds, 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, Eqn 1. The unknowns are A_o , 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_o , 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 by either moving the diodes further from the main row or by using a fibre optic array to channel light onto a single diode.

For the IN 718 measurements it would appear that even with this problem, the scatter on repeat measurements is significantly less using the 40 diode system than that obtained by Overfelt et al [2] using a 3 diode system, (see Figure 19).

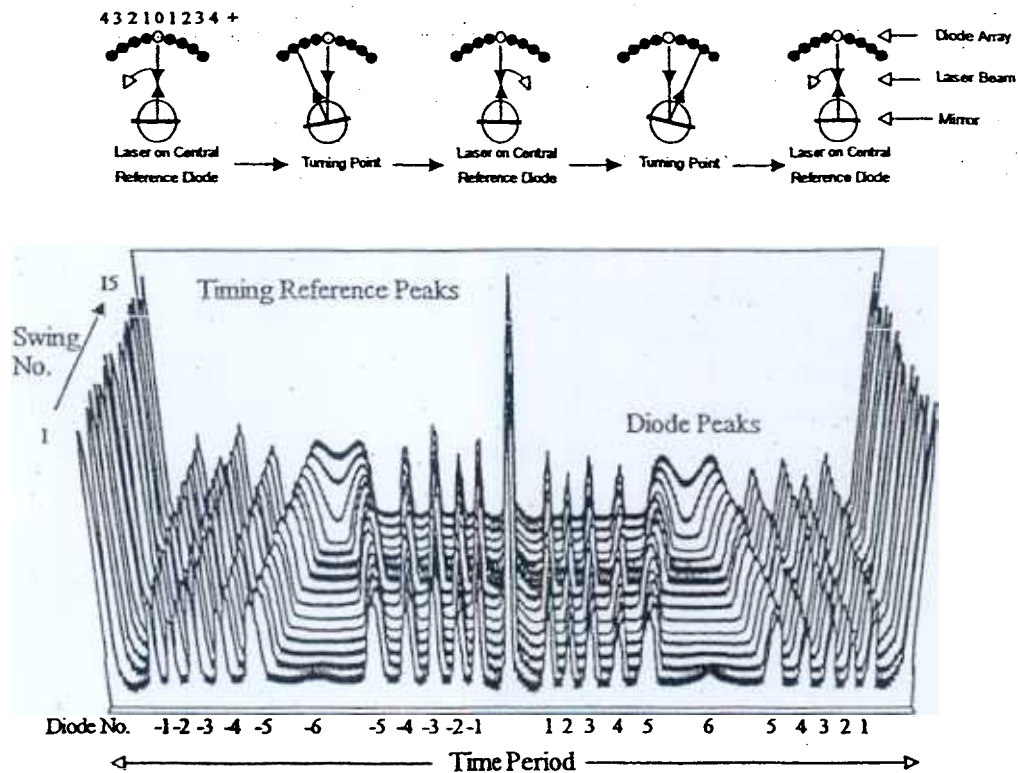


Figure 2 Diode voltage as a function of time for successive swings

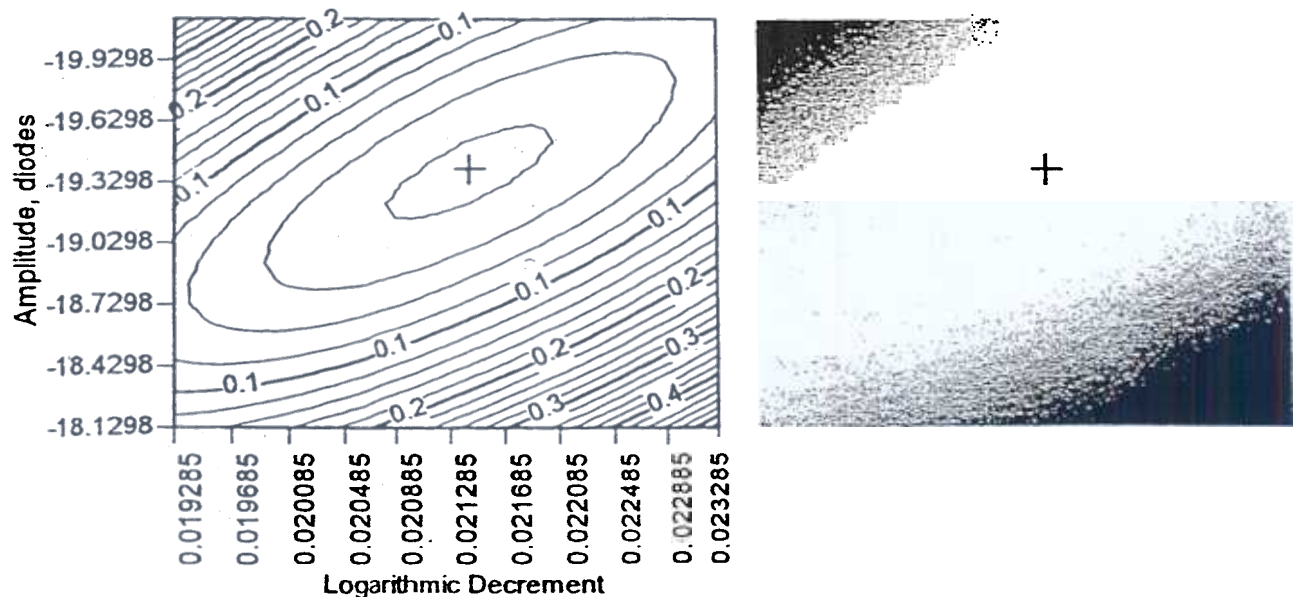


Figure 3 Example of minimisation surface. The true minimum is indicated with a cross.

Once the logarithmic decrement, δ , has been calculated it can be inserted into the Roscoe [3] equation (Eqn 2). These are considered to be the most rigorous, but different workers [3] used different approximations to aid the computations, especially before the general availability of computers and electronic calculators.

The equation is as follows:

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

where

η	=	viscosity
ρ	=	density
R	=	radius of liquid sample
h	=	height of liquid sample
τ	=	time period
δ	=	logarithmic decrement
I_e	=	moment of inertia for empty system.

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

$$p = \left(\frac{\pi \rho}{\eta \tau} \right)$$

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

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

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

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

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 radius, inertia and logarithmic decrement of the empty crucible are all corrected for their relevant temperature dependencies.

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) the times at which peaks appeared and their widths as a function of time for successive swings; (iii) at the bottom, the reconstituted oscillation curve which is split into two halves.

Figure 5 shows example oscillation curves for LM25 at various temperatures. At 610 °C the material is semi-molten. It should be noted for the solid and semi-liquid specimen, 19 °C and 590 °C, that the 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.

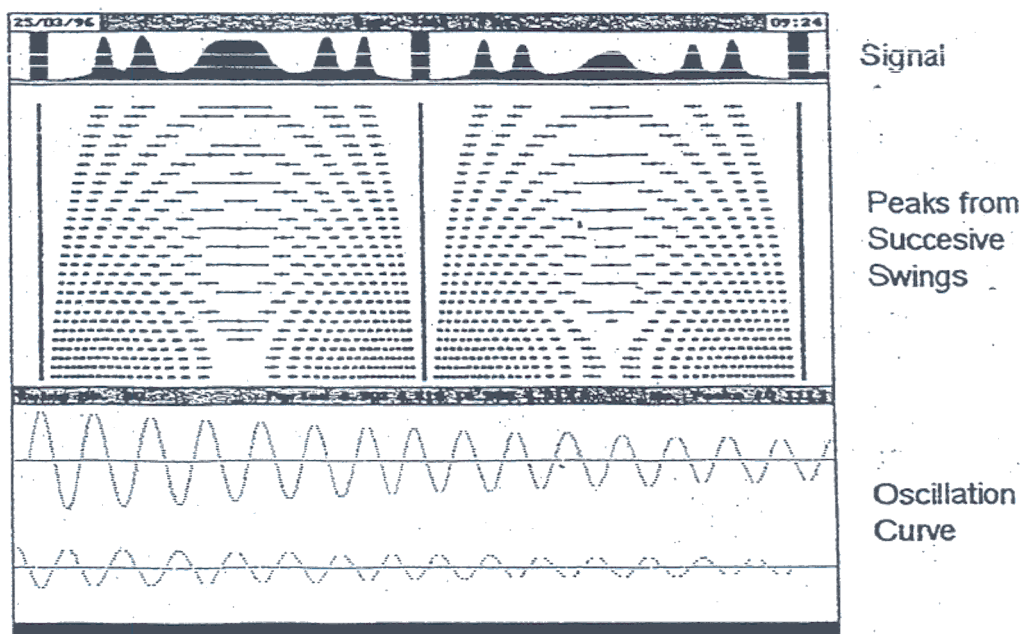


Figure 4 Screen snapshot showing data capture and oscillation curve.

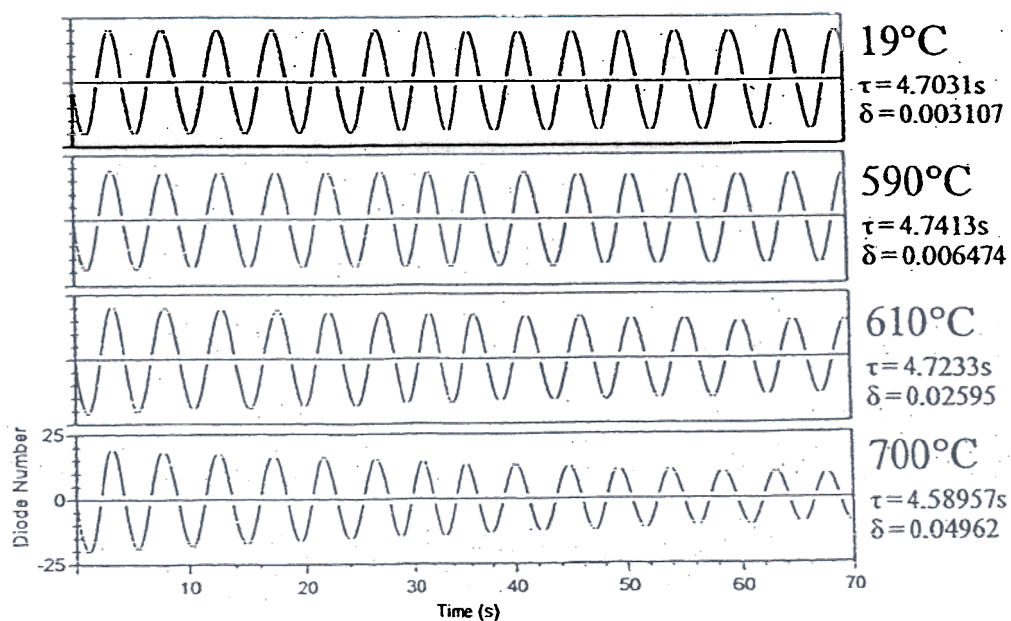


Figure 5 Example of the oscillation curves for LM25 at 610 °C the specimen is semi-molten.

4 RESULTS

The following results are arranged in the order of increasing melting points of the metal followed by increasing T_{liq} for the commercial alloys. The various runs are identified in the diagrams by numerals. The data are presented as Arrhenius plots where the viscosity (in mPas) is plotted on a logarithmic scale.

4.1 MERCURY

Crucible: (i) Graphite; Mo cap
(ii) Al_2O_3

Gas: Ar

Purity:

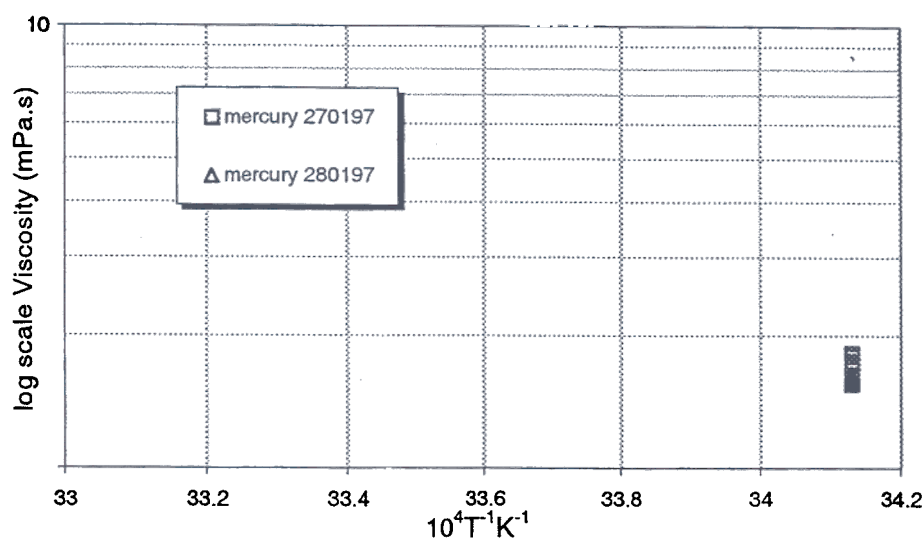


Figure 6 Logarithm of viscosity (mPa.s) of mercury as a function of reciprocal temperature (K^{-1})

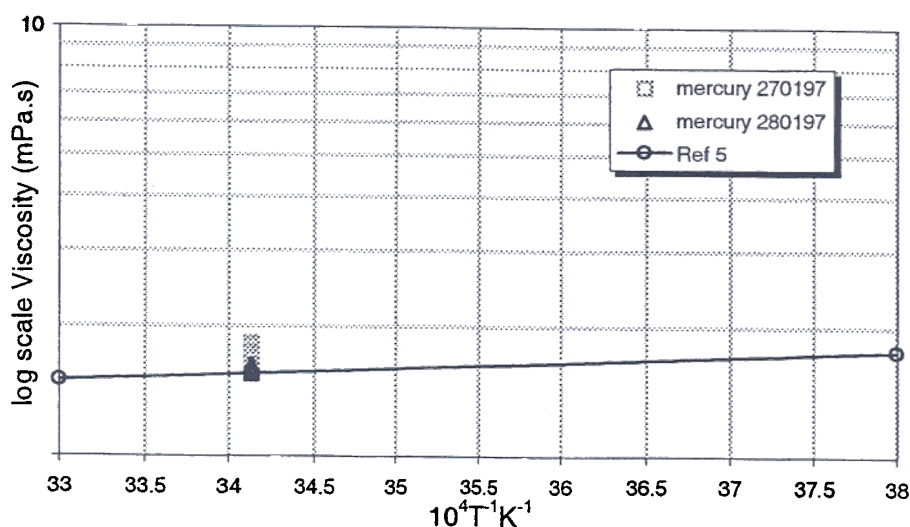


Figure 7 Logarithm of viscosity as a function of reciprocal temperature for Hg showing present results compared with literature values [5].

4.2 TIN

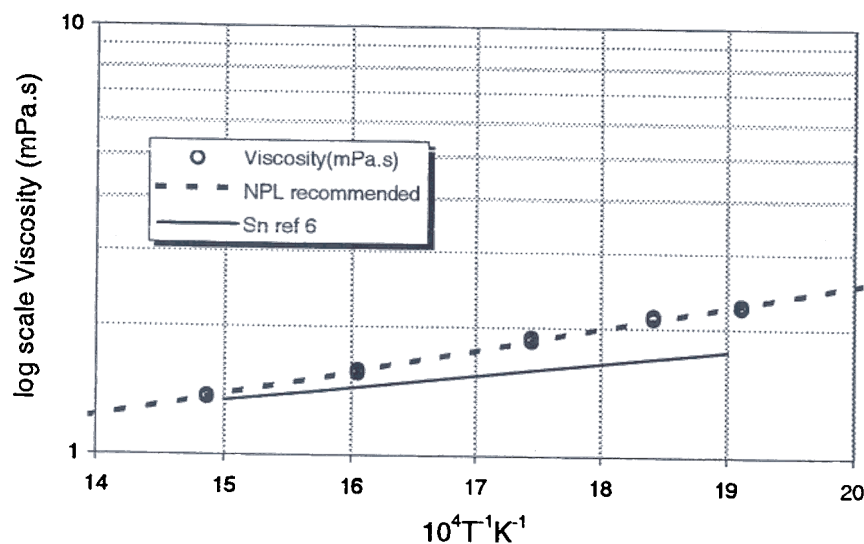
Crucible: Graphite; Mo capGas: ArPurity: Chempure

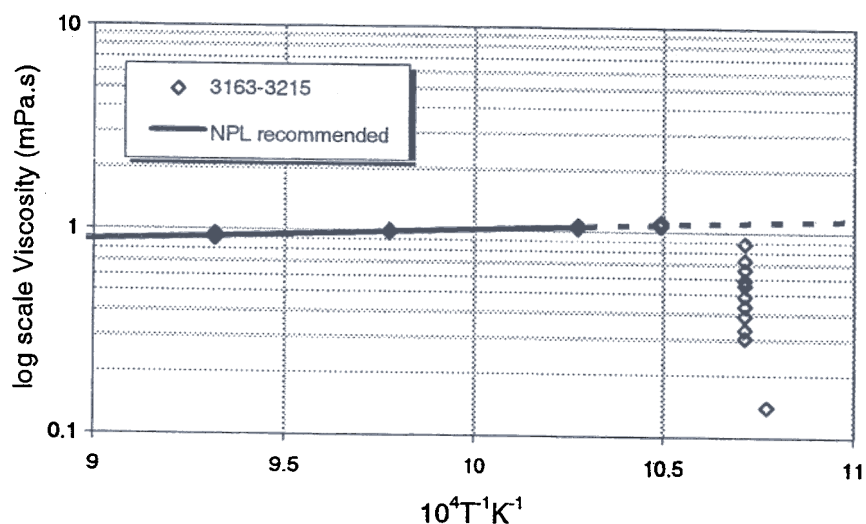
Figure 8 Logarithm of viscosity (mPas) of Sn as a function of reciprocal temperature (K^{-1}); measured by NPL and with recommended values from [6].

Recommended: $\log_{10} \eta \text{ (mPas)} = -0.647 + (526/T)$
(T is in K)

Comments:

The measured viscosities are about 10% higher than those adopted by Iida and Shiraishi [6].

4.3 ALUMINIUM

Crucible: Al₂O₃/Mo capGas: ArPurity: SpecpureFigure 9 Logarithm of viscosity (mPas) of Al as a function of reciprocal temperature (K⁻¹)

Recommended: $\log_{10}\eta$ (mPas) = $-0.562 + (567/T)$
(T is in K)

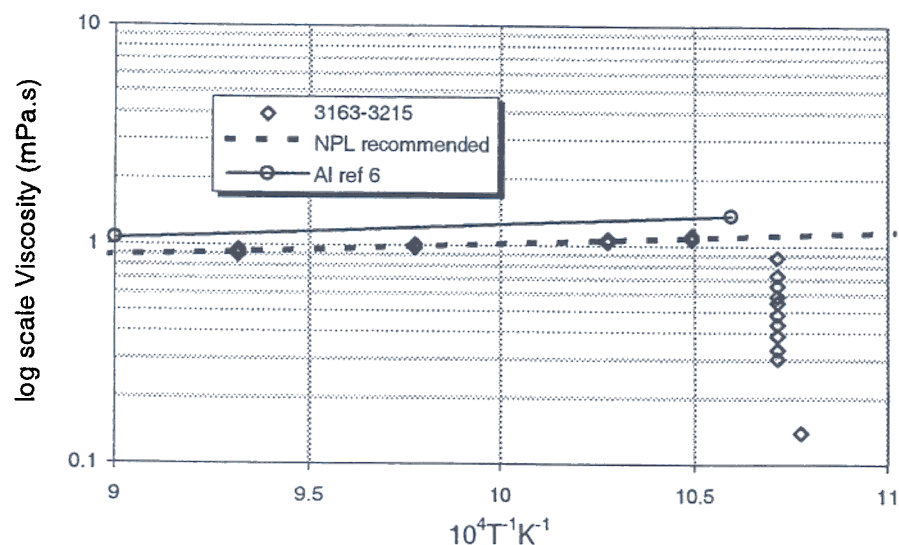


Figure 10 Logarithm of viscosity as a function of reciprocal temperature for aluminium showing present results, - - -, with limits of literature values, —○—, [6,4].

Comments:

The recommended values lie at the lower limit of the reported range. The large scatter in the literature values is probably a result of the oxide films formed on the surface of the Al. Note (i) apparent low η values, (ii) increased scatter in results at melting points (measured > 657 °C cf recommended value of 660 °C).

4.4 COPPER 'CGO'

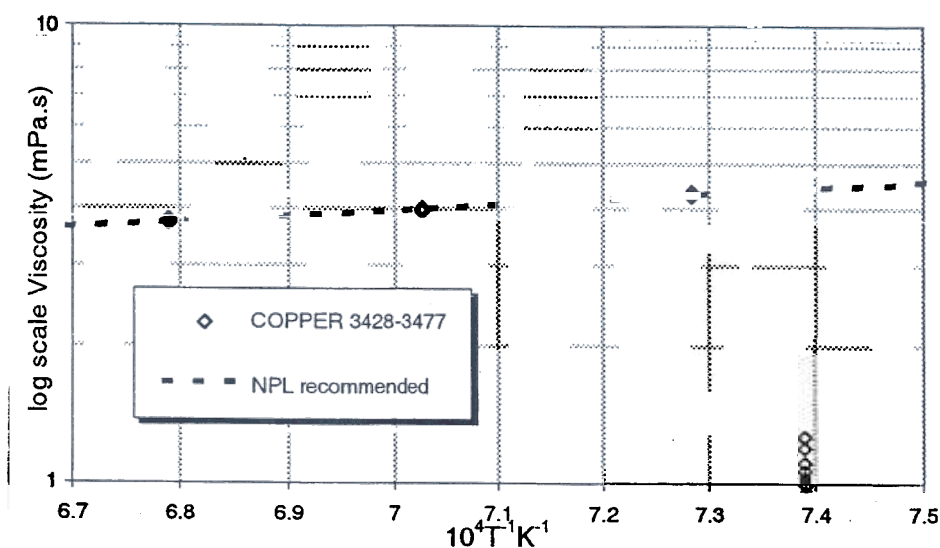
Crucible: Graphite/Mo capGas: ArPurity: 99.95%

Figure 11 Logarithm of viscosity (mPas) of copper as a function of reciprocal temperature (K^{-1})

Recommended: $\log_{10}\eta$ (mPas) $-0.279 + (1254/T)$
(T in K)

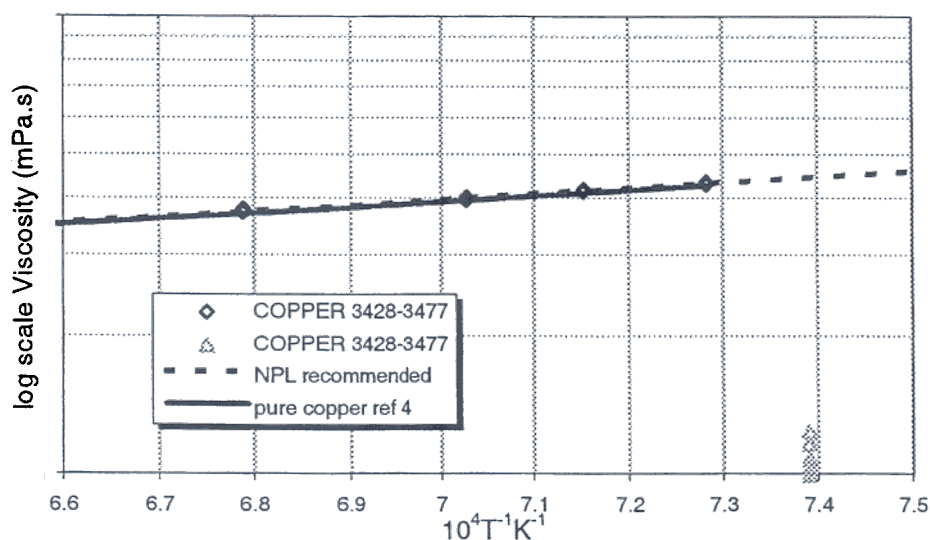


Figure 12 Logarithm of viscosity as a function of reciprocal temperature for copper showing comparison of present investigation with literature data [6,4].

Comments:

There is excellent agreement between the measured values and recommended literature values. Note (i) apparently low η values and (ii) increased scatter in results at melting point.

4.5 NICKEL (ATF)

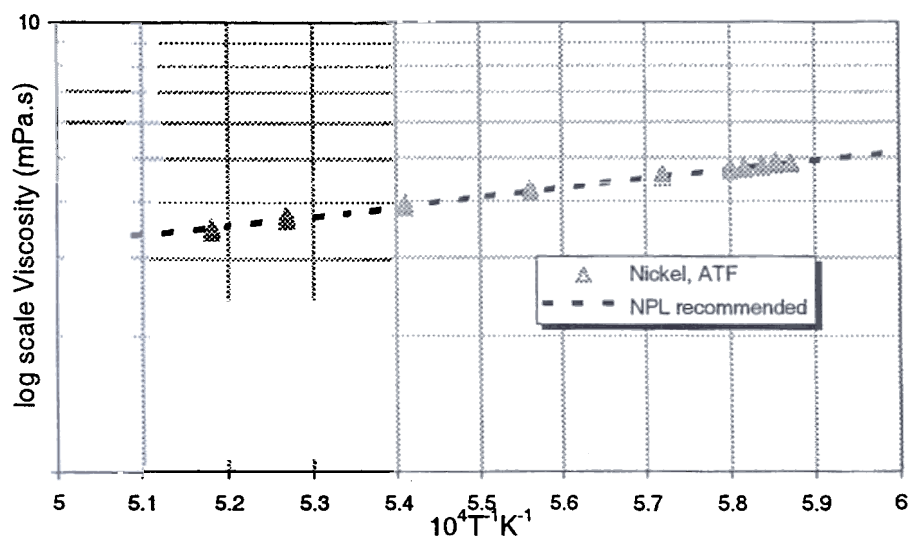
Crucible: Al₂O₃, Mo capGas: ArPurity: 99.98%

Figure 13 Logarithm of viscosity (mPas) of Ni as a function of reciprocal temperature (K⁻¹).

Recommended: $\log_{10} \eta \text{ (mPas)} = -0.509 + (2038/T)$
(T is in K)

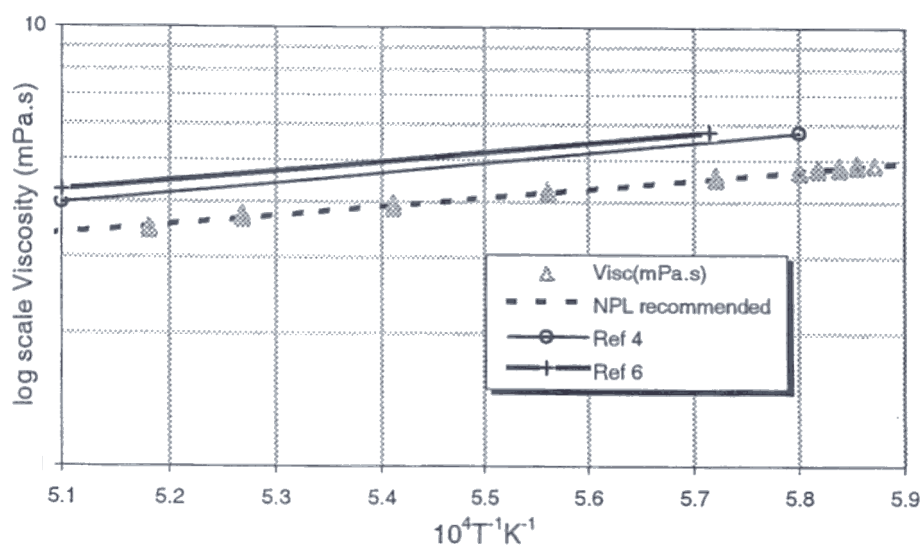


Figure 14 Logarithm of viscosity as a function of reciprocal temperature for nickel showing the comparison of present results with literature values [6,4].

Comments:

The values reported by Iida and Shiraishi [6] are ca 10% higher than those measured in this study.

4.6 IRON (ACF)

Crucible: Al_2O_3 , Mo cap Gas: Ar Purity: 99.8

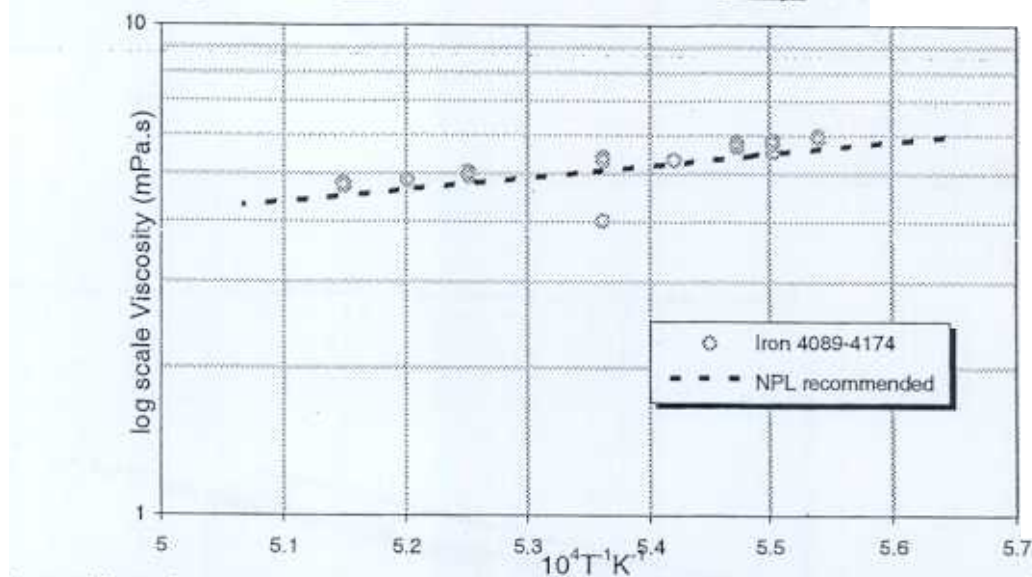


Figure 15 Logarithm of viscosity (mPas) of Fe as a function of reciprocal temperature (K^{-1})

Recommended: $\log_{10}\eta$ (mPas) = $-0.622 + (2478/T)$
(T is in K)

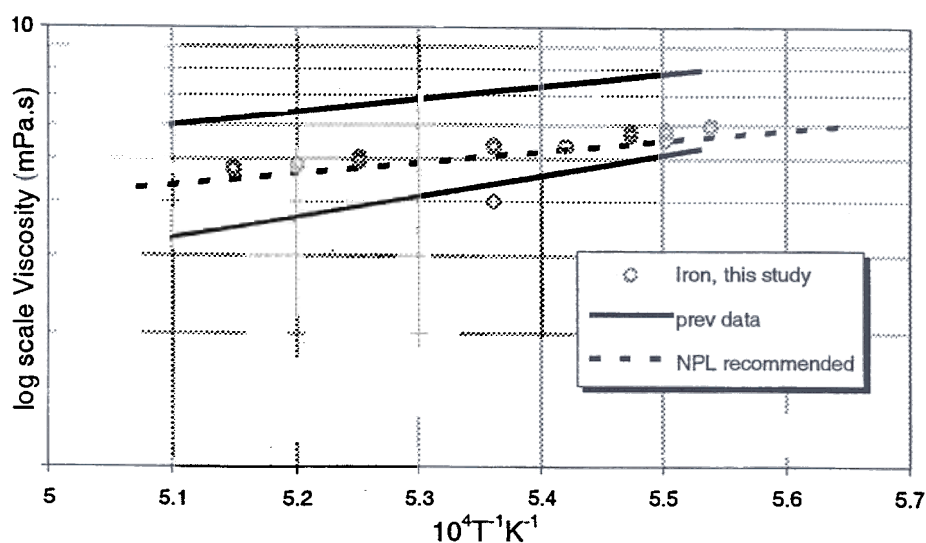


Figure 16 Logarithm of viscosity as a function of reciprocal temperatures for iron showing the results of present investigation, ----; and the limits of previously reported data in the literature [6,4], —

Comments:

The results obtained in this investigation are in excellent agreement with those reported recently by Yamamura et al [7].

4.7 Cu/Sn ALLOY

Crucible: Al₂O₃Gas: ArIdentifier: ATN

Composition, mass %

Cu	Sn	
90%	10%	Synthetic

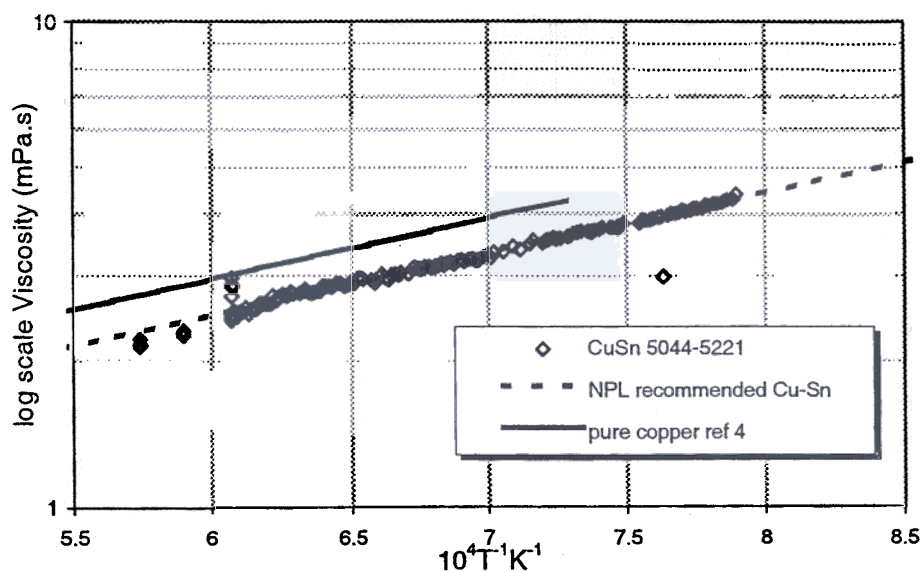


Figure 17 Logarithm of viscosity (mPas) of Cu/Sn alloy as a function of reciprocal temperature (K^{-1}). The measurements were carried out dynamically. The values for pure copper are shown as —

Recommended: $\log_{10}\eta(\text{mPas}) = -0.362 + (1263/T)$
(T in K)

Note these measurements were obtained for the dynamic mode.

Comments:

The viscosity of the alloy is lower than that of pure Cu, this may be related to the fact that Sn has a low viscosity.

4.8 Ni-ALLOY IN 718

Crucible: Al₂O₃, Mo capGas: ArIdentifier: EMT

Composition, (mass %)

Al	C	Co	Cr	Cu	Fe	Mn	Mo	Nb	Ni	Si	Ti
0.5	0.08	1	19	0.3	16.7	0.35	3.1	5.2	52.5	0.35	0.9

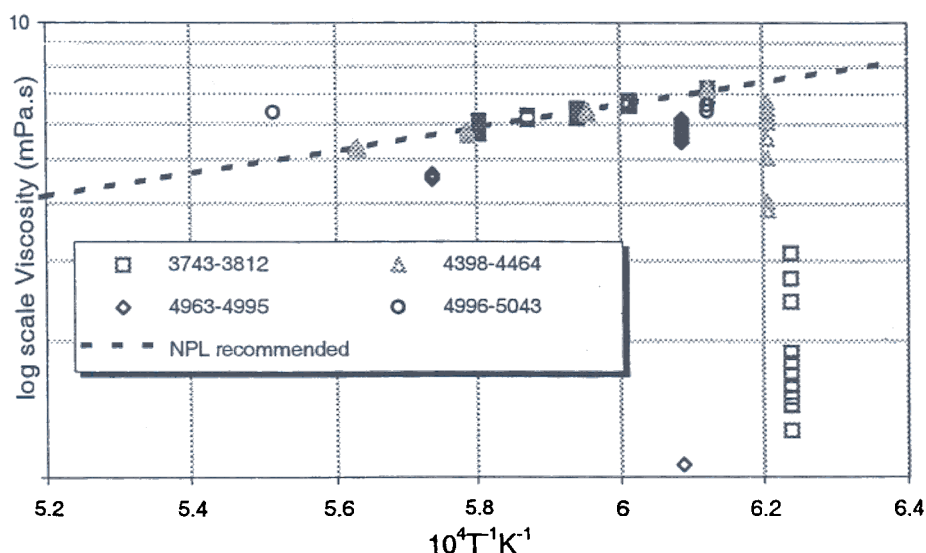


Figure 18 Logarithm of viscosity (mPas) of IN 718 as a function of reciprocal temperature (K⁻¹)

Recommended: $\log_{10}\eta(\text{mPas}) = -0.680 + (2500/T)$
(T in K)

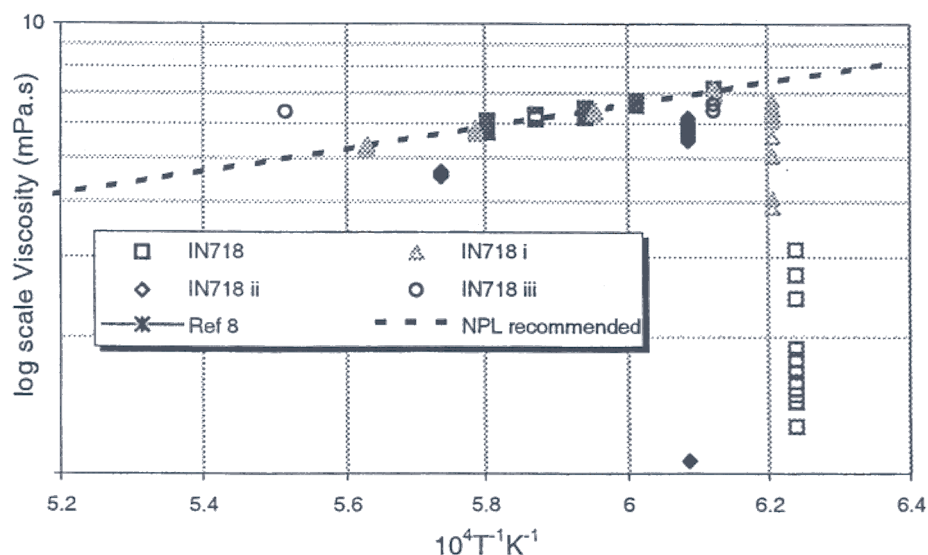


Figure 19 Logarithm of viscosity as a function of reciprocal temperature for IN 718; - - this investigation; -x-, Overfelt *et al* [8]

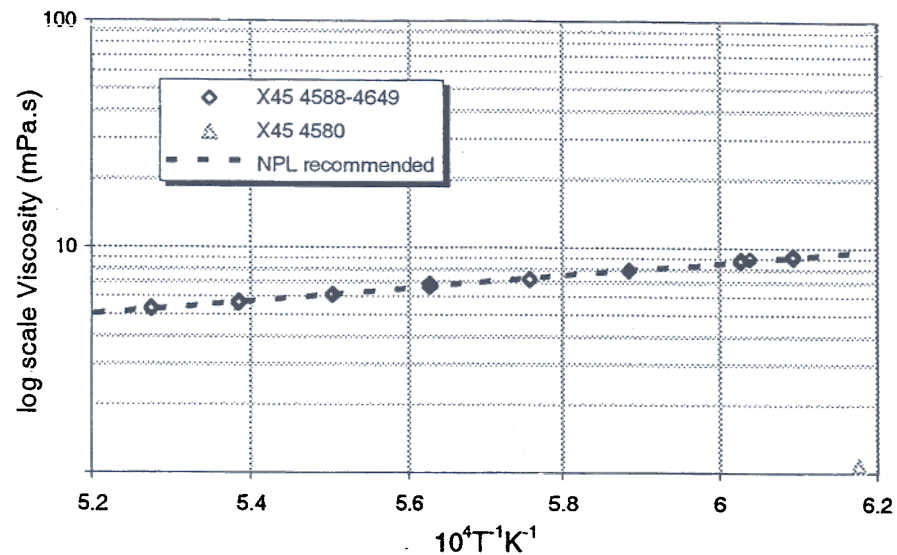
Comments:

There is excellent agreement with the values reported by Overfelt [8]. Note the apparent decrease in viscosity and large scatter for data < T_{liq} at 1337 °C which is in excellent

4.9 Co ALLOY - X45

Crucible: Al₂O₃, Mo capGas: ArIdentifier: EMYComposition (mass %)

Co	C	Cr	Fe	Mn	Ni	P	S	Si	W
52.6	0.25	25.5	2.0	0.7	10.5	0.04	0.04	0.8	7.5

Figure 20 Logarithm of viscosity (mPas) of X45 as a function of reciprocal temperature (K⁻¹)

Recommended: $\log_{10}\eta$ (mPas) = $-0.797 + (2884/T)$
(T in K)

4.10 EUTECTOID STEEL

Crucible: Al₂O₃, Mo cap

Gas: Ar

Identifier: PAF

Composition in mass %

	Fe	C	Si	Mn	Cr	Ni	Al	S	P	N
BST analysis	98.4	0.74	0.24	0.52	0.018	0.02	0.001	0.013	0.009	0.0054

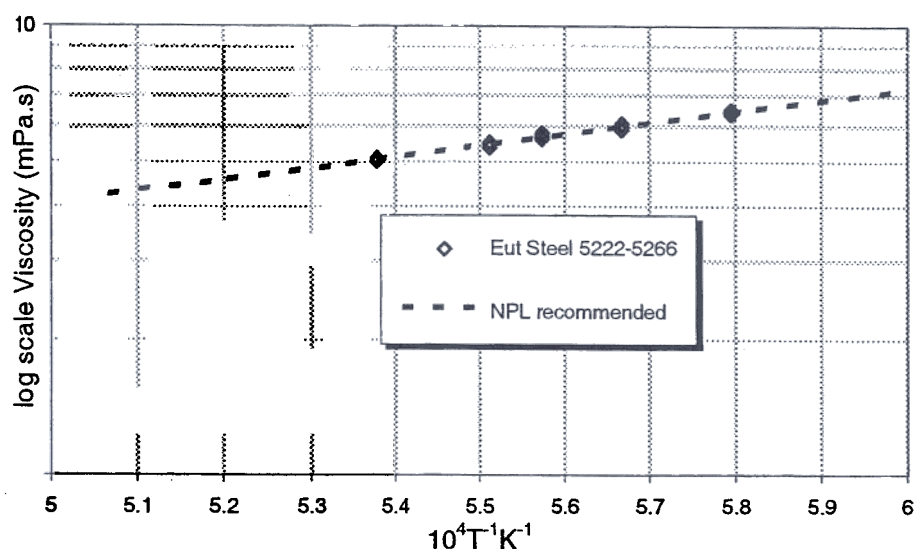


Figure 21 Logarithm of viscosity (mPas) of eutectoid steel as a function of reciprocal temperature (K⁻¹)

Recommended: $\log_{10}\eta$ (mPas) = $-0.6272 + (2478/T)$
(T in K)

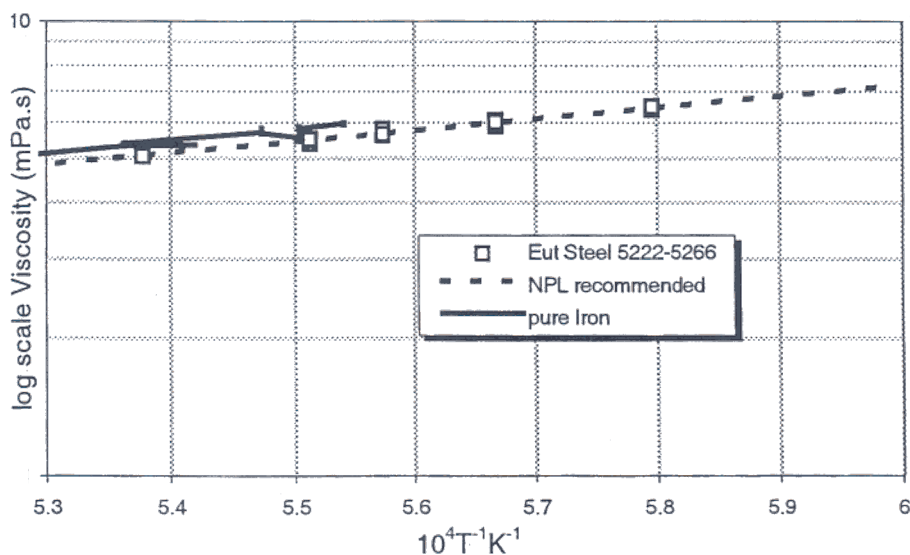


Figure 22 Logarithm of viscosity as a function of temperature for eutectoid steel; present results compared with those obtained for pure Fe, —.

Comments:

The viscosity of the steel is only slightly higher than that of pure Fe.

4.11 LOW CARBON STEEL

Crucible: Al₂O₃, Mo cap
Composition in mass %

Gas: Ar

Identifier: ATQ

	Fe	C	Mn	P	Cu	Al	Si	S	Ni	N
BST analysis	99.6	0.05	0.21	0.013	0.03	0.04	0.01	0.016	0.02	0.0056

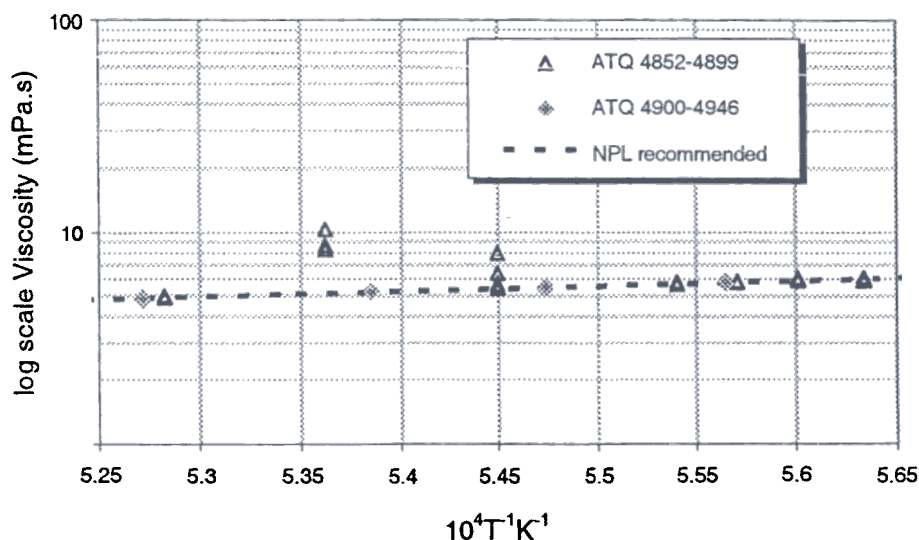


Figure 23 Logarithm of viscosity (mPas) of low carbon steel as a function of reciprocal temperature (K⁻¹)

Recommended: $\log_{10}\eta$ (mPas) = $-0.653 + (2543/T)$
(T in K)

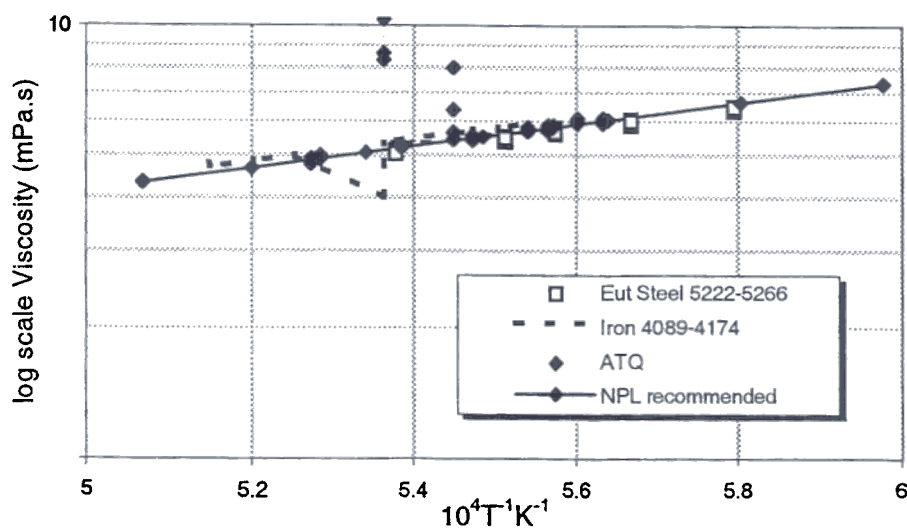


Figure 24 Logarithm of viscosity as confirmation of reciprocal temperature; —◆— present results; - - -, Fe; □, eutectoid steel.

Comments:

There is little difference in the viscosities of steels and pure Fe.

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