A Simplified Phenomenological Theory of Viscosity for Liquid Metals

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This study simplifies the phenomenological theory of viscosity previously proposed by the present authors. This simplified theory has only two thermodynamic properties and one parameter, as opposed to its predecessor, which has many, allowing for easy calculation of the liquid viscosities. The viscosity of liquid metals, an excellent test for checking the validity of the liquid theory, can be calculated using the equation based on the simplified theory. The calculated results by the current theory turn out to be good for the liquid metals, including sodium, potassium, rubbidium, cesium, lead and mercury.

Keywords: Theory of viscosity, Viscosity of liquid, Liquid metals.

Introduction

The viscosities of liquid metals and alloys play an important role in liquid metal processing. Interest in the viscosity of liquid metals stems both from practical considerations, such as their use as atomic reactor coolants and philosophic considerations such as the fact that their structural simplicity makes them good media in which to test current theories of the liquid state. However, the high temperatures and critical contamination problems inherent in liquid metal viscometry have given rise to conflicting experimental results as well as to the anomalous viscosity dispute that has dotted the literature.

During the last 50 years various expressions for the viscosity of liquids have been presented. Representative examples are the expressions based on the statistical kinetic theory of Born and Green[1] and Kirkwood *et al.*,[2] the model theory of Frenkel,[3] Eyring[4] and Andrade,[5] the corresponding states theory of Helfand and Rice,[6] the hard sphere theory of Longuet-Higgins and Pople,[7] and so forth. Unfortunately, none of these expressions provides entirely satisfactory results when applied to liquid metals. A liquid metal is a severe test for any liquid theory to that extent. Low vapor pressures at melting, small differences in volume between liquid and solid, and rather large temperature ranges of the liquid state can lead to large differences between experimental and calculated values for thermodynamic properties and viscosities.

A phenomenological theory of viscosity that was proposed by the authors has been successfully applied to normal liquid,[8] water,[9] and helium,[10] which exhibits abnormal behavior compared to other ordinary liquids.

In the present paper, we apply the simplified expression of this theory to liquid metals adequately

Theory

According to the previous theory of viscosity,[8] the viscosities of fluids through which a phonon propagates with a velocity V_{ph} and a mean free path λ_{ph} can be given as follows:

$$\eta = (P_K + P_I)\, \lambda_{ph}/V_{ph}, \qquad (1)$$

where P_K and P_I are the kinetic pressure and internal pressure of fluids, respectively.

V_{ph} and λ_{ph} are expressed as follows:

$$V_{ph} = c_o = (\gamma/\rho\beta_{\mathrm{T}})^{1/2} \qquad (2)$$

$$\lambda_{ph} = 1/(\pi d^2 N_{ph}), \qquad (3)$$

where c_o, N_{ph}, ρ, β_{T}, d and γ, respectively, are the velocity of sound, the phonon number density, the densities of fluids, the isothermal compressibility, the collision diameter of phonon, and the heat capacity ratio, C_P/C_V. Therefore we have

$$\eta = (P_K + P_I)\, \frac{(\rho\beta_T/\gamma)^{1/2}}{\pi d^2 N_{ph}}. \qquad (4)$$

To calculate the viscosity, we have to know the thermodynamic properties, such as ρ, P, α_P, β_T, and γ. But, in general, all thermodynamic values of a fluid are not known. Therefore, we refined the above equation to reduce the data reqired .

The quantities on the right hand side of Eqn. (4) are intrinsic properties for the fluid in equilibrium state, therefore, we can simplify the above formula using the thermodynamic relation between them.

Eqn. (4) is reformulated as follows:

$$\eta = P_K\!\left(1 + \frac{P_I}{P_K}\right)(\lambda_{ph}/c_o) \qquad (5)$$

Of above equation, the ratio of P_K and c_o can be expressed as a simple form, using the following thermodynamic relationship.

$$\frac{C_P}{C_V} - 1 = \frac{TV\alpha_P^2}{C_V\beta_T} = \frac{MTc_o^2\alpha_P^2}{C_P} \qquad (6)$$

And the result is as follows:

$$\frac{P_K}{c_o} = \frac{(MT)^{1/2}}{V}[C_V(1-1/\gamma)]^{1/2} \qquad (7)$$

By introducing Eqn. (7) into Eqn. (5), we have

$$\eta_{fluid} = \frac{(MT)^{1/2}}{V}[C_V(1-1/\gamma)]^{1/2}\left(1+\frac{P_I}{P_K}\right)\lambda. \qquad (8)$$

In a liquid state where the intermolecular forces of neighbors are so high that the internal pressure is equal to the kinetic pressure, $P_I \doteqdot P_K$, the equation of liquid viscosity is simplified as follows:

$$\eta_{liquid} = 2\frac{(MT)^{1/2}}{V}[C_V(1-1/\gamma)]^{1/2}\lambda. \qquad (9)$$

Calculations and Results

If it is assumed that the number density of the phonon is proportional to the density of molecules in the fluid, it can be expressed as

$$N_{ph} = f(N/V), \qquad (10)$$

where the proportionality factor f is taken equal to the vacancy fraction in the fluid $(V-V_s)/V$, V_s being the fluid volume at closest packed state.

The collision diameter, d, can be calculated using the Sutherland method[11] which is given by

$$d = d_o(1+1.8T_b/T)^{1/2}, \qquad (11)$$

where T_b is the normal boiling temperature, and d_o is the values of d at infinite temperature. The parametric value d_o can be calculated from the close packed volume, V_S, and the structural information as follows:

$$d_o = (\alpha V_S/N)^{1/3}, \qquad (12)$$

where α is a proportionality constant that has the order of magnitude of unity and may be obtained readily for all types of regular spacings from the solid geometry of the structures. In this calculation, we take α as 0.44.

The parametric values of V_S for various fluids are listed in Table 1. The only parametric value of Vs is the one obtained by fitting the calculated viscosity to the experimental data.

The calculated viscosities of sodium, potassium, rubbidium, cesium, mercury and lead are listed in Table 2.

Discussion

In spite of the practical and theoretical importance, few theories have been developed for the viscosity of liquid metals. The viscosity equation (Eqn. 9) can be applied successfully to liquid metals, an exellent check of the validity of the

Table 1. Parametric Values used in Calculation

	Na	K	Rb	Cs	Pb	Hg
V_s (cm³/mol)	23.46	48.41	58.40	70.32	19.28	14.47

Table 2. Comparison of Calculated and Observed Viscosities of Liquid Metals

	T/°K	$\eta_{calc.}/\mu$-poise	$\eta_{obs.}/\mu$-poise
Na	376.7	6628	6860
	440.6	4205	5040
	523.0	3308	3810
	623.0	2844	2690
	973.0	2241	1820
K	342.9	5146	5150
	440.4	3265	3310
	523.0	2760	2680
	623.0	2473	1910
Rb	311.0	6789	6734
	320.9	6180	6258
	371.7	4643	4844
	413.5	4016	4133
	493.1	3383	3234
Cs	316.4	6285	6299
	371.6	4555	4753
	413.5	3972	4065
	441.0	3730	3760
	483.9	3462	3430
Pb	729.0	21378	20590
	842.0	17964	17000
	976.0	15995	13490
	1117.0	14831	11850
Hg	253.0	18379	18500
	273.0	17430	16800
	293.0	16879	15500
	373.0	15422	12700
	473.0	14446	10100

liquid theory. To calculate the liquid viscosities by using Eqn. (9), we need the data for C_p and C_v. Since we can not find the experimental values for C_v, we can use the value of C_v from the following simple equation by Walter and Eyring[12] for the liquid metals.

$$C_v = 6V_s/V + 3(V - V_s) \quad \text{cal mole}^{-1}\text{deg}^{-1} \qquad (13)$$

There may remain some arguments in using the approximations of Eqn. (10) and (11), since these equations are semi-empirical. Better approximations for N_{ph} and d_o may be available. However no adjustable parameters are involved in either equation. Although 1.8 is used in Eqn. (11), it changes slightly with the material of interest. But we chose 1.8 for the liquid metals. The calculated viscosities are in good agreement with the observed ones. If we could use the experimental values instead of the value of C_V from Eqn. (13), better calcuated results would be found. Some theories of viscosity have been applied to liquid metals, but none of these provides satisfactory results from an engineering point of view. The significant structure theory, which has been used widely for many liquids, shows large deviations for liquid metals.[13] Notwithstanding the simplicity of this equation, we hope it may be useful for engineering applications.

Conclusion

Of all the current theories of viscosity, the phenomenological theory of viscosity, previously proposed by the authors, can be applied most easily to many systems with acceptable results. But they could not be used for the calculation of the viscosity of liquid metals because of the many thermodynamic properties involved. The theory is simplified as in Eqn. (9) for liquids, using the thermodynamic relations, and therefore, this theory can be used for the calculation of the viscosity of liquid metals. No adjustable parameters are involved in this equation. Through this simple viscosity equation, **v**iscosities of liquid metals are calculated as easily as in normal liquids. The calculated and observed values are in good agreements.

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