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SOLUTE DIFFUSION
IN LIQUID METALS

by B. N. Bhat

George C. Marshall Space Flight Center

Marshall Space Flight Center, Ala. 35812

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16. ABSTRACT A gas model of diffusion in liquid metals is presented. In this model, ions of liquid metals are assumed to behave like the molecules in a dense gas. Diffusion coefficient of solute is discussed with reference to its mass, ionic size, and pair potential. The model is applied to the case of solute diffusion in liquid silver. An attempt has been made to predict diffusion coefficients of solutes with reasonable accuracy.			
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TABLE OF CONTENTS

	Page
INTRODUCTION	1
THE GAS MODEL OF DIFFUSION	1
EXPERIMENTAL EVIDENCE	4
Thermotransport	4
Clusters in Liquid Metals	4
Temperature Dependence of Diffusion Coefficients.	4
Solute Diffusion	4
SOLUTE DIFFUSION IN LIQUID SILVER	4
CONCLUSIONS	9
REFERENCES.	11

SOLUTE DIFFUSION IN LIQUID METALS

INTRODUCTION

While the mechanism of diffusion in solid metals seems to have been well understood, the process of diffusion in liquid metals has been puzzling. There is no generally accepted mechanism of diffusion in liquid metals. Several models of diffusion have been proposed, namely, fluctuation model [1,2], free volume model [3,4], and hole theory [5]. These models have been reviewed by Nachtrieb [6,7]. All the models have some degree of success in predicting the diffusion coefficients of liquid metals. Yet the disparity between theory and experiments is large in many cases. A similar situation exists in the case of solute diffusion in liquid metals, which has been reviewed by Gupta [8]. A later work by Ejima, Ingaki, and Kameda [9] also shows there is no satisfactory agreement between the theories and experiments in the case of diffusion of sulfur, selenium and tellurium in liquid silver.

Inadequacy of the current models is further exemplified by the results of thermotransport experiment [10,11], where mass transport occurs because of a temperature gradient. This is a second-order effect and hence provides a sensitive test of the validity of different models of diffusion. It turns out that none of the current models explains the results of thermotransport experiments. In fact, the hole theory predicts values for the heat of transport which are in direct contradiction to the experimental observations.

Hence, there is a lot to be desired about our understanding of diffusion in liquid metals. It does not seem to be a matter of refining the

current theories. But it calls for a basic change in the approach to diffusion in liquid metals. We need a theory which would explain the observed experimental results. In view of the partial success of the current theories, the new model would combine the features of the previous models. The gas model of diffusion, which is the subject of this paper, seems to satisfy the above requirements.

THE GAS MODEL OF DIFFUSION

The gas model assumes that the ions in a liquid metal act like the molecules in a dense gas. Theory of diffusion in gases has been well developed [12,13], and hence it can be used to describe the process of diffusion in liquid metals. There are some difficulties, however. In the kinetic theory of dilute gases it is assumed that there are only two-body collisions and that the molecular diameter is small as compared to the average distance between the molecules. Both these assumptions are valid in dilute gases, but in dense gases, these assumptions must be reconsidered. Enskog was the first to make advance in this direction by developing a kinetic theory of dense gases made up of rigid spherical molecules. For this special model there are no three-body or higher-order collisions. By considering only two-body collisions and by taking into account the finite size of the molecules, he was able to graft a theory of dense gases into the dilute gas theory developed earlier. He showed that the frequency of collisions in a gas made up of rigid spheres differs by a factor χ from that in a gas made up of point particles. The diffusion coefficient (D_{12}) in a dense binary gas mixture is given by [13].

$$(D_{12})_{\text{dense}} = \frac{(D_{12})_{\text{dilute}}}{\chi} \quad (1)$$

where

$$\chi \cong 1 + \frac{2}{3} \pi n_1 \sigma_1^3 \left(\frac{\sigma_1 + 4\sigma_2}{4\sigma_1 + 4\sigma_2} \right) + \frac{2}{3} \pi n_2 \sigma_2^3 \left(\frac{\sigma_2 + 4\sigma_1}{4\sigma_1 + 4\sigma_2} \right)$$

n_1, n_2 = number density of components 1 and 2 respectively,

and

σ_1, σ_2 = molecular diameter of components 1 and 2 respectively

There is little variation in the value of χ for dilute binary systems, and hence the expression for $[D_{12}]_{\text{dilute}}$ will be sufficiently accurate for the present discussion.

The diffusion coefficient in a dilute binary gas mixture is given by [13]

$$D_{12} = \frac{0.002628 \sqrt{T^3 [m_1 + m_2] / 2m_1 m_2}}{p \sigma_{12}^2 \Omega_{12}^{(1,1)*}(T_{12}^*)} \quad (2)$$

where

D_{12} = diffusion coefficient, in cm^2/sec ,

p = pressures in atmospheres,

T = temperature in degrees K,

$$T_{12} = kT/\epsilon_{12}$$

$\sigma_{12}, \epsilon_{12}/k$ = molecular potential energy parameters of 1-2 interaction in angstroms and degrees Kelvin respectively,

m_1, m_2 = molecular masses of components 1 and 2 respectively,

$\Omega_{12}^{(1,1)*}(T_{12}^*)$ = collision integral with the physical significance that it indicates the deviation of the model from the idealized rigid sphere model,

and

$$\sigma_{12} = \frac{\sigma_1 + \sigma_2}{2} = \text{for a rigid sphere model}$$

where σ_1, σ_2 are the ionic diameters of components 1 and 2 respectively.

When equation (2) is written for a single component, the expression for self-diffusion coefficient is obtained

$$D_{\text{self}} = \frac{0.002628 \sqrt{T^3/m}}{p \sigma^2 \Omega^{(1,1)*}(T^*)} \quad (2a)$$

where m and σ refer to the mass and size of the diffusing species.

For a liquid metal the pressure p is not the same as the pressure of a gas. One must consider the internal pressure which is equivalent to the cohesive energy of the liquid metal [14]. An accurate value of p cannot be determined at the present time. However, the value of p would be nearly the same when one considers the diffusivity of different solutes in a single solvent. The value of the collision integral $\Omega_{12}^{(1,1)*}$ is unity for rigid spheres. For a

normal liquid metal, its value is greater than unity when T_{12}^* is unity or less. The value of $\Omega_{12}^{(1,1)*}(T_{12}^*)$ decreases with increasing temperature, but, slowly. (Refer to the tables in Reference 13).

From the expression (2) it is evident that the diffusion coefficient varies at $T^{3/2}$. If D_{12} is plotted against $T^{3/2}$, one would obtain a straight line if $\Omega_{12}^{(1,1)*}$ were constant. Since $\Omega_{12}^{(1,1)*}(T_{12}^*)$ decreases with temperature, one would obtain a curved line which is concave upwards. The degree of concavity will depend on the values of $\Omega_{12}^{(1,1)*}$, which in turn depends on the type of pair potential. Conversely, one could determine the pair potential between the atoms from a plot of D_{12} versus $T^{3/2}$.

Expression (2) shows the variables in solute diffusion, namely, atomic mass and ionic size. It predicts that the diffusion coefficient varies inversely as the square root of the reduced mass, and square of the effective diameter σ_{12} . If the pair potentials are nearly the same for a set of solute elements in a given solvent, the relative diffusion coefficients of these solutes can be determined in a simple manner by use of expression (2).

Dense gas approach to diffusion in liquid metals is not totally new. Ascarelli and Paskin [15] have used a dense gas formulation to calculate the self-diffusion coefficient of mercury, indium and tin. Their theory employs van der Waals' concept of a dense fluid in which the attractive potential energy is thought of as a uniform negative potential. This is justified for liquid metals on the basis of pseudopotential theory according to which the attractive part of the interaction energy between electronically screened ions is

comparable to that found in rare gas liquids [16]. In their free volume model of diffusion Cohen and Turnbull do maintain that the diffusing atom does indeed move with gas kinetic velocity [3]. The fluctuation in density as envisaged by Swalin [1] is quite conceivable in a dense gas. This view is also supported by computer simulation of dense gases by Rahman [17] and Paskin and Rahman [18].

The above efforts have treated the liquid metals as essentially homogeneous. There are other efforts that consider a liquid metal may be heterogeneous. In his theory of significant liquid structures [19] Eyring assumes that a liquid metal consists of an intermixing of crystalline and gaseous fractions. More recently McLachlan has presented some evidence about the gaseous fraction in liquid metals [20].

The idea that a liquid may be heterogeneous comes naturally from the theory of Larson [21,14]. According to his theory a liquid metal is an aggregate of solid-like, liquid-like and vapor-like atoms. Their proportion is determined by the relative probability of occurrence of these atoms. Above the melting point the probability of a liquid-like atom is more than that of a solid-like atom. The probability of solid-like atoms decreases with increasing temperature. Hence, there will be some solid (less than 50 percent) in a molten metal and its percentage decreases with increasing temperature. A liquid atom is the one which is gas-like in one (or two) dimension and solid-like in other two (or one) dimensions. Since the diffusion coefficient of solid-like atoms is much smaller than that of gas-like atoms, the observed diffusion coefficient will correspond to that of gas-like atoms and its value will lie in between the values of diffusion coefficients in solid and gas phase.

EXPERIMENTAL EVIDENCE

There are many experimental results that seem to support the gas model. They are described in this section.

Thermotransport [10,11]

In a thermotransport experiment where a binary alloy is held in a temperature gradient for a sufficiently long time, segregation is generally observed. This segregation can be described by a quantity called the heat of transport, which is sensitive to the mechanism of diffusion. The previous models have failed to explain the heat of transport either in magnitude or in sign, whereas the gas model explains it satisfactorily. Hence it may be deemed that the gas model is the most satisfactory model of diffusion in liquid metals.

Clusters in Liquid Metals [22]

Clusters have been observed in liquid metals by Egelstaff in neutron diffraction studies. It has also been suggested that diffusion could occur through rotation and translation of clusters. According to Larson's theory (which supports the gas model) clusters of solid can exist in liquid metals near the melting temperature. The theory further suggests that the number and/or size of clusters will decrease with increasing temperature. In the opinion of this writer, the contribution of the clusters to the overall diffusion coefficient will be small, since the cooperative movement of several atoms is a much less likely event as compared with a binary collision.

Temperature Dependence of Diffusion Coefficients

Diffusion coefficients of solutes are generally reported in the form

$$D = D_0 \exp(-Q/RT) \quad (3)$$

where Q is the apparent activation energy. If the variation of p and $\Omega_{12}^{(1,1)*}(T_{12}^*)$ with the temperature are neglected, one obtains a value for the apparent activation energy from equation (2),

$$Q = -R \frac{d \ln D}{d \frac{1}{T}} = \frac{3}{2} RT \quad (4)$$

which is a function of temperature. Hence a plot of $\ln D$ versus $1/T$ will yield a curved line that is concave upwards. This has been observed in several metals [23].

Solute Diffusion

Experimental results of solute diffusion studies in liquid metals appear to be in good agreement with the gas model of diffusion. This is discussed in the following section, for the case of solute diffusion in liquid silver.

SOLUTE DIFFUSION IN LIQUID SILVER

Diffusion of solute in liquid silver has been studied in detail. The current models of

diffusion explain the diffusion behavior of indium, tin, antimony, and gold in silver with some success[8] by taking into account the valence of the solute. This approach fails when it comes to transition element solutes (iron, nickel, cobalt and ruthenium) or solutes in group VIA (sulfur, selenium and tellurium). The gas model, however, works satisfactorily for all the solutes Table 1 lists the pertinent data on the solutes along with the references.

Figure 1 shows the diffusion coefficients of solutes in liquid silver plotted against $T^{3/2}$. Many lines appear to be linear and some are curved and are concave upwards. With the exception of sulfur (which is a nonmetal) they are all grouped together. There is no particular trend noticeable (with respect to either valence or atomic mass or ionic size). The same curves are replotted in Figure 2 after dividing the value of the diffusion coefficient D_{12} by the factor $\sqrt{(m_1 + m_2)/m_1 m_2 / \sigma_{12}^2}$, which takes into account the mass and the ionic size of the solute element. The modified diffusion coefficient is denoted by D'_{12}

$$D'_{12} = 0.002628/p\Omega_{12}^{(1,1)*}(T_{12}^*) \quad (5)$$

In Figure 2, the curves are observed to separate into three distinct groups

1 Those with the lowest D'_{12} values are the transition elements; Fe, Ni, Co and Ru.

2. Those with intermediate D'_{12} values are the elements indium, tin, antimony, which are to the right of silver in the periodic table; gold, which is immediately below silver; and zinc, which is adjacent to copper, lying immediately above silver in the periodic table. All these solutes have similar inner ionic core,

and their diffusion coefficients (D'_{12}) are comparable to D'_{11} , the self-diffusion coefficient of liquid silver

3 Those with the highest values for D'_{12} are the elements sulfur, selenium, and tellurium, which belong to the group VIA.

The value of D'_{12} depends on the values of pressure p and collision integral $\Omega_{12}^{(1,1)*}(T_{12}^*)$, which depend upon the 1-2 pair potential. Figure 2 is thus largely a manifestation of the difference in the pair potential for solute elements in liquid silver. The diffusion coefficients of sulfur, selenium, and tellurium are widely separated in Figure 1, but they are much closer together in Figure 2. This will be the case if sulphur, selenium, and tellurium have similar pair potentials in silver. This is to be expected since all these elements belong to the group VIA of the periodic table, have a valence of two, and accept electrons from the conduction band. There are small variations in the values of D'_{12} within each group, but these differences are much smaller than the differences in the values of D'_{12} between the groups. The variation in the slope of the D'_{12} versus $T^{3/2}$ plot is a measure of the temperature dependence of $\Omega_{12}^{(1,1)*}(T_{12}^*)$, which is different for each solute.

Similar considerations apply to the solutes in the second group of Figure 2, namely, gold, indium, tin, antimony, and zinc. These solutes have similar ionic cores. Gold and zinc are the extremes within this group. This may be due to the fact that they belong to different periods in the periodic table of elements. Indium, tin, and antimony are in the same row as silver and hence their D'_{12} values may be expected to be even closer. Figure 2 shows that this is indeed the case. Interestingly, the slope of the

TABLE 1. PERTINENT DATA ON THE SOLUTES

Group	Element	Atomic Mass	Ionic Radius (Å)	Diffusion Coefficient in Liquid Ag at 961°C (1234°K) (D_{12}) cm ² /sec	Reference
1	S	32.1	1.84	5.50×10^{-5}	9
	Se	79.0	1.91	2.84	9
	Te	127.6	2.11	2.32	9
2	Au	197.0	1.37	2.27	24
	Ag	107.9	1.26	2.55	25
	In	114.8	0.81	3.65	26
	Sn	118.7	0.71	3.75	26
	Sb	121.8	0.62	4.30	26
	Zn	65.4	0.74	3.90	27
3	Fe	55.8	0.74	2.35	27
	Co	58.9	0.72	2.38	27
	Ni	58.7	0.69	2.69	27
	Ru	101.7	0.67	2.27	27

curve D'_{12} versus $T^{3/2}$ falls from Ag→In→Sn→Sb, in that order, suggesting a valence dependence of $\Omega_{12}^{(1,1)*}$

The transition elements, which form the third group, have the lowest values of D'_{12} . It is a common experience for transition metals to behave differently from other metals and here they have the lowest diffusion coefficients (D'_{12}). This suggests that the values of p and $\Omega_{12}^{(1,1)*}(T_{12}^*)$ (or either one of them) for transition metals are higher than those for other solutes.

It is clear that one needs to have accurate knowledge of the pair potentials to evaluate the collision integral $\Omega_{12}^{(1,1)*}$. At the present time pair potentials are being evaluated for pure metals, and it may be several years before the pair potentials are available for alloys. In the meantime, one can obtain a reasonable estimate of diffusion coefficient of a solute element which belongs to a particular group as envisaged in the previous paragraphs. Since the average of D'_{12} is known for that group, the value of D_{12} can be calculated from the expression

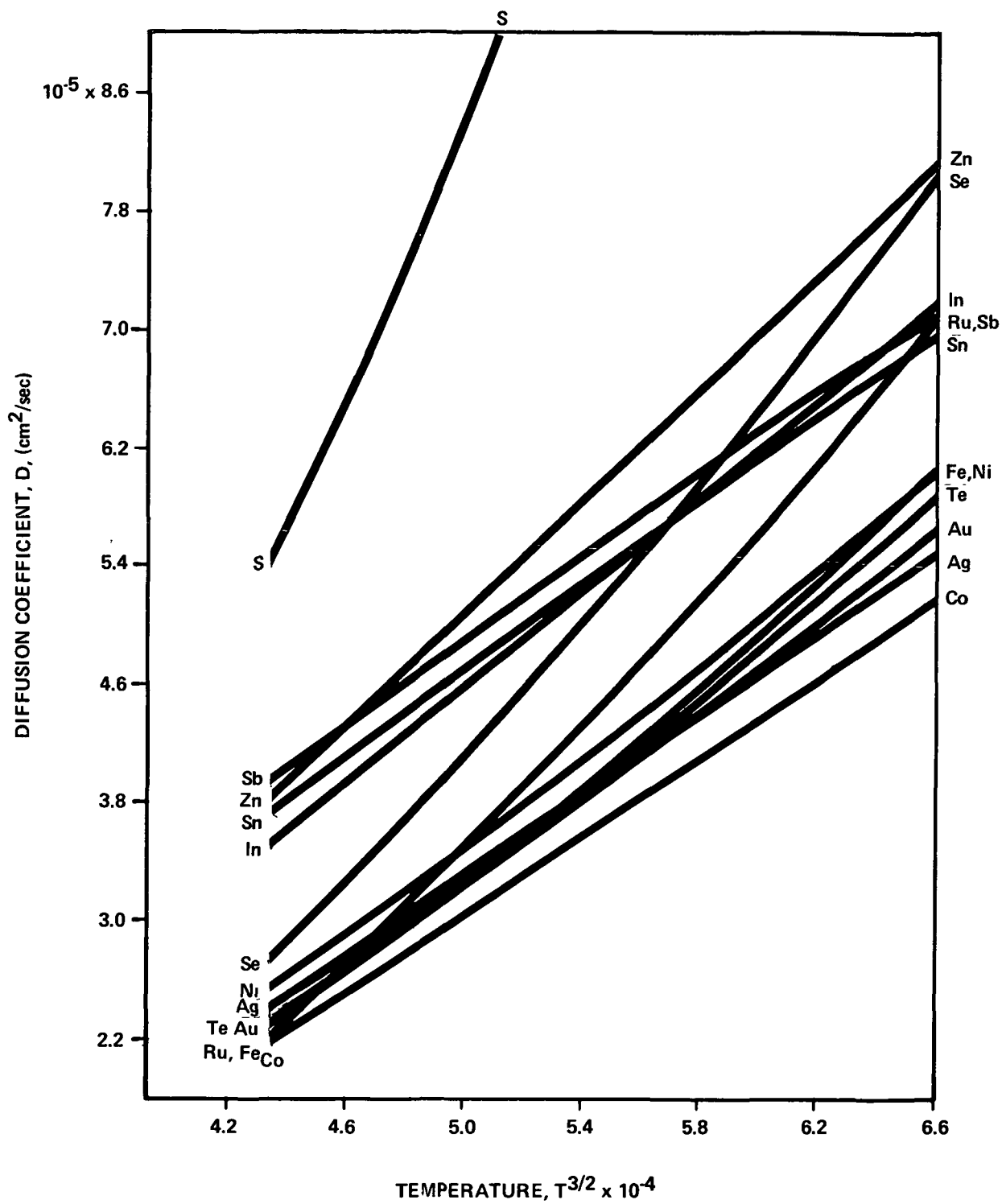


Figure 1 Diffusion coefficients of various solutes in liquid silver.

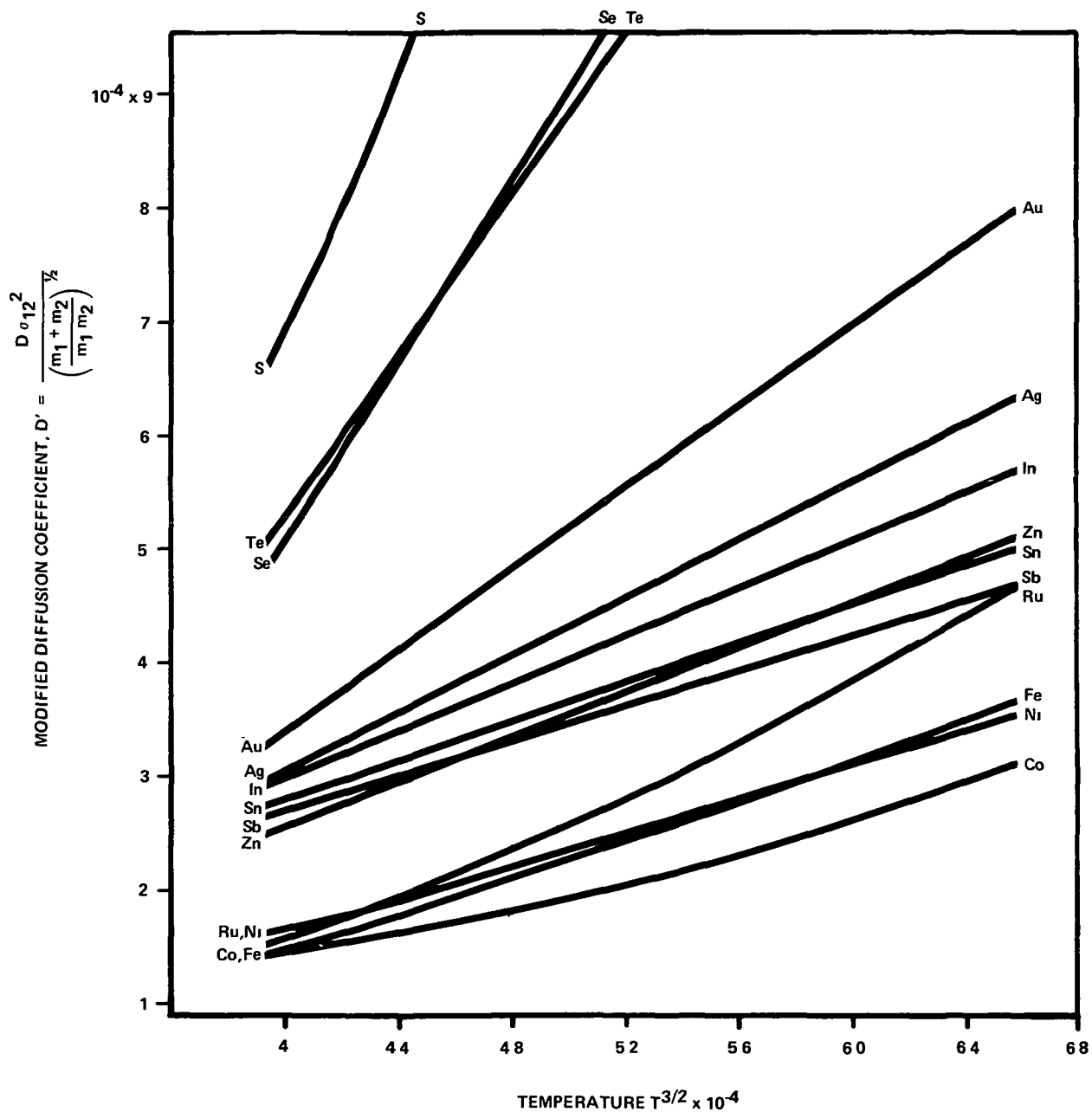


Figure 2 Plot of modified diffusion coefficients of various solutes in liquid silver.

$$D_{12} \cong \frac{(D'_{12})_{\text{group}} \sqrt{m_1 + m_2}}{\sigma_{12}^2} \quad (6)$$

Table 2 lists the values of D'_{12} for the three groups at the melting point. From this table the value of D'_{12} for the second group is 2.84. The value of diffusion coefficient of cadmium in liquid silver, for example, may now be obtained from equation (6) and is found to be

$$D_{\text{Cd in Ag}} = 3.07 \text{ cm}^2/\text{sec at } 961^\circ\text{C (1234}^\circ\text{K)} \quad (7)$$

Thus, the gas model provides a simple way of estimating the solute diffusion coefficients.

CONCLUSIONS

The diffusion of solutes in liquid metals can be satisfactorily described by use of a gas model in which ions of metals are assumed to behave like the molecules in a dense gas. The diffusion coefficient of a solute is a function of its mass, ionic size and pair potential. The model has been applied to solute diffusion in liquid silver. It is found that the solutes form groups in their diffusion behavior, depending on their position in the periodic table. It is possible to predict the diffusion coefficient of a solute with reasonable accuracy.

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TABLE 2. VALUES OF D'_{12} FOR THE THREE GROUPS AT MELTING POINT

Group	Element	Diffusion Coefficient at 961°C (1234°K)	D'_{12} Average
		$D'_{12} = \frac{D_{12} \sigma_{12}^2}{\sqrt{(m_1 + m_2)/m_1 m_2}}$	
1	S	6.57	5.48
	Se	4.82	
	Te	5.05	
2	Au	3.28	2.84
	Ag	2.97	
	In	2.91	
	Sn	2.74	
	Sb	2.65	
	Zn	2.48	
3	Fe	1.42	1.49
	Co	1.44	
	Ni	1.58	
	Ru	1.53	

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