International Journal of Thermophysics, Vol. 20, No. 6, 1999

Viscosity Measurements of Industrial Alloys Using the Oscillating Cup Technique[1]

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Molten metal processing can be effectively simulated using state-of-the-art computer algorithms, and manufacturers increasingly rely upon these tools to optimize the design of their operations. Reliable thermophysical properties of the solid, solid + liquid, and liquid phases are essential for effective computer simulation. Commercially available instruments can measure many of the required properties of molten metals (e.g., transformation temperatures, thermal conductivity, specific heat, latent heat, and density). However, there are no commercially available instruments to characterize several important thermophysical properties (e.g., emissivity, electrical resistivity, surface tension, and viscosity). Although the literature has numerous examples of measurements of surface tension using the sessile drop and the oscillating drop techniques, literature references are sparse with regard to measurements of emissivity, electrical resistivity, and viscosity. The present paper discusses the development of an oscillating cup viscometer and its application to characterizing the viscosity of fully molten industrial alloys. The theory behind the oscillating cup technique is reviewed, and the design details of the current instrument are discussed. In addition, experimental data of the viscosity of several nickel-based superalloys are presented.

KEY WORDS: molten metals; oscillating cup; superalloy; viscosity.

1. INTRODUCTION

Casting manufacturers increasingly rely upon state-of-the-art computational models of their casting processes to ensure the highest casting quality

[1] Paper presented at the Thirteenth Symposium on Thermophysical Properties, June 22–27, 1997, Boulder, CO, U.S.A.

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0195-928X/99/1100-1791$16.00/0

while maintaining the lowest possible costs. A critical need for the industry is the development of both publicly available as well as proprietary databases of critically evaluated thermophysical property data of industrial alloys in their molten and solidifying states. Although measurements of the viscosities of molten metals have been reported using several techniques, the dominant technique at moderate to high temperatures is the oscillating cup technique. In this method, a molten metal is contained within a ceramic vessel suspended by a torsional pendulum. Torsional oscillations are then induced, and the resulting motion is damped primarily by viscous dissipation within the molten metal under investigation. The viscosity of a molten sample can be determined by measuring the time period and decay of the oscillations. The principal advantages of this technique are its mechanical simplicity and the ability to measure the time period and amplitude decay with great precision.

The motion of a torsional pendulum undergoing damped oscillations can be described by

$$\theta(t) = \theta_0 \exp\left(\frac{\delta}{\tau} t\right) \cos\left(\frac{2\pi}{\tau} t + \psi\right) \tag{1}$$

where $\theta(t)$ is the time-dependent angular displacement, θ_0 is the initial angular displacement, δ is the logarithmic decrement of the amplitude of oscillation, τ is the period of oscillation, t is the time, and ψ is the oscillatory phase shift. Only the logarithmic decrement and the time period need to be measured for calculating the viscosity of a molten sample. These can be obtained by a best fit of Eq. (1) to the observed motion of the torsional suspension system.

A number of analytical equations have been theoretically developed and experimentally tested to relate the observed time period and decrement of the oscillating assembly to the sample viscosity [1–7]. Roscoe's approximation [4, 5] has been widely used and is considered to provide accurate values of viscosity [8], especially when a small empirical correction factor is employed [9]. Abe et al. [10] note that Roscoe's approximation is consistently within 0.6–1.5% of the more rigorous analytical solutions. In fact, application of Roscoe's corrected formula has been shown to accurately reproduce calibration quality viscosity data for mercury, lead, tin, bismuth, and indium obtained using the well-accepted capillary technique [9].

For an oscillating cylindrical vessel, Roscoe's corrected equation for the viscosity μ is [9]

$$\frac{\delta}{\rho} = \zeta \left\{ A \left(\frac{\mu}{\rho}\right)^{1/2} - B\left(\frac{\mu}{\rho}\right) + C\left(\frac{\mu}{\rho}\right)^{3/2} \right\} \tag{2}$$

where

$$A = \frac{\pi^{3/2}}{I}\left(1 + \frac{R}{4H}\right) HR^3\tau^{1/2}$$

$$B = \frac{\pi}{I}\left(\frac{3}{2} + \frac{4R}{\pi H}\right) HR^2\tau$$

$$C = \frac{\pi^{1/2}}{2I}\left(\frac{3}{8} + \frac{9R}{4H}\right) HR\tau^{3/2}$$

R is the internal radius of the oscillating vessel, I is the moment of inertia of the torsional assembly including the sample, H is the height of the molten metal, ρ is the density of the molten metal, and ζ is an experimentally determined correction factor dependent upon the construction of the suspension system and the design and materials of the oscillating vessel. The correction factor must be evaluated for the specific apparatus utilized from experiments with low melting-point metals of known viscosity, e.g., mercury, lead, tin, etc. Iida and Guthrie [9] report an average correction factor of approximately 1.04 for a series of investigations. All dimensions and torsional inertias must be corrected for thermal expansion effects. An iterative numerical procedure of successive approximation is required to solve Eq. (2) for the unknown viscosity.

2. EXPERIMENTAL

Figure 1 shows the oscillating viscometer used in this study [11]. The inertia bar/crucible assembly is suspended with a single 56 cm long and 0.25 mm diameter Pt–10% Rh wire. The vacuum system is designed to enable crucible/sample exchange without mishandling the suspension wire and inadvertantly changing its elastic properties. Solid samples $\sim$5 cm long $\times \sim$1 cm in diameter were placed in the bottom of flat-bottomed, high-purity alumina crucibles. Torsional impulses to the oscillator for initial excitation were generated through a rotary vacuum feedthrough by a computer-driven stepping motor at the top of the system. An inertia bar can be exchanged to alter the overall inertia and period of the oscillator system. Two bars of known inertia were utilized to enable calculation of the unknown inertia of the overall oscillator system. A HeNe laser is reflected from a mirror mounted on the inertia bar/crucible assembly, and the oscillations of the reflected laser beam are detected by two photodiodes at fixed angular positions. The photodiode signals are monitored by a personal computer. A 3.2-cm-diameter high-purity alumina tube with o-ring

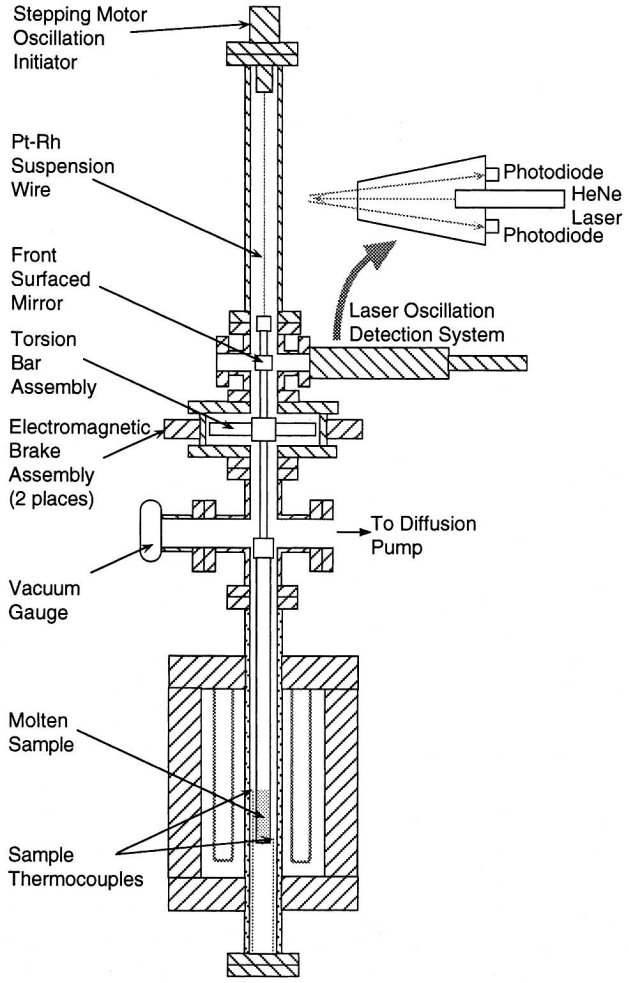

Fig. 1. Schematic of the high-temperature oscillating cup viscometer.

seals serves as the vacuum furnace chamber. The oscillator's vacuum system is pumped with a 5-cm diffusion pump from the flange shown. High-purity argon gas at a pressure of 30 mTorr was used to suppress excess vaporization of the metal and minimize the atmospheric drag on the oscillator. A resistively heated clam-shell furnace regulated by a PID controller is wrapped around the alumina retort tube. Temperature control of ± 0.5°C at 1500°C is realized with this system. As shown in Fig. 1, two Pt–10% Rh thermocouples, axially spaced 5 cm apart, are located within

the retort adjacent to the crucible/sample to ensure an axial temperature uniformity of $\pm 0.5°C$ in the sample region. The thermocouples were calibrated to within 1 K with a pure nickel reference sample. In addition, prior to beginning experimentation, a separate sample with an internal thermocouple was utilized to determine the temperature difference between the actual sample temperature and the external thermocouple readings. This calibration was repeatable to within 1 K.

Extremely straight, flat-bottomed crucibles were prepared by sealing one end of an extruded high-purity alumina tube with alumina cement and then completely curing the cement for 0.5 hr at $\sim 200°C$. The specimen-crucible assembly was inserted into the alumina retort tube placed in the center of the vertical furnace system and evacuated to approximately 30 mTorr. The retort was back-filled with argon and then reevacuated two additional times. The furnace was heated to the operational temperature regime at the rate of $400°C \cdot h^{-1}$.

Small amounts of additional damping arise due to internal friction in the suspension wire and viscous damping due to the low-pressure cover gas. These extraneous torques also act against the motion of the pendulum and are linearly additive to the viscous torques from the samples. These contributions are often determined by running separate experiments with empty crucibles to eliminate liquid damping. However, in the present investigation, the additional decrements were most easily determined for each sample/crucible assembly by repeating oscillation experiments after the samples froze (preventing viscous damping from the molten samples). This approach eliminated extra handling of the torsional suspension system and also maintained a constant axial load on the suspension wire for the determination of each sample's intrinsic decrement. The corrections in decrement were typically of the order of 15%.

Oscillations were initiated by quickly rotating the stepping motor at the top of the pendulum by a total angle of 7°. Data were taken at 100 Hz over the course of approximately 35 oscillation cycles. The logarithmic decrement and oscillation period were obtained by curve fitting Eq. (1) to the times that the reflected HeNe laser beam crossed the two photodetectors [11]. Correlation coefficients of the curve fits typically exceeded 0.9995. After the completion of data acquisition during an experiment, electromagnetic braking coils were energized to stop the oscillations of the crucible assembly over a period of approximately 1 min. Fifteen minutes was allowed between individual experiments to enable the fluid motions in the crucible to completely damp out.

A Roscoe equation correction factor of 1.025 was determined from room-temperature experiments with mercury. This is consistent with the average literature value of $\zeta = 1.04$ [9].

Table I. Compositions (wt %) of the Nickel-Based Superalloys Investigated

Alloy	Ni	Cr	Fe	Nb	Mo	Ti	W	Ta	C	Al	Co
IN 718	BAL	18.3	18.9	4.6	2.0	0.8	—		0.0	0.6	—
IN 939	BAL	22.2	0.1	1.5	—	3.9	2.2	2.0	0.1	2.0	18.5
Mar-M-247	BAL	8.4	—	—	0.7	1.0	10.0	3.0	0.2	5.5	10.0

Samples of three nickel-based superalloys were investigated. The compositions of the superalloys are given in Table I.

3. RESULTS AND DISCUSSION

3.1. Experimental

The absolute viscosities of the three superalloys were experimentally evaluated over the temperature range of 1375 to 1500°C. Density data of the molten alloys were characterized in separate investigations or were theoretically estimated [12–14]. Two samples were utilized for each set of experiments and none showed any evidence of crucible reactions around their circumferences or their bottoms and only minimal oxidation on their free surfaces (tops). Viscosity measurements were made both during the heating cycle (three measurements at each temperature) and during the cooling cycle (three measurements at each temperature) for each sample. No systematic discrepancies in viscosity determinations were detectable between the samples or between whether the samples were being heated or cooled for the measurements. The viscosity data were averaged at each temperature for each alloy, and the temperature dependence of these mean viscosities are shown in Table II.

Figure 2 shows the behavior of the log of the viscosity versus $1/T$ for each of the three nickel-based superalloys. The data for each alloy are linear over this limited temperature range. Through a least-squares curve fit procedure, the Arrhenius equations describing the behavior were determined to be

$$\text{Alloy 718:} \quad \mu(\text{mPa} \cdot \text{s}) = 0.18 \exp\left(\frac{50.2}{RT}\right) \tag{3}$$

$$\text{Alloy 939:} \quad \mu(\text{mPa} \cdot \text{s}) = 0.092 \exp\left(\frac{60.3}{RT}\right) \tag{4}$$

$$\text{Mar-M-247:} \quad \mu(\text{mPa} \cdot \text{s}) = 0.077 \exp\left(\frac{64.9}{RT}\right) \tag{5}$$

Table II. Temperature Dependence of the Viscosity of the Nickel-Based Superalloys Shown

Temperature (°C)	Viscosity (mPa·s)		
	IN718	IN939	Mar-M-247
1350	7.40	–	–
1375	7.12	7.61	–
1380	–	7.33	8.48
1400	6.43	7.06	8.00
1420	–	6.54	7.98
1425	6.17	6.39	–
1440	–	6.15	7.31
1450	6.02	6.15	–
1460	–	6.09	6.98
1475	5.69	5.86	–
1480	–	5.79	6.73
1500	–	5.46	6.04

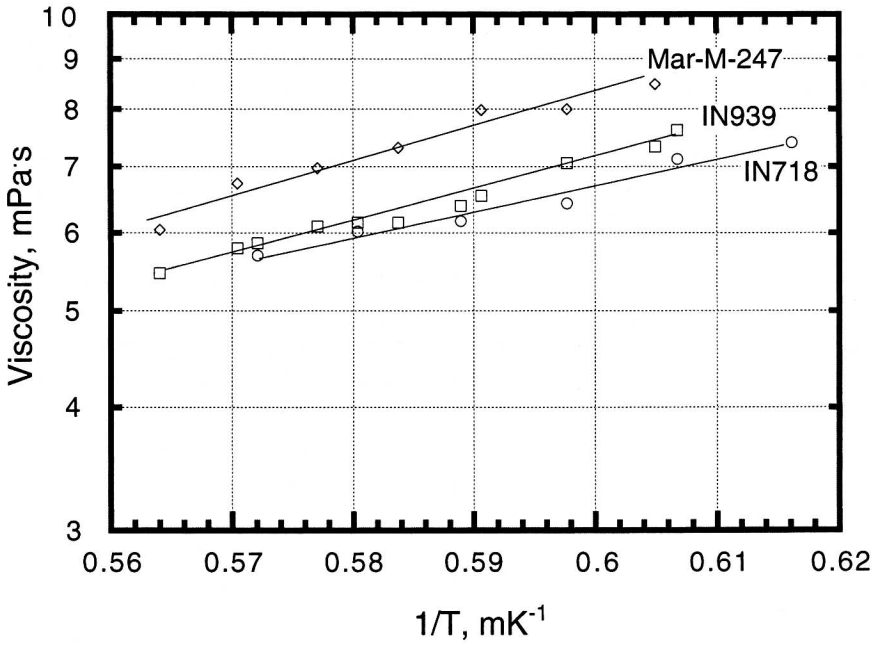

Fig. 2. Arrhenius plot ($\log \mu$ versus $1/T$) of the measured viscosity of the three superalloys.

where R is the universal gas constant ($0.008314\ \text{kJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$) and T is the absolute temperature in K. The correlation coefficients of the curve fits to the experimental data were all greater than 0.98. The activation energies for viscous flow were found to be $50.2\ \text{kJ} \cdot \text{mol}^{-1}$ for superalloy 718, $60.3\ \text{kJ} \cdot \text{mol}^{-1}$ for superalloy 939, and $64.9\ \text{kJ} \cdot \text{mol}^{-1}$ for Mar-M-247.

3.2. Uncertainty Estimate

The total uncertainty, ΔG, in any experimental measurement can be estimated using the procedure of Moffat [15]. When j independent variables are utilized in a function G, the individual contributions, ΔX_i, to the total uncertainty, ΔG, can be estimated by the root-sum-square method. Thus

$$\Delta G = \left[\left(\frac{\partial G}{\partial X_1} \Delta X_1 \right)^2 + \left(\frac{\partial G}{\partial X_2} \Delta X_2 \right)^2 + \cdots + \left(\frac{\partial G}{\partial X_j} \Delta X_j \right)^2 \right]^{1/2} \tag{6}$$

where the partial derivative of G with respect to X_i is the sensitivity coefficient for the function G with respect to the measurement X_i.

The internal diameter of the crucible was carefully measured prior to sealing the bottom. Since the extruded alumina tubes had been fired prior to the low-temperature cement curing step and were thus quite stable, further changes in diameter from additional sintering effects during cement curing were neglected. The inertia of the torsion assembly and the oscillating vessel radius can both be calculated to within 0.1% using the thermal expansion coefficients of the materials utilized. Since the correlation coefficients of the curve fits for determining decrement and period typically exceeded 0.9995, it is estimated that the percentage uncertainty in decrement and period is approximately $\pm 0.1\%$. The molten metal density is generally known to no better than within 2% [10]. Although the thermal expansion of the crucible and alloy are usually well established up to the melting region of the alloy [10], expansion of the alloy due to melting and free surface meniscus effects preclude knowing the height of the liquid column to better than about 2%. The uncertainty in sample temperatures are approximately 2.5 K or 0.15%. At a temperature of 1350°C, a simple calculation using Eq. (3) for superalloy 718 shows a corresponding uncertainty in measured viscosity of $0.05\ \text{mPa} \cdot \text{s}$ (0.8%). Finally, although the Roscoe equation correction factor was carefully evaluated at room temperature for the current experimental apparatus ($\zeta = 1.025$), its application at elevated temperatures is a cause for concern and its uncertainty must also be of the order of 1% ($\zeta = 1.025 \pm 0.01$), consistent with the average correction factor of 1.04 [9].

Table III. Uncertainty Estimates for Oscillating Cup Measurement of Viscosity of Superalloy 718

Parameter	Estimated $\pm 2\sigma$ confidence limits (%)	Viscosity change (mPa · s)	Viscosity change squared $(\text{mPa} \cdot \text{s})^2$
Assembly inertia, $I = 495\ \text{g} \cdot \text{cm}^2$	0.10	0.02	0.0004
Oscillating vessel radius, $R = 0.483$ cm	0.10	0.02	0.0004
Molten metal height, $H = 5.32$ cm	2.00	0.33	0.1089
Molten metal density, $\rho = 7.35\ \text{g} \cdot \text{cm}^{-3}$	2.00	0.12	0.0144
Measured decrement, $\delta = 0.00543$	0.10	0.02	0.0004
Measured oscillation period, $\tau = 2.435$ s	0.10	0.01	0.0001
Correction factor, $\zeta = 1.025$	1.00	0.17	0.0289
Temperature measurement, $T = 1648$ K	0.15	0.05	0.0025
Total uncertainty in viscosity, mPa · s $[\sum (\Delta\mu_i)^2]^{1/2}$			0.395
Total % uncertainty in viscosity (5.9 mPa · s)			6.7

Moffat's uncertainty estimation procedure [15] was numerically applied to the corrected Roscoe equation, Eq. (2), and the temperature uncertainty for superalloy 718 and the results are shown in Table III. The total estimated measurement uncertainty (95% confidence limits) from the oscillating cup technique is approximately 6.7% for the viscosity of superalloy 718. The largest contributors to the total uncertainty are the uncertainties in molten metal height, the Roscoe equation correction factor, and the molten alloy density.

4. SUMMARY

The oscillating vessel technique is an excellent technique for measuring the viscosity of molten metals when crucible contamination is not a concern. No reactions between the superalloys and the alumina crucibles were seen in this investigation. The total estimated measurement uncertainty (95% confidence limits) is 6.7% for the viscosity of these superalloys. The largest contributors to uncertainty in the oscillating cup measurements are uncertainties in the molten metal height, the Roscoe equation correction factor, and the molten alloy density.

The viscosities of three nickel-based superalloys exhibited Arrhenius behavior between 1350 and 1500°C and are described by an equation of the type

$$\mu(\text{mPa} \cdot \text{s}) = \mu_0 \exp\left(\frac{Q}{RT}\right)$$

where R is the universal gas constant ($0.008314\ \text{kJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$) and T is the absolute temperature in K. The correlation coefficients of the curve fit to the experimental data were greater than 0.98. The activation energies (Q) for viscous flow were found to be 50.2, 60.3, and 64.9 kJ · mol-1 for nickel-based superalloys 718, 939, and Mar-M-247, respectively.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the financial support received from NASA's Office of Space Access and Technology under Grant NAGW-1192, from ARPA under Agreement MDA972-93-2-0001, and from the Investment Casting Cooperative Arrangement (Howmet Corporation, PCC Airfoils Inc., General Electric Aircraft Engines, United Technologies Corp., and UES Inc.) chaired by Howmet Corporation.

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