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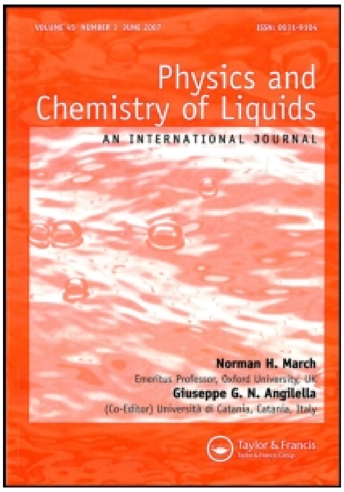

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Liquid Metal Transport Properties†

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Abstract—Viscosity and self-diffusivity data for ten liquid metals are correlated by use of the theory of corresponding states and the reducing factors defined by Helfand and Rice. The transport data are used to determine relative values of the pair-potential energy parameters, which are found to have a linear relationship with melting point. By using density data, it is possible to construct reduced correlations for diffusivity and viscosity as functions of reduced temperature without explicitly choosing distance parameters.

1. Introduction

Knowledge of the transport properties, viscosity and diffusivity, of liquid metals is important for a number of existing and potential applications, including the use of liquid metals as coolants and fuels in nuclear reactors. Fast breeder reactors, as well as small reactors designed to generate power aboard spacecraft, will probably have liquid metal coolants. Reactors that generate power by the fusion of deuterium and tritium will employ " blankets " of liquid lithium as a moderator and coolant and for tritium breeding.(1)

One objective of theoretical work on transport properties of liquid metals is the establishment of a sound basis for the extrapolation of existing data to higher temperatures and the development of techniques for estimating properties where data are lacking. Furthermore, theories that successfully correlate data are likely to afford insight into the nature of the liquid state.

In this paper, the theorem of corresponding states is used to develop correlations of the viscosity and diffusivity of liquid metals as functions of temperature. The reduced values of viscosity, diffusion, volume, and temperature as defined by Helfand and Rice(2) are used to construct the correlations.

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Probably the first use of the theorem of corresponding states to correlate the properties of liquid metals was Chapman's viscosity correlation.(3) Chapman showed that reduced viscosity (defined somewhat differently than by Helfand and Rice) could be correlated with reduced volume and temperature by the adjustment of the energy parameter of the interatomic potential energy function. The distance parameter was assumed to be the interatomic distance in the close-packed crystal at 0 °K (Goldschmidt diameter). The energy parameters determined from the viscosity data were then found to vary linearly with melting temperature. Thus, they can be estimated for those liquid metals whose viscosities have not been measured. Chapman(4) also used the theorem of corresponding states to correlate the heat capacities of several low-melting-point liquid metals. The same energy and distance parameters, ϵ/k and σ, were used as in the viscosity correlation. These parameters were also used by the present writer(5) to correlate isothermal compressibility data for the liquid alkali metals, cesium, rubidium, potassium, and sodium. From this correlation, the isothermal compressibility versus temperature curve for liquid lithium was predicted. The predicted values are in good agreement with data reported a year later by Novikov *et al.*(6)

The transport properties, viscosity, self-diffusivity, and thermal conductivity of a number of cryogenic liquids have been correlated with temperature (along the saturation curve) by Preston, Chapman, and Prausnitz,(7) using the reduced quantities as defined by Helfand and Rice. The data for viscosity and thermal conductivity were used to determine, first, the relative values of the energy parameters for the cryogenic liquids, and second, the relative values of the distance parameters. The parameters thus found agree approximately with the energy and distance parameters for the Lennard–Jones potential determined from gas-phase viscosity data. Preston *et al.* showed that, with these parameters, reduced correlations of viscosity, self-diffusivity, and thermal conductivity with temperature can be constructed.

The procedure adopted here for determining the pair-potential energy parameters is similar to that of Preston *et al.*; by combining the data for viscosity, self-diffusivity, and density for ten liquid metals, the relative values of the energy parameters are determined without making any assumptions about the distance parameter. However, instead of correlating only reduced self-diffusivity with temperature, the correlations employ these reduced transport properties multiplied by the reduced volume raised to the power required to cancel the distance parameter from the resulting product. In this way the choice of a distance parameter can be avoided altogether.

2. Determination of the Energy Parameters

Helfand and Rice⁽²⁾ have shown that by assuming an intermolecular potential of the form suggested by Pitzer:⁽⁸⁾

$$u = \epsilon u^*(r/\sigma), \tag{1}$$

where ϵ and σ are characteristic energy and distance constants and u^* is a universal function of r/σ, it is possible to derive a law of corresponding states for the transport properties. The reduced transport properties are

$$\text{Reduced viscosity,} \quad \eta^* = \frac{\eta\sigma^2}{m^{1/2}\epsilon^{1/2}} \tag{2}$$

and

$$\text{Reduced diffusivity,} \quad D^* = \frac{Dm^{1/2}}{\epsilon^{1/2}\sigma}. \tag{3}$$

The reduced state variables are

$$\text{Reduced temperature,} \quad T^* = \frac{Tk}{\epsilon}, \tag{4}$$

$$\text{Reduced volume,} \quad V^* = \frac{V}{N\sigma^3}, \tag{5}$$

and

$$\text{Reduced pressure,} \quad P^* = \frac{P\sigma^3}{\epsilon}. \tag{6}$$

Neglecting quantum mechanical effects, universal functions relating the reduced transport properties with T^* and P^* are

$$\eta^* = \eta^*(T^*, P^*) \tag{7}$$

and

$$D^* = D^*(T^*, P^*). \tag{8}$$

TABLE 1 Sources of Density and Transport Data for Ten Liquid Metals

Metal	Viscosity	Diffusivity	Density
Mercury	S. Erk, *Z. Physik*, **47**, 886 (1928). Ch. Chaililov, *Zh. Tekhn. Fiz.* **8**, 1249 (1938).	E. F. Broome and H. A. Walls, *TMS-AIME* **242**, 2177 (1968).	A. V. Grosse, *J. Phys. Chem.* **68**, 3419 (1964).
Gallium	K. E. Spells, *Proc. Phys. Soc.* **48**, 299 (1936).	E. F. Broome and H. A. Walls, *TMS-AIME* **245**, 739 (1969).	W. H. Hoather, *Proc. Phys. Soc.* **48**, 699 (1936).
Sodium	C. T. Ewing *et al.*, *J. Phys. Chem.* **58**, 1086 (1954). N. A. Kalakutskaya, *High Temp.* **6**, 436 (1968).	M. W. Ozelton and R. A. Swalin, *Phil. Mag.* **18**, 441 (1968). R. E. Meyer and N. H. Nachtrieb, *J. Chem. Phys.* **23**, 1851 (1955).	C. T. Ewing *et al.*, *J. Phys. Chem.* **58**, 1086 (1954). N. A. Kalakutskaya, *High Temp.* **6**, 436 (1968).
	C. T. Ewing *et al.*, *J. Am. Chem. Soc.* **73**, 1168 (1951).		E. I. Gol’tsova, *High Temp.* **4**, 348 (1965).
Indium	V. A. Lodding, *Z. für Natur.* **11A**, 200 (1956).	A. F. Crawley and H. R. Thresh, *TMS-AIME* **245**, 424 (1969).	P. J. McGonigal *et al.*, *J. Inorg. Nucl. Chem.* **24**, 1012 (1962).
Lithium-7	N. T. Ban *et al.*, *Phys. Rev.* **128**, 6 (1962).	J. S. Murday and R. M. Cotts, *J. Chem. Phys.* **48**, 4938 (1968).	E. I. Gol’tsova, *High Temp.* **4**, 348 (1965).
Tin	E. Gebhardt, *Z. Metallk.* **46**, 669 (1955).	G. Careri *et al.*, *Nuovo Cimento* **10**, 1088 (1958).	A. D. Kirshenbaum and J. A. Cahill, *Trans. ASM* **55**, 844 (1962).
	F. A. Kanda and R. P. Colburn, *Phys. and Chem. of Liquids* **1**, 159 (1968).		
Lead	D. Ofte and L. J. Wittenberg, *TMS-AIME* **227**, 706 (1963).	S. J. Rothman and L. D. Hall, *J. Metals* **8**, 199 (1956).	E. Gebhardt, *Z. Metallk.* **46**, 90 (1955).
Zinc	E. Gebhardt and K. Detering, *Z. Metallk.* **50**, 379 (1959).	N. H. Nachtrieb *et al.*, *J. Phys. Chem.* **67**, 2353 (1963).	E. Gebhardt, *Z. Metallk.* **50**, 379 (1959).

TABLE 1—*continued*

Metal	Viscosity	Diffusivity	Density
Silver	E. Gebhardt and G. Worwag, *Z. Metallk.* **42**, 358 (1951).	L. Yang *et al.*, *TMS-AIME* **230**, 426 (1964). V. G. Leak and R. A. Swalin, *TMS-AIME* **230**, 426 (1964).	E. Gebhardt, *Z. Metallk.* **42**, 358 (1951).
Copper	G. Cavalier, " Measurement of the Viscosity of Under-cooled Molten Metals," in *Physical Chemistry of Metallic Solutions and Intermetallic Compounds*, National Physical Laboratory, Symposium No. 9 (London, 1958), Vol. II, paper 4D.	J. Henderson and L. Yang, *TMS-AIME* **221**, 72 (1961).	G. Urbain and L. D. Lucas, " Densities of Molten Silver, Copper, and Iron," in *Physical Chemistry of Metallic Solutions and Intermetallic Compounds*, National Physical Laboratory, Symposium No. 9 (London, 1958), Vol. II, paper 4E.

The reduced equation of state is

$$V^* = V^*(T^*, P^*). \tag{9}$$

In general, the transport properties and the densities of liquid metals show much greater dependence on temperature than on pressure. Therefore, we assume that pressure can be neglected in Eqs. (7), (8), and (9). Since η^*, D^*, and V^* are functions of T^*, their combinations are also functions of T^*. Therefore,

$$\frac{\eta^* V^*}{D^*} = f(T^*), \tag{10}$$

and, by substitution of the defining Eqs. (2), (3), and (5),

$$\frac{\eta^* V^*}{D^*} = \frac{\eta V}{DmN} = \frac{\eta V}{DM} = \frac{\eta}{D\rho} \tag{11}$$

$$= \frac{\nu}{D} = \text{ratio of kinematic viscosity to diffusivity.}$$

The data sources for the density, viscosity, and self-diffusivity for ten liquid metals are given in Table 1. Figure 1 is a plot of the ratio of kinematic viscosity to self-diffusivity as a function of log $1/T$ for these liquid metals. (Smoothed values of the data have been used in constructing Fig. 1.) The family of approximately parallel curves shown in Fig. 1 can be superimposed by choosing values of ϵ/k that have the correct relative magnitudes and plotting ν/D as a function of $1/T^* = \epsilon/kT$. This demonstrates the validity of Eq. (10).

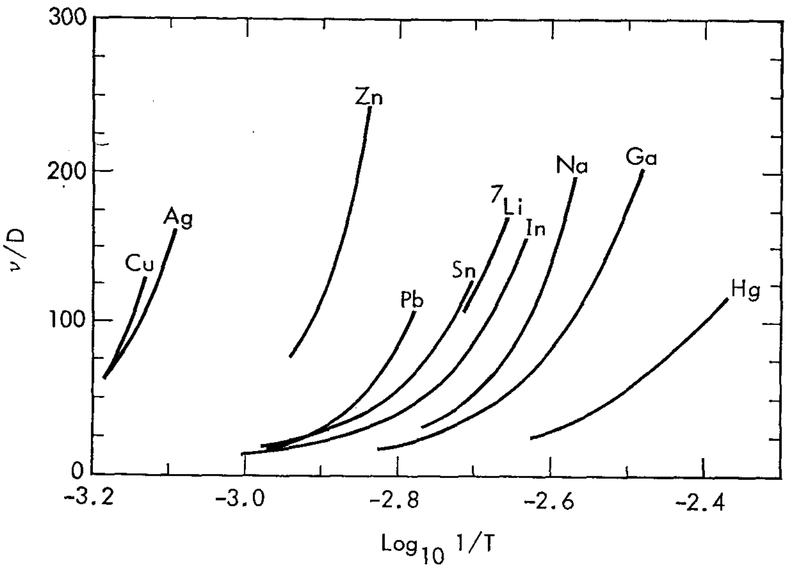

Figure 1. Ratio of kinematic viscosity, ν, to self-diffusivity, D, for 10 liquid metals.

All other values of ϵ/k are relative to the value of ϵ/k for sodium. Table 2 lists the values of ϵ/k found by superimposing the curves of Fig. 1. As found by Chapman, the resulting ϵ/k values bear an approximate linear relationship to the melting point: $\epsilon/k = 5.2\,T_m$.

A value of $\epsilon/k = 1970$ °K is assigned to sodium. This is the effective Lennard–Jones parameter for sodium as determined by Ling(9) from experimental X-ray scattering data. This is the same value used by Chapman.(3,4)

TABLE 2 Energy Parameters

Metal	Melting point (°K)	Ref. 3 ϵ/k(°K)	Ref. 3 $\frac{\epsilon/k}{T_m}$	This Work ϵ/k(°K)	This Work $\frac{\epsilon/k}{T_m}$
Hg	234.3	1250	5.33	1200	5.12
Ga	303.0	—	—	1730	5.71
Na	371.0	1970	5.31	1970	5.31
In	429.3	2500	5.82	2230	5.19
^{7}Li	453.7	2350	5.18	2380	5.25
Sn	505.0	2650	5.25	2510	4.97
Pb	600.0	2800	4.66	2750	4.57
Zn	692.7	4700	6.78	3770	5.44
Ag	1234.0	6400	5.19	6250	5.07
Cu	1356.0	6600	4.87	6440	4.75

3. Correlations for Viscosity and Diffusivity

In order to directly apply the Helfand and Rice parameters to the correlation of viscosity and diffusivity, relative values of σ, the distance parameter, would be required. By multiplying η^* and D^* by V^* raised to the power required to cancel σ from the resulting expression, the selection of explicit values for σ can be avoided. The dimensionless expressions to be correlated with T^* are thus

$$\text{for viscosity: } \eta^*(V^*)^{2/3} = \frac{N^{1/3}\eta V^{2/3}}{(MR\epsilon/k)^{1/2}}; \tag{12}$$

$$\text{for diffusivity: } D^*/(V^*)^{1/3} = \frac{N^{1/3}M^{1/2}D}{(R\epsilon/k)^{1/2}V^{1/3}}. \tag{13}$$

In Figs. 2 and 3 the correlations for viscosity and diffusivity are plotted as indicated by Eqs. (12) and (13). The resulting graphs appear to correlate the data well. The correlation for self-diffusivity has a spread of ±20% if the high-temperature values for lead are neglected.

4. Use of the Correlations

The use of the correlations in Figs. 2 and 3 can be illustrated by calculating the viscosity and self-diffusivity of liquid iron. The viscosity of iron has been measured but the diffusivity has not.

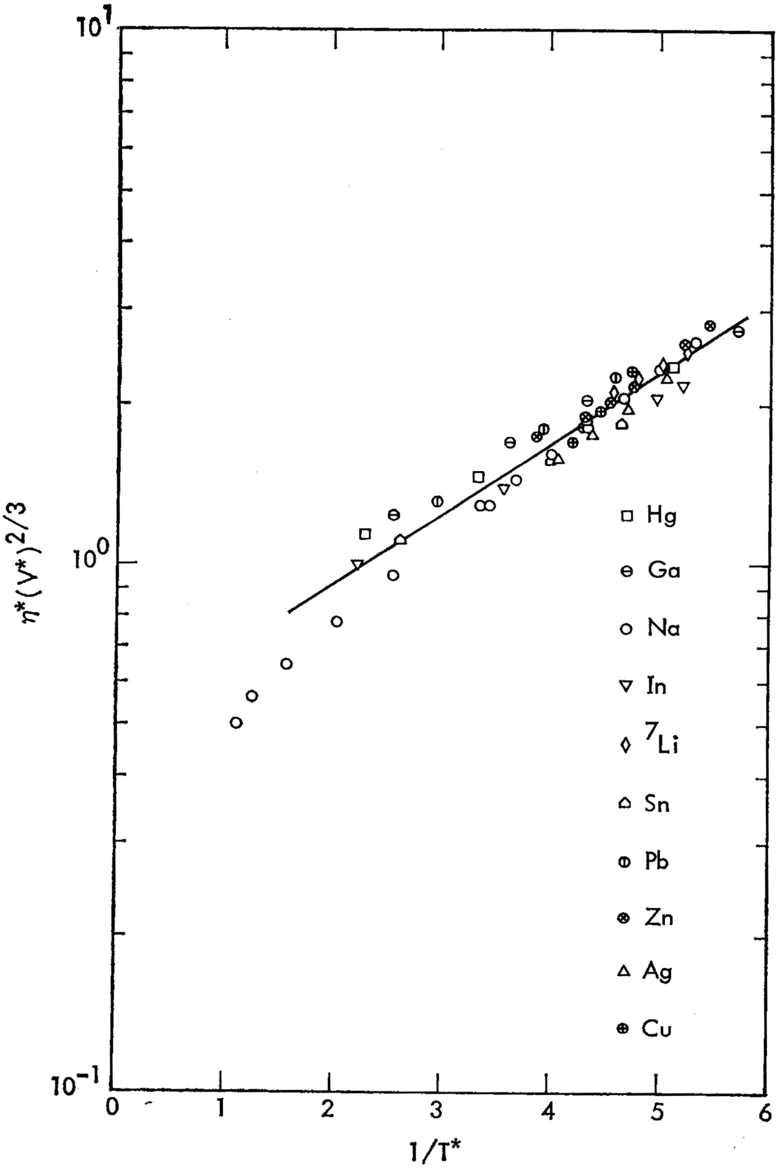

Figure 2. Viscosity correlation.

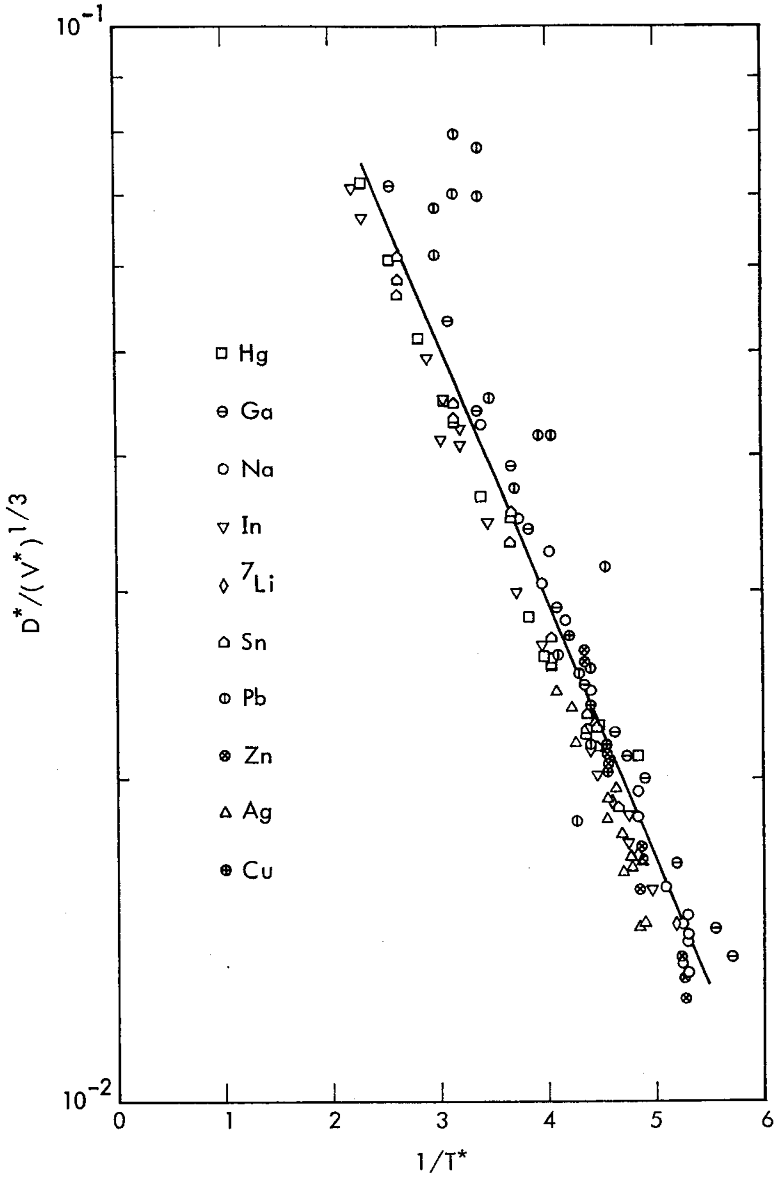

Figure 3. Self-diffusivity correlation.

According to Eq. (12), the viscosity, η, can be expressed as:

$$\eta = \eta^*(V^*)^{2/3}\frac{(MR\epsilon/k)^{1/2}}{N^{1/3}}\cdot\frac{1}{V^{2/3}}. \tag{14}$$

Similarly, by rewriting Eq. (13), we have for the diffusivity, D:

$$D = D^*/(V^*)^{1/3}\frac{(R\epsilon/k)^{1/2}}{N^{1/3}M^{1/2}}V^{1/3}. \tag{15}$$

In Eqs. (14) and (15), the groups $\eta^*(V^*)^{2/3}$ and $D^*/(V^*)^{1/3}$ are given by the correlations of Figs. 2 and 3, respectively, as a function of reduced temperature, T^*.

The energy parameter, ϵ/k, for iron is estimated from the melting temperature of iron, 1809 °K: $\epsilon/k = 5.2T_m = 5.2(1809\ °\text{K}) = 9400\ °\text{K}$. With this value of ϵ/k, the absolute temperature can be converted to reduced temperature,

$$T^* = \frac{T}{\epsilon/k}. \tag{16}$$

Values of the viscosity have been calculated using Eq. (14) and Fig. 2. Values for the density of liquid iron are from Lucas.$^{(10)}$ In Fig. 4 the calculated values are compared with Cavalier's$^{(11)}$ data and with the calculated values of Grosse.$^{(12)}$ Agreement with the

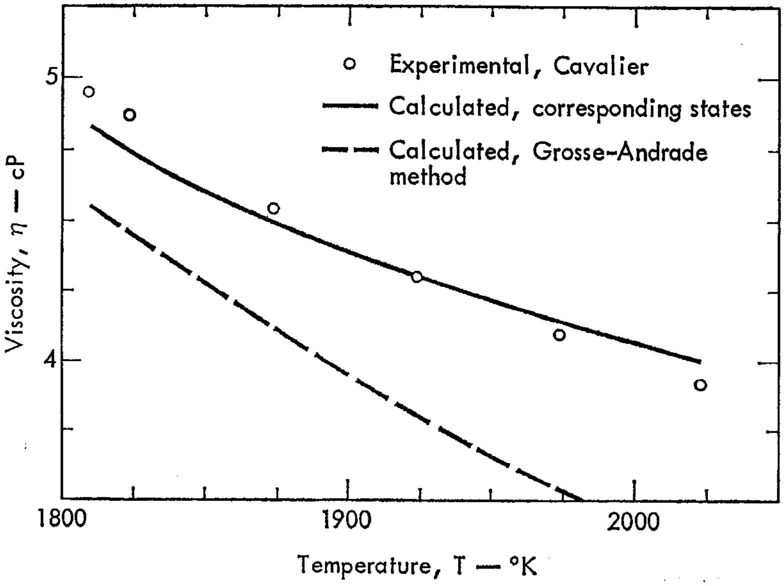

Figure 4. Viscosity of liquid iron.

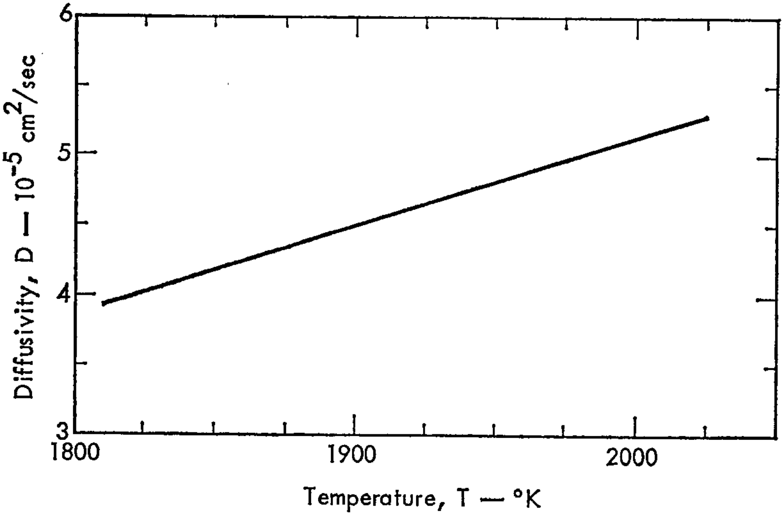

Figure 5. Calculated values for self-diffusivity of liquid iron.

data is good. Since the corresponding-states method produced values of viscosity which are in good agreement with the data, one has some confidence in using the same value of ϵ/k, 9400 °K, to predict the self-diffusivity of liquid iron. This can be done with Eq. (15), Fig. 3, and Lucas's density data. The calculated values are given in Fig. 5 over the temperature range from the melting point to 2025 °K.

5. Conclusions

The corresponding-states theory, employing the simple reduced transport coefficients defined by Helfand and Rice, has been used to correlate viscosity and self-diffusivity data for the ten liquid metals for which both measurements have been reported. The correlation of the transport properties of these metals can be used to extrapolate existing data and to estimate the transport properties of those metals for which no data exist. Despite the use of a different expression for the reduced viscosity, the values of ϵ/k found here have the same linear relationship to melting point as the ϵ/k values determined by Chapman in his analysis of viscosity data for the liquid metals. By introducing reduced volume into the correlation, it is possible to avoid an explicit choice of the distance parameter.

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Notation

Italic Letters

D	Diffusivity, cm^2/sec
k	Boltzmann's constant, 1.380×10^{-16} erg/°K
M	Atomic weight, g/mole
m	Mass of an atom, g/atom
N	Avogadro's number, 6.024×10^{23} molecules/mole
P	Pressure, dynes/cm^2
r	Interatomic distance, Å
T	Temperature, °K
T_m	Melting point, °K
u	Intermolecular pair potential, ergs
V	Molecular volume, cm^3/mole

Greek Letters

ϵ	Pair-potential energy parameter, ergs
ϵ/k	Pair-potential energy parameter, °K
η	Viscosity, g/cm-sec
ν	Kinematic viscosity, cm^2/sec
ρ	Density, g/cm^3
σ	Pair potential distance parameter, Å

Reduced Quantities

D^*	Reduced diffusivity $= \dfrac{Dm^{1/2}}{\epsilon^{1/2}\sigma}$
P^*	Reduced pressure $= \dfrac{P\sigma^3}{\epsilon}$
V^*	Reduced volume $= \dfrac{V}{N\sigma^3}$
T^*	Reduced temperature $= \dfrac{Tk}{\epsilon}$
η^*	Reduced viscosity $= \dfrac{\eta\sigma^2}{m^{1/2}\epsilon^{1/2}}$

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