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Oscillating Cup Viscosity Measurements of Aluminum Alloys: A201, A319 and A356[1]

D. Wang[2] and R. A. Overfelt[2, 3]

An oscillating cup viscometer was developed to measure the absolute viscosities of molten metals. Previous experiments established the capability of the apparatus to characterize the viscosities of molten nickel-based superalloys. However, modifications to the instrument and its theoretical analysis were required for reliable measurements on molten aluminum alloys, presumably due to their lower densities and lower viscosities. The theoretical literature for the fluid flow inside an oscillating cup is reviewed, and a working equation without any correction factor is developed for the improved viscometer. Some design parameters of the viscometer that directly affect the accuracy of viscosity estimation by using the working equation are discussed. A special vertical furnace was adopted to uniformly heat a longer cylindrical sample (10 mm inner diameter and 120 mm length) with a temperature difference of less than 2°C over the sample length. The measuring procedure was also improved to get more accurate motion parameters. It is estimated that the working equation and improved instrument provide an uncertainty of less than 4%. In addition, applications and experimental data are presented for pure aluminum and three aluminum alloys: A201, A319, and A356.

KEY WORDS: aluminum alloys; molten metals; oscillating cup; viscosity.

1. INTRODUCTION

The oscillating cup viscometer has become a primary technique to measure the absolute viscosities of high temperature liquids [1–3]. In the viscometer, a high temperature liquid (such as a molten metal) is contained within

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[2] Department of Mechanical Engineering, 201 Ross Hall, Auburn University, Auburn, Alabama 36849, U.S.A..

[3] To whom correspondence should be addressed. E-mail: overfra@auburn.edu

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a crucible suspended by a wire to form a torsional pendulum, which induces torsional oscillation motion. This motion is damped primarily by viscous dissipation within the viscous liquid inside the crucible. The viscosity of the liquid can be calculated by an analytical or numerical solution of the equations of motion of the oscillating cup system. The principal advantages of this technique are its mechanical simplicity and the ability to measure the time period and amplitude decay with great precision.

Since the 1960s, a number of successful viscometers and their working equations have been developed to measure the viscosities of liquids at high temperature [4–6]. But, there are still large discrepancies between laboratories, sometimes amounting to 50%. It is commonly considered that the errors come from the different kinds of viscometers and viscosity estimation methods. Further study of the principle of viscometers and their working equations is still very important to improve these measurement techniques and obtain reliable viscosity data for science and industry.

Roscoe [7, 8] proposed an straightforward approximate method to calculate the viscosity from the measured motion parameters. Advantages of the method are that it is simple to use and easy to understand, so this method is still often employed [2]. Kestin and Newell [9] and Beckwith and Newell [10] provided another analytical method and working equation to calculate viscosities of liquids from measurements of the oscillation parameters. This method does not require a correction factor since an exact solution of the equation of motion of oscillating cup systems is given. One of the primary advantages of this method is its mathematical rigor with calculation errors less than 0.01%. Thus, this method is preferred by many investigators. Ohta et al. [11] used this method for their viscometer with oscillations of a spherical body. Torklep and Oye [12] also used it for their new-generation oscillating cup viscometer, and they presented a set of simplified calculation formulae. Nunez et al. [3] also used the method for their new high-temperature viscometer to measure the viscosity of molten salts. However, because of the elimination of the correction factor, this method cannot correct for any other errors which come from the viscometer itself or the measuring procedure, such as the determination of a stiffness parameter, inertial damping and inertial moment of the oscillation system, or data acquisition, curve fit of a harmonica function, nonlinearity of oscillations of the system, or turbulent flow in the liquid. These effects also can introduce error in the viscosity measurement that cannot be corrected in the working equation. Thus, clear understanding of the theory of the oscillating cup is required to obtain reliable data.

In this paper, a new working equation without a correction factor is developed for systems with small dimensionless radii. The details of the equation are given by motion analysis of the system that defines the relationships among viscosity and the damping oscillation motion parameters.

2. MOTION ANALYSIS

In an oscillating cup system, a cup with a viscous liquid is suspended by an elastic wire. The cup is forced to rotate through an angle along the wire axis and then held motionless (see Fig. 1). When the cup is released, it will freely oscillate. If the oscillation is considered as a simple harmonic motion, the two oscillation motion parameters, oscillation frequency ω and damping parameter Δ, can be measured by a curve fit technique. The viscosity of an experimental liquid can be calculated from the two motion parameters, other physical parameters of the system, and size of the sample.

For an empty oscillating cup, a simple harmonic oscillation can be described as

$$\alpha(t) = \alpha_0(t)\, e^{-\Delta_0 \omega_0 t} \sin(\omega_0 t + \phi) \tag{1}$$

where $\alpha(t)$ is the angular displacement of the body from equilibrium, $\alpha_0(t)$ is the initial angular displacement, ϕ is an oscillatory phase shift, ω_0 is an angular frequency of the cup system without the liquid, and Δ_0 is a logarithmic decrement of the amplitude of the oscillation without the viscous liquid, which is caused by the internal friction of the wire and the resistance in the surrounding air.

Based on mechanical dynamics, the equation of the cup without liquid can be written as

$$I_0 \omega_0^2 \left[\frac{d^2\alpha(\tau)}{d\tau^2} + 2\Delta_0 \frac{d\alpha(\tau)}{d\tau} + (1 + \Delta_0^2)\, \alpha(\tau) \right] = 0 \tag{2}$$

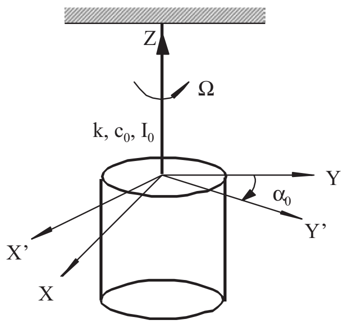

Fig. 1. Torsional pendulum.

with initial conditions,

$$\text{for} \quad t=0, \quad \alpha(t)=\alpha_0 \quad \text{and} \quad \frac{d\alpha(t)}{dt}=0.$$

The angular frequency and damping parameter are

$$\omega_0^2=\frac{k}{I_0}-\left(\frac{c_0}{2I_0}\right)^2 \tag{3}$$

$$\Delta_0=\frac{c_0}{2I_0\omega_0} \tag{4}$$

where I_0, c_0, and k are, respectively, initial moment of the cup, damping coefficient of the suspended wire, and stiffness coefficient of the suspended wire. τ is a dimensionless unit of time,

$$\tau=\omega_0 t \tag{5}$$

If the oscillating cup contains a viscous liquid, the friction force that comes from the liquid can be considered as an external applied force and placed on the right side of Eq. (2). So, the motion equation of the cup system with the viscous liquid becomes

$$I\omega_0^2\left[\frac{d^2\alpha(\tau)}{d\tau^2}+2\Delta_0\frac{d\alpha(\tau)}{d\tau}+(1+\Delta_0^2)\,\alpha(\tau)\right]=M(\tau) \tag{6}$$

where I is an initial moment of the cup system with the liquid and $M(\tau)$ is a torque around the z axis caused by the friction force of a liquid. By using the Laplace transform, the above motion equation is rewritten as the function of a complex frequency s.

$$[(s+\Delta_0)^2+1]\,\bar{\alpha}(s)-\frac{\bar{M}(s)}{I\omega_0^2}=(s+2\Delta_0)\,\alpha_0 \tag{7}$$

where $\bar{\alpha}(s)$ and $\bar{M}(s)$ are, respectively, the transforms of $\alpha(\tau)$ and $M(\tau)$.

If the liquid inside the oscillating cup is considered as an ideal viscous fluid, the rate of flow is a function of the stress. The ratio of applied shear stress to the rate of shear for an ideal viscous body is the viscosity, η. If the viscous body is a Newtonian fluid, η is a constant. The torque $M(\tau)$ caused by the total friction in the liquid is a function of the velocity of the liquid

$$M(\tau)=\eta\iint_A r^2\frac{\partial\Omega}{\partial n}\,d\sigma \tag{8}$$

in which r is the radius of the cylindrical cup, Ω is an angular velocity around the z axis, A denotes the surface of contact between the liquid and the oscillating cup, and n is the normal direction of the fluid motion. If the viscosity is expressed by a relative kinematic viscosity, μ, Eq. (8) is rewritten as

$$M(\tau) = \mu\rho \iint_A r^2 \frac{\partial \Omega}{\partial n}\, d\sigma \tag{9}$$

where

$$\mu = \eta / \rho$$

and ρ is the density of a liquid.

The equation of motion of the liquid is described by the Navier–Stokes equation:

$$\rho \frac{\partial \vec{u}}{\partial t} + \vec{u} \cdot \nabla \vec{u} = \rho g - \nabla p + \mu\, \Delta \vec{u} \tag{10}$$

The usual assumption of no secondary motion is invoked and the nonlinear terms in Eq. (10) are eliminated. Based on this assumption, Eq. (10) for cylindrical polar coordinates, α, r, and z (see Fig. 2) is rewritten as

$$\frac{\partial \Omega}{\partial t} = \mu \left\{ \frac{\partial^2 \Omega}{\partial r^2} + \frac{3}{r} \cdot \frac{\partial \Omega}{\partial r} + \frac{\partial^2 \Omega}{\partial z^2} \right\} \tag{11}$$

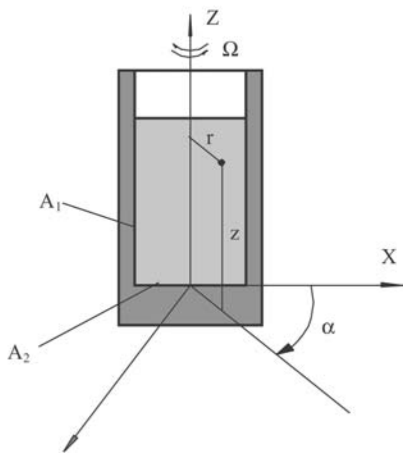

Fig. 2. Fluid in the oscillating sample cup.

When R and H are, respectively, the radius and height of a cylindrical sample, the boundary conditions to be satisfied by Ω are

- initial condition: $t=0$, $\Omega(r, z, t)=0$. The fluid is initially at rest.
- boundary conditions: $r=R$, $\Omega(r, z, t)=d\alpha/dt$. There is no slip at the boundary between the fluid and the cup. α is an angle of the pendulum system from its equilibrium position.

It is convenient to use dimensionless space coordinates by adopting an average boundary layer thickness, $\delta=\sqrt{\mu/\omega_0}$. Equation (11) is rewritten, with $\xi=r/\delta$ and $\eta=z/\delta$, as

$$sw=\frac{\partial^2 w}{\partial \xi^2}+\frac{3}{\xi}\cdot\frac{\partial w}{\partial \xi}+\frac{\partial^2 w}{\partial \eta^2} \tag{12}$$

where w is another dimensionless variable,

$$w=\frac{\bar{\Omega}}{\omega_0(s\bar{\alpha}-\alpha_0)}. \tag{13}$$

Equation (13) also results in a simplified boundary condition

$$w(\xi, \eta, s)=1 \qquad \text{on } A$$

Equation (12) can be solved by the well-known separation of variables technique [9].

$$w(\xi, \eta, s)=\frac{\cosh\sqrt{s}(\eta_0-\eta)}{\cosh\sqrt{s}\,\eta_0}+\sum_{m=0}^{\infty}\frac{\xi_0\beta_1(s_m\xi)}{\xi\beta_1(s_m\xi_0)}(\xi, s)\sin\frac{(2m+1)\,\pi\eta}{2\eta_0} \tag{14}$$

with

$$s_m^2=s+\left(\frac{(2m+1)\,\pi}{2\eta_0}\right)^2 \tag{15}$$

and

$$\xi_0=\frac{R}{\delta}, \qquad \eta_0=\frac{L}{\delta}, \tag{16}$$

and β_i denotes a Bessel function of ith order.

3. VISCOSITY ESTIMATION EQUATION

After the solution of the equation of motion for liquid flow in an oscillating cup is obtained, the friction force can be calculated using Eq. (9). Substituting this force into the system motion equation provides the system equation as

$$\frac{\bar{\alpha}(s)}{\alpha_0} = \frac{1}{s} - \frac{1+\Delta_0^2}{s[(s+\Delta_0)^2+1+D(s)]}, \tag{17}$$

with

$$D(s) = +\frac{\rho\delta^4}{I} s \iint_A \xi^2 \frac{\partial\omega}{\partial n} d\sigma \tag{18}$$

Using the solution of the motion Eq. (14), we obtain the analytic equation for $D(s)$

$$D(s) = s^2 \frac{I'}{I} \left\{ \frac{\tanh(\sqrt{s}\,\eta_0)}{\sqrt{s}\,\eta_0} + \frac{32s}{\pi^2\xi_0} \sum_{m=0}^{\infty} \frac{1}{(2m+1)^2 s_m^3} \frac{\beta_2(s_m\xi_0)}{\beta_1(s_m\xi_0)} \right\} \tag{19}$$

where

$$I' = \pi\rho H R^4/2$$

By inversion of the Laplace transform, the angular position now can be written as the integral,

$$\alpha(\tau) = \frac{\alpha_0}{2\pi i} \int_C e^{s\tau} \left\{ \frac{1}{s} - \frac{1+\Delta_0^2}{s[(s+\Delta_0)^2+1+D(s)]} \right\} ds \tag{20}$$

along any vertical contour C in the right-hand half of the complex plane.

Equation (20) can be evaluated by residue theory since the only singularities of the integrand are poles [9]. If we get the k roots, S_k of the following equation:

$$(S_k+\Delta_0)^2+1+D(S_k) = 0 \tag{21}$$

then Eq. (20) is solved as

$$\alpha(\tau) = -\alpha_0 \sum_k \frac{(1+\Delta_0^2)\exp(S_k\tau)}{S_k[2S_k+2\Delta_0+D(S_k)]} \tag{22}$$

The ultimate purpose is to deduce the value of viscosity from the observed behavior of the system oscillation. Assuming the oscillation of the cup system with viscous liquid is a damped harmonic motion, the motion, $\alpha(\tau)$, can be represented by simple harmonic motion plus a fast decay transient $f(t)$. Thus

$$\alpha(\tau) = \alpha_0 e^{-\Delta\theta\tau} \sin(\theta\tau + \phi) + f(\tau) \tag{23}$$

where $\theta = \omega/\omega_0$, and Δ is total damping observed with liquid in the cup. After a few oscillation cycles, the experimental system will behave as a simple harmonic oscillation according to

$$\alpha(t) \approx e^{-\Delta\theta\tau} \cos(\theta\tau + \phi) \tag{24}$$

where S_k, one of the roots of Eq. (21), is

$$S_k = \theta(-\Delta \pm i) \tag{25}$$

Substituting the above solution into Eq. (21), we obtain two equations by taking the real and imaginary parts,

$$\text{Re } D[\theta(-\Delta \pm i)] = -1 + \theta^2 - (\Delta\theta - \Delta_0)^2 \tag{26}$$

$$\text{Im } D[\theta(-\Delta \pm i)] = \pm 2\theta(\Delta\theta - \Delta_0) \tag{27}$$

and

$$D[(-\Delta \pm i)\,\theta] = (-\Delta \pm i)^2\,\theta^2 \frac{I'}{I} \left\{ \frac{\tanh(\sqrt{(-\Delta \pm i)\,\theta}\,\eta_0)}{\sqrt{(-\Delta \pm i)\,\theta}\,\eta_0} + \frac{32(-\Delta \pm i)\,\theta}{\pi^2 \xi_0} \sum_{m=0}^{\infty} \frac{1}{(2m+1)^2\, s_m^3} \frac{\beta_2(s_m \xi_0)}{\beta_1(s_m \xi_0)} \right\} \tag{28}$$

Torklep [12] used the three lowest terms of the Bessel expansion and two lowest terms of tanh function to give a simple and approximate viscosity estimation formula,

$$D(s) = s^2 \frac{I'}{I} \left(\begin{array}{l} \dfrac{4}{s^{1/2}\xi_0} - \dfrac{6}{s\xi_0^2} + \dfrac{3}{2s^{3/2}\xi_0^3} + \dfrac{3}{2s^2\xi_0^4} + \cdots \\ \quad + \dfrac{1}{s^{1/2}\eta_0} - \dfrac{16}{\pi s \xi_0 \eta_0} + \dfrac{9}{s^{3/2}\xi_0^2\eta_0} - \dfrac{8}{\pi s^2 \xi_0^3 \eta_0} + \cdots \end{array} \right) \tag{29}$$

Substituting Eq. (29) into Eqs. (26) and (27), Torklep then obtained a simplified viscosity estimation formula,

$$\frac{I'}{I}\left[-A(\Delta p+q)\,\theta^{1/2}\xi_0^{-1}+B\Delta\theta\xi_0^{-2}+Cp\theta^{3/2}\xi_0^{-3}+D\theta^2\xi_0^{-4}\right]$$
$$=-1/\theta^2+1^2-(\Delta-\Delta_0/\theta)^2 \qquad (30)$$

$$\frac{I'}{I}\left[A(p-\Delta q)\,\theta^{1/2}\xi_0^{-1}-B\theta\xi_0^{-2}+Cq\theta^{3/2}\xi_0^{-3}\right]=2\omega(\Delta\omega-\Delta_0) \qquad (31)$$

where

$$A=4+R/H$$
$$B=6+(16/\pi)(R/H)$$
$$C=(3/2)+9(R/H)$$
$$D=1.5-(8/\pi)(R/H)$$
$$p=1/\{2[\Delta+(1+\Delta^2)^{1/2}]\}^{1/2}$$
$$q=1/2p$$
$$\theta=\omega/\omega_0$$
$$\xi_0=R(2\pi\rho/\eta_D T)^{1/2}$$
$$I'=\pi\rho HR^4/2$$

When the dimensionless radius, ξ_0 is larger than 10, the approximate error of Torklep's simplified equation (Eqs. (30) and (31)) is less than 0.1%. This requirement can be satisfied for many oscillating cup viscometers. But for viscometers with ξ_0 less than 10, Torklep's equation will result in additional calculation error. In the case, it is recommended to use Eqs. (26)–(28), which still have high calculation accuracy for any value of the dimensionless radius.

4. APPLICATION

Figure 3 shows an oscillating cup viscometer from this study. In the system, an inertia bar with a crucible is suspended with a single steel wire of 56 cm length and 0.254 mm diameter. Solid samples were placed in the bottom of flat-bottomed graphite crucibles. The graphite crucible was coated by boron nitride to avoid chemical reaction between crucible and samples. The sample sizes were 1 cm diameter and 12 cm length. The end

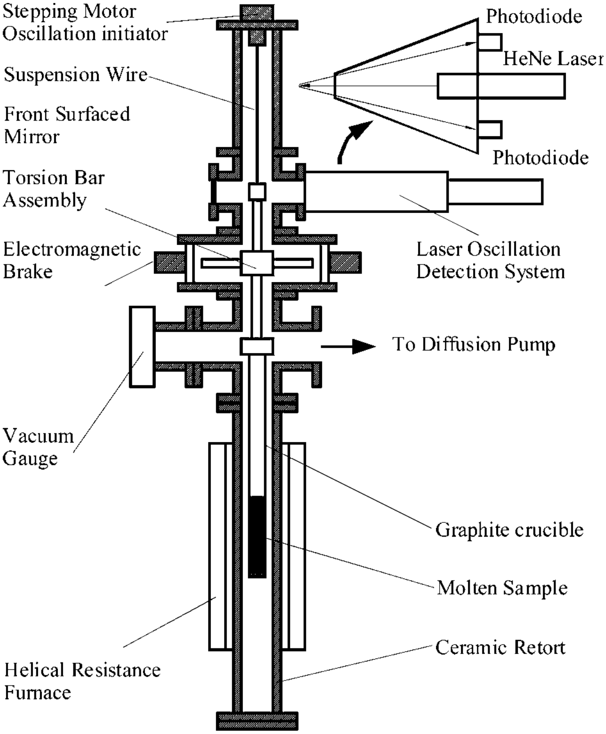

Fig. 3. Schematic of the oscillating cup viscometer.

effect of the samples was neglected here due to the small diameter and long length. Torsional impulses to the oscillator for initial excitation were generated through a rotary vacuum feed-through by a computer-driven stepping motor at the top of the 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 working vacuum chamber is pumped with a diffusion pump to 2×10^{-3} torr. A temperature-controlled furnace is used to heat the alumina retort tube and melt the sample. Two Pt-10% Rh thermocouples, axially spaced outside the crucible at the top and bottom of the sample, are used to ensure axial temperature uniformity on the test sample.

The system is initially motionless at an off equilibrium position of 5 degrees. When a test begins, a stepping motor at the top of the pendulum quickly returns the pendulum to equilibrium position, resulting in oscillation with an initial angle of 5 degrees and with an initial velocity of zero. The oscillating motion data were collected by a PC computer. A curve fit is used to fit measured timing-position data to a simple harmonic oscillation equation Eq. (25) and obtain the two main motion parameters, logarithmic decrement and oscillation period.

Previous experiments established the capability of the apparatus to characterize the viscosity of molten nickel-based superalloys [4]. The instrument has been recently improved to measure low density and low viscosity molten aluminum alloys. The following improvements have been performed on the instrument.

- The viscosity estimation model presented above is used to calculate the viscosity from the measured motion parameters.
- Length of a sample is increased from 5 to 12 cm. The larger dimension of the samples increases the damping ratein Eqs. (30) and (31) and ensures accurate convergence of the working equation.
- A new furnace was used to uniformly heat the larger cylindrical samples with an axial temperature difference controlled below 1°C. This also minimizes buoyant forces which induce the second-order terms in the Navier–Stokes equation and causes the simplified liquid flow calculation model of Eq. (11) to be incorrect.
- The viscosities of aluminum alloys are quite low, so the decay of oscillation is much slower. In order to characterize the viscosity of these materials, more oscillation cycles are used to get the decrement of the oscillation. The measurement time increased from 100 to 400 s, which ensures the accuracy of the curve fit.

After improving the viscometer and adopting the working equation without a correction factor, the accuracy and repeatability are much improved for low viscosity, low-density aluminum alloy samples. The viscosities of pure aluminum and three aluminum alloys were measured in the present study. The samples were machined to 1 cm diameter and 12 cm length. The chemical compositions are shown in Table I. Three measurements were made at each temperature, and the average data are listed in Table II and Fig. 4. The logarithms of viscosities vs. $1/T$ for the measurement data are shown in Fig. 5. Empirical Arrhenius-type equations for the materials were determined to be:

$$\text{Pure Aluminum,} \quad \mu = 0.205\exp(1780\ \mathrm{K}^{-1}) \tag{32}$$

$$\text{A201} \quad \mu = 0.214\exp(1690\ \mathrm{K}^{-1}) \tag{33}$$

$$\text{A319} \quad \mu = 0.219\exp(1480\ \mathrm{K}^{-1}) \tag{34}$$

$$\text{A356} \quad \mu = 0.157\exp(1850\ \mathrm{K}^{-1}) \tag{35}$$

Table I. Chemical Compositions of Samples (wt%)

Sample	Ag.	Cu	Si	Fe	Mg	Mn.	Ti	AL
Pure AL	–	–	–	–	–	–	–	99.995
A201	0.59	4.7	< 0.05	0.05	0.28	0.31	0.21	Remainder
A319	–	3.01	6.1	0.68	0.3	0.71	–	Remainder
A356	–	–	6.9	0.08	0.34	–	0.013	Remainder

The liquidus temperatures of these alloys were determined by DSC, which ensured the measured viscosities are in the all liquid zones of the alloys. The liquid flows of alloys in mushy zones will become non-Newtonian. In that case, Eq. (9) is not satisfied and the viscosities of two-phase fluids cannot be obtained by the above method. Another technique is being investigated to measure viscosities of metals in the mushy zone. Densities of the molten alloys were characterized in a separate investigation.

Table II. Viscosity Measurements of Aluminum Samples (mPa · s)

Temperature (°C)	Pure Al	A201	A319	A356
678	1.345			
688			0.999	
690				1.045
706	1.284	1.214		
727		1.127		
733				0.987
735	1.183			
742			0.947	
755		1.106		
760	1.149			
765			0.929	
766		1.086		
783	1.095			
787		1.051		
788				0.924
789			0.903	
808		1.021		
815	1.049		0.857	
840			0.831	0.821
841	1.037			
866			0.811	

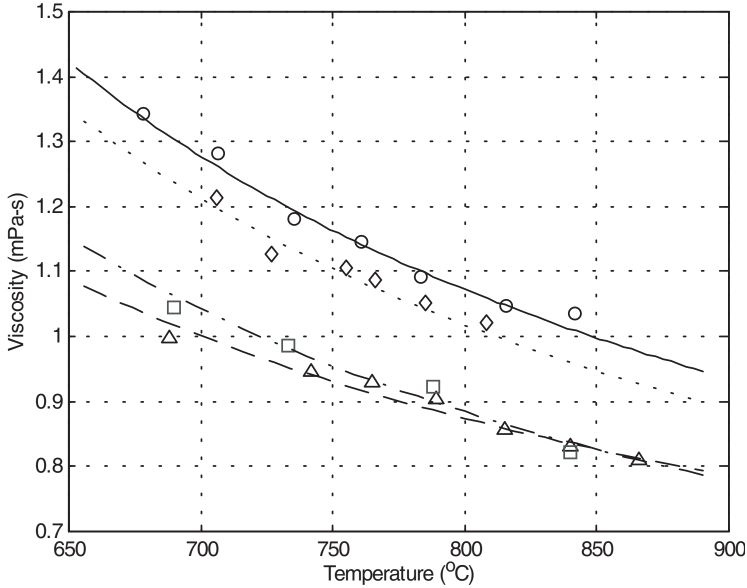

Fig. 4. Viscosities of the aluminum samples [○, pure aluminum, Eq. (32); ◇, A201, Eq. (33); △ A319, Eq. (34); □ A356, Eq. (35)].

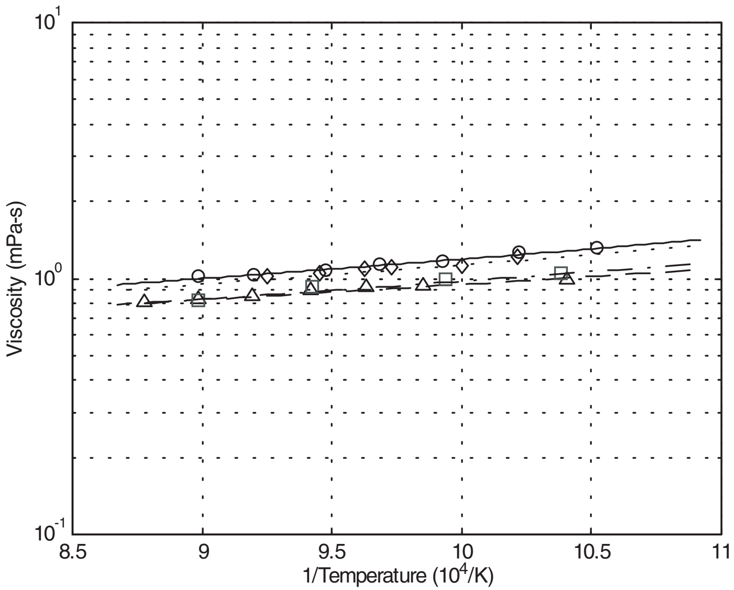

Fig. 5. Viscosities of the aluminum samples vs. reciprocal of absolute temperature [○, pure aluminum, Eq. (32); ◇ A201, Eq. (33); △ A319, Eq. (34); □ A356, Eq. (35)].

5. CONCLUSION

A working equation to calculate the viscosity of fluids from measured oscillating motion parameters without any correction factor was developed for an improved oscillating cup viscometer. The technique was utilized to measure viscosities of molten aluminum alloy samples with low densities and low viscosities. The experimental data are very repeatable. The study of the working equation shows it has very high mathematical accuracy, but special attention should be taken when obtaining oscillation parameters and other system design parameters.

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REFERENCES

1. W. Brockner, K. Torklep, and H. A. Oye, *Ber. Bunsenges Phys. Chem.* **83**:1 (1979).
2. P. Banerjee and R. A. Overfelt, *Int. J. Thermophys.* **20**:1791 (1999).
3. V. M. B. Nunes, F. J. V. Santos, and C. A. Nieto de Castro, *Int. J. Thermophys.* **19**:427 (1998).
4. R. A. Overfelt, C. A. Matlock, and M. E. Wells, *Metallurgical and Materials Transactions* **27B**:698 (1996).
5. B. Knapstad, P. A. Skjolsvik, and H. A. Oye, *Ber Bunsenges. Phys. Chem.* **94**:1156 (1990).
6. M. R. Hopkins and T. C. Toye, *Proc. Phys. Soc.* **B63**:773 (1950).
7. R. Roscoe, *Proc. Phys. Soc.* **72**:576 (1958).
8. R. Roscoe and Bainbridge, *Proc. Phys. Soc.* **72**:585 (1958).
9. J. Kestin and G. F. Newell, *Z. Angew. Math. Phys.* **8**:433 (1957).
10. D. A. Beckwith and G. F. Newell, *Z. Angew. Math. Phys.* **8**:450 (1957).
11. T. Ohta, O. Borgen, W. Brockner, D. Fremstad, K. Grjotheim, K. Torklep, and H. A. Oye, *Ber. Bunsenges Phys. Chem.* **79**:335 (1975).
12. K. Torklep and H. A. Oye, *J. Phys. E Sci. Instrum.* **12**:875 (1979).