THE DIFFUSIVITIES OF GASES IN PURE LIQUID METALS

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ABSTRACT

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The diffusion of gases in liquid metals is important for industrial and theoretical reasons. This research investigates the applicability of four selected theories for predicting diffusion coefficients of hydrogen, nitrogen, and oxygen in pure liquid metals. Experimentally-determined diffusion coefficients are compared with values predicted by four theories which were chosen for their predictive capacity and their theoretical value: hydrodynamical theory, absolute reaction-rate theory, vibrational displacement theory, and modified Enskog theory. No experimental datum is more accurate than ±2%, no theoretical prediction is more accurate than ±10%; the most difficult cases are those with hydrogen, those with silver, and those with gallium. For the oxygen-silver system, experimental diffusivity values are larger than those expected from any theory. The modified hydrodynamical theory and the modified Enskog theory appear the most useful for predicting coefficients for nitrogen in liquid iron and for oxygen in liquid metals. A proposed extension of these theories is necessary for accurate prediction of the coefficient of hydrogen.

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Chapter 1:

OVERVIEW AND REVIEW OF THE FOUR SELECTED THEORIES

1.1. Importance of Diffusion of Gases in Liquid Metals

Diffusion of gases in liquid metals is important for several reasons. First, in liquid-metal cooled fission and fusion reactors, it appears that many important corrosion mechanisms are limited by diffusion of contaminant gas in the liquid metal (Addison, 1972; Johnson, 1977; Klueh, 1984; Steinmeyer et al., 1981; Stewart and Sze, 1977). Second, the chemical, electrical, mechanical, and thermal properties of solid structural metals depend on the rate of gas absorption or desorption before and during solidification which is determined by diffusion of gas in the liquid metal (Birnbaum and Wert, 1972; Fromm and Hörz, 1980; Guy and Hren, 1974). Third, gas diffusion data and theories are necessary to provide information about the structure of liquids in general and liquid metals in particular (Ashcroft and Lekner, 1966; Reddy, 1984; Swamy and Reddy, 1984; Weber and Stillinger, 1984). There is no consistent general theory or model of the liquid metallic state which at the present predicts the physical, thermodynamic, and transport properties (Coffey et al., 1984; Hafner, 1977).

1.2. Four Theories of Liquid-State Diffusion

In historical order, the four selected theories of liquid-state diffusion are: hydrodynamical theory (Einstein, 1905), absolute reaction-rate theory (Eyring, 1936), vibrational displacement theory (Nachtrieb, 1967), and modified Enskog theory (Protopapas and Parlee, 1976). The first three theories yield simple mathematical expressions containing physical constants and measured properties of the liquid metal and provide a reasonable fit for self-diffusion data in liquid metals. The mathematical expressions of the modified Enskog theory, while somewhat more complicated than those of the other authors, have only one free parameter (covalent diameter of the gas) and are relatively simple to use.

Other excluded theories of diffusion include cluster theory (Egelstaff, 1962), free-volume theory (Cohen and Turnbull, 1959), corresponding-states theory (Pasternak and Olander, 1967), fluidity theory (Hildebrand, 1971; Sridhar and Potter, 1977), fluctuation theory (Swalin, 1959), and mobility theory (Walls and Upthegrove, 1964). These theories generally yield complicated mathematical expressions containing a number of physical properties of the gas-liquid pair, some of which are difficult to determine. A brief overview of the four selected theories follows.

1.2.1. The Hydrodynamical Theory

The hydrodynamical theory assumes solute particles are enough larger than solvent particles so that the solvent behaves as a continuum. The solute particle is often assumed to be spherical with a size independent of solute-solvent interaction; recently however, corrections have been proposed to account for these effects (Brenner, 1979, 1981; Emi, 1972).

For dilute solution with constant activity coefficients, the Nernst-Einstein equation gives (Levine, Chapter 16, 1978)

$$D_{AB} = kT\left(\frac{v_A}{F_A}\right) \tag{1}$$

where D_{AB} is the diffusion coefficient of A in B, k is Boltzmann's constant, T is absolute temperature, v_A is the average velocity of

A, and F_A is the average frictional force acting on A. For low Reynolds number flow, this force is (Sutherland, 1905; Lamb, 1945)

$$F_A = 6\pi\mu_B \, v_A \, r_A \, \frac{2\mu_B + r_A \, \beta_{AB}}{3\mu_B + r_A \, \beta_{AB}} \tag{2}$$

where μ_B is the solvent viscosity, r_A is the solute radius, and β_{AB} is a coefficient of sliding friction. Two limiting cases of Equation (2) are the no-slip condition, $\beta_{AB} = \infty$, and the no-stick condition, $\beta_{AB} = 0$. For the no-slip condition, Equation (2) reduces to Stokes's law:

$$F_A = 6\pi\mu_B \, v_A \, r_A \tag{3}$$

For the no-stick condition, Equation (2) becomes

$$F_A = 4\pi\mu_B \, v_A \, r_A \tag{4}$$

Substituting Equations (3) and (4) into Equation (1) gives two limiting expressions for the diffusion coefficient:

$$D_{AB} = \frac{kT}{6\pi\mu_B \, r_A} \tag{5}$$

and

$$D_{AB} = \frac{kT}{4\pi\mu_B \, r_A} \tag{6}$$

For diffusion of gases in liquid metals, where r_A is assumed to be the solute van der Waals radius, Equation (6) gives better results than Equation (5). However, according to the theoretical analysis of Lamm (1938) and of Lamm and Sjostedt (1938), if the diffusing particles are smaller than the solvent particles, the numerical factor 4 in Equation (6) is still too large.

Evans et al. (1981) showed experimentally that the hydrodynamical theory is not applicable for small solutes (e.g., argon and methane) diffusing in polyatomic organic liquids because this theory assumes solutes are much larger than solvents. They state that, based on a corresponding states-type analysis, past agreement between the hydrodynamical theory and experimental diffusivity data was purely accidental due to the narrow atomic density ranges over which experiments were performed. Their explanation cannot be tested for gas-liquid metal systems because there are not enough experimental data on a variety of gas-liquid metal systems over a broad atomic density range.

Equation (6) can be modified to

$$D'_{AB} = \frac{kT}{4\pi\mu_B\, r'_A} \tag{7}$$

where r'_A, the solute radius, is now a function of temperature and is given by (Protopapas and Parlee, 1976)

$$r'_A = r_c\,(1 - 0.112\,(T/T_m)^{\frac{1}{2}})10^{-8} \text{ (in cm)} \tag{8}$$

where T_m is the melting point of the liquid metal and r_c is the covalent radius of the solute. Protopapas and Parlee give no reason why the covalent radius is used in Equation (8). Examination of the data suggest a better fit is available if the radius were related to the ionic character of the solution. Thus, r'_A must be considered a free parameter which, by nature, is always close to the covalent radius. The results of the modified hydrodynamical theory, Equation (7), are seen to be consistently smaller than experimental results, but they are better than the results of the original hydrodynamical theory, Equation (6).

1.2.2. The Absolute Reaction-Rate Theory

In its original form, the absolute reaction-rate theory assumes particles are all of the same kind. Diffusion occurs through the action of holes (i.e., vacant sites). Accordingly, the diffusing particle jumps from its equilibrium position over a potential energy barrier into a nearby hole in the surrounding liquid (Eyring, 1936; Glasstone et al., Chapter 9, 1941). The sum of the energy to form a hole and the energy needed to surmount the barrier is called the activation energy for diffusion. Once a particle jumps into a hole, it must remain there long enough to dissipate the energy it acquired. For viscosity and self-diffusion, respectively, it can be shown that

$$\mu_B = kT \frac{\lambda_1}{\lambda_2 \lambda_3} \frac{1}{\lambda_v^2 kv} \tag{9}$$

and

$$D_{SELF} = \lambda_D^2 k_D \tag{10}$$

where λ_3 is the interparticle distance in the direction of viscous flow, λ_1, and λ_2 are interparticle distances perpendicular to λ_3, and λ_v and k_v are the jump-distance and absolute rate constant, respectively, for viscous flow. Similarly, λ_D and k_D are the jump-distance and absolute rate constant for diffusive flow. Consistency requires that the viscosity and self-diffusion coefficient of a liquid be related because the models are identical and because the jump-distance and absolute rate constants of the two processes are identical:

$$\lambda_v^2 k_v = \lambda_D^2 k_D \tag{11}$$

Substituting Equation (11) into Equation (9) and then Equation (9) into Equation (10) gives

$$D_{SELF} = \frac{kT}{\mu_B} \frac{\lambda_1}{\lambda_2 \lambda_3} \qquad (12)$$

For spherical particles in a quasi-crystalline lattice, this equation simplifies further because the interparticle distances equal the jump distance, which equals twice the solvent radius:

$$\lambda_1 = \lambda_2 = \lambda_3 = 2r_B \qquad (13)$$

where r_B is the solvent radius; thus

$$D_{SELF} = \frac{kT}{2\,\mu_B\,r_B} \qquad (14)$$

In 1958, Eyring modified this theory by assuming gas-like properties for particles which jump into holes, and assuming solid-like properties for all neighboring particle-hole pairs. The idea is that a liquid should have thermodynamic and transport properties intermediate between those of the gas and solid states. For the self-diffision coefficient of a liquid this (significant structures theory) gives (Eyring et al., 1958, 1960)

$$D_{SELF} = \frac{kT}{\xi\,\mu_B\,\lambda_D} = \frac{kT}{12\,\mu_B\,r_B} \qquad (15)$$

where ξ is the number of nearest neighbors in the same plane. For quasi-hexagonal close-packed liquids, $\xi = 6$ (Ree et al., 1958). Alternatively (Eyring and Ree, 1961; Ree et al., 1964),

$$D_{SELF} = \frac{kT}{\xi \frac{\lambda_2 \lambda_3}{\lambda_1} \mu_B} = \frac{kT}{6} \left(\frac{N_{AV}}{V_s\sqrt{2}}\right)^{1/3} \qquad (16)$$

where N_{AV} is Avogadro's number and V_S is the molar volume of the solid metal at its melting point.

For self-diffusion, Equations (14) - (16) are similar to expressions derived from hydrodynamical theory (e.g., Equations (5) and (6)); however, for Eyring's self-diffusion correlation to apply to impurity diffusion, all properties of the liquid metal solution (density, mean frequency of vibration, viscosity, jump distance, and absolute rate constant) must be identical to those in self-diffusion. These assumptions imply that the diffusion coefficient is independent of the diffusing gas species - a result which is not supported by experiment. Absolute reaction-rate theory remains valuable, as Equations (14) and (15) can be rewritten as

$$D' = \frac{kT}{2\,\mu_B\,r'_B} \qquad (17)$$

and

$$D' = \frac{kT}{12\,\mu_B\,r'_B} \qquad (18)$$

respectively; where r'_B, the solvent radius, is now a function of temperature. Choosing r'_B as that given by Protopapas and Parlee (1974),

$$r'_B = \frac{1}{2}\,1.288\left(\frac{AW}{\rho_m}\right)^{1/3}\left(1 - 0.112(T/T_m)^{1/2}\right)10^{-8}\ \text{(in cm)} \qquad (19)$$

where AW is the liquid metal atomic weight, ρ_m is the liquid metal density at its melting point, and T_m is the melting temperature. The results from this modification of absolute reaction-rate theory are seen to bracket all the experimental results (Chapter 3, Section 3).

1.2.3. The Vibrational Displacement Theory

Nachtrieb (1967) proposed his vibrational displacement theory because of the observation that the self-diffusion coefficient of liquid metals is linearly proportional to temperature. Also, Nachtrieb was able to show that experimental data is just as well represented by a linear relation (D vs. T) as by an Arrhenius-type, activated state, relation (ln D vs. 1/T). This implys that diffusion in liquid metals is not a simple, thermally activated process.

Vibrational displacement theory proposes that every particle is enclosed in a cage composed of its nearest neighbors. From equipartition of energy (which is valid at high temperatures), every particle is assumed to vibrate with energy

$$\overline{E} = 3kT = \tfrac{1}{2}\kappa\overline{r^2} \tag{20}$$

where $\overline{E}$ is the average energy, κ is the force constant of the metal (Pauling, Chapter 7, 1960), and $\overline{r^2}$ is the mean-squared thermal amplitude. From Equation (20),

$$\overline{r^2} = \frac{6kT}{\kappa} \tag{21}$$

At this point, $\overline{r^2}$ is inserted into the Einstein diffusion equation for random walks:

$$D = \frac{1}{6}\,\overline{r^2}\,\tau \tag{22}$$

where D is the diffusion coefficient and τ is the characteristic time of diffusion. This result provides an accurate prediction for self-diffusivity at the melting point if the vibrations are all in phase, and if τ is approximately equal to the Debye frequency (ν_D) of the solid metal. The Debye frequency is usually given in terms of a Debye temperature:

$$\theta_D = \frac{h\ \nu_D}{k} = \frac{h\ \tau}{k} \tag{23}$$

where θ_D is the Debye temperature and h is Planck's constant. Combining Equations (22) and (23) yields an equation which predicts identical diffusion coefficients for all gases in a given liquid metal:

$$D = \frac{k^2\ \theta_D\ T}{h\ \kappa} \tag{24}$$

The results from this theory are seen to be consistently much smaller than experimental data (Chapter 3, Section 4).

1.2.4. The Modified Enskog Theoery

Thorne, in an unpublished work (Hirchfelder et al., Chapter 9, 1964; Chapman and Cowling, Chapter 16, 1970), extended the Enskog theory of pure liquids (Enskog and Svenskii, 1921; Enskog, 1922) to binary liquid mixtures. According to Thorne, the binary diffusion coefficient is given by

$$D_{12} = D'_{12}/g_{12} \tag{25}$$

where D'_{12} is the binary diffusion coefficient in dilute solution and g_{12} is the pair-correlation (or radial distribution) function. For hard spheres in dilute solution,

$$D'_{12} = \frac{3}{8n\ \sigma_{12}^2} \left(\frac{kT\ (m_1 + m_2)}{2\pi m_1\ m_2} \right)^{1/2} \tag{26}$$

where m_1 and m_2 are atomic masses of the two components, n is the total particle density, and σ_{12} is the center-to-center distance between two dissimilar hard spheres in contact:

$$\sigma_{12} = \frac{\sigma_1 + \sigma_2}{2} \tag{27}$$

Here σ_1 and σ_2 are the hard-sphere diameters of the gas and liquid metal, respectively.

Lebowitz (1964) derived expressions for the pair-correlation function at contact between similar and dissimilar hard spheres. In an early paper, Protopapas and Parlee (1975) corrected Lebowitz's expression for dissimilar hard spheres:

$$g_{12} = \frac{2 - \eta}{(2 + \eta)(1 - \eta)^3}\left(1 + \frac{\eta}{2}\left(1 - 3\,\frac{(\sigma_1 - \sigma_2)(\eta_1 - \eta_2)}{(\sigma_1 + \sigma_2)(\eta_1 + \eta_2)}\right)\right) \tag{28}$$

where η_1 and η_2 are the packing fractions of the two components given by

$$\eta_i = \left(\frac{\pi}{6}\right) n_i \sigma_i^{\,3} \tag{29}$$

and

$$\eta = \eta_1 + \eta_2 \tag{30}$$

Taking a different tack, Alder, et al. (1974) used molecular dynamics to determine correction factors for the Enskog theory based on mass ratio, diameter ratio, and packing fraction of the hard-sphere particles. Thus, Equation (25) becomes

$$D_{12} = (CF)\, D'_{12}/g_{12} \tag{31}$$

where CF is the correction factor.

Protopapas and Parlee used the data of Alder et al., and an idea first suggested by Dymond and Alder, to obtain temperature-dependent expressions for hard-sphere diameters which are then incorporated into the correlation (Dymond and Alder, 1966, 1968, 1970; Protopapas and Parlee, 1974, 1976). The idea is to use hard-sphere diameters which shrink with increasing temperature, thus giving, effectively, soft spheres. This idea combines the mathematical simplicity of hard spheres with the reality of soft spheres. Expressions obtained by Protopapas and Parlee for the hard-sphere diameters are given by Equations (8) and (19) (multiply these equations by 2 to get diameters). Generally, the results from this modified Enskog theory are larger than experimental results (Chapter 3, Section 5).

Chapter 2:

EXPERIMENTAL METHODS IN GAS-LIQUID METAL DIFFUSION STUDIES

The main experimental techniques for measuring diffusion coefficients of gases in liquid metals are the capillary-cell methods and electrochemical methods; the latter are used for oxygen diffusion only. A brief overview of these methods follows.

2.1. Capillary-Cell Methods

Capillary-cell measurements are either steady-state or unsteady-state. In either case, gas is dissolved in a stagnant sample of liquid metal and the volume of gas absorbed is measured as a function of time. The rate of gas absorption is an accurate measure of diffusivity because convection and surface effects appear to have little effect (El-Tayeb and Parlee, 1967; Birnbaum and Wert, 1972). In the former method, the system is allowed to reach steady state before gas volumes are recorded (Sacris and Parlee, 1970).

The unsteady-state method (Figure 1) typically assumes semi-infinite boundary conditions (e.g., Mizikar et al., 1962):

$$V = \frac{d^2 \rho_m C_s (60\pi Dt)^{1/2}}{200\rho_g} \tag{32}$$

where V is the STP (standard temperature and pressure) volume of gas absorbed, d is the capillary cell diameter, ρ_m is the liquid metal density at temperature T, ρ_g is the STP density of absorbed gas, C_s is the gas saturation concentration in the liquid metal, D is the diffusion coefficient, and T is time. A plot of V vs. $t^{\frac{1}{2}}$ should be a straight line whose slope can be used to calculate D.

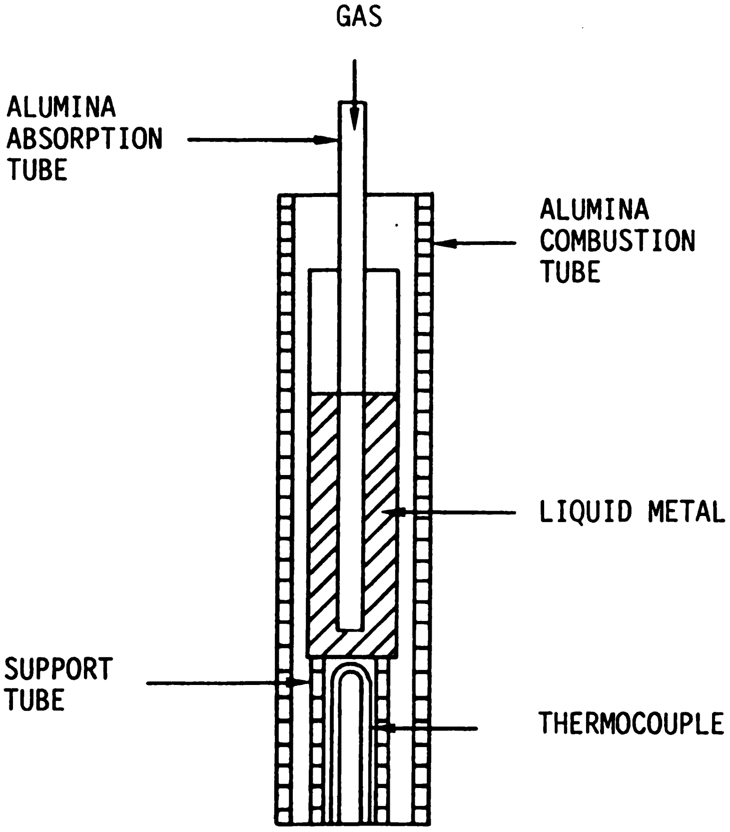

Figure 1. Schematic of the unsteady-state capillary-cell (after El-Tayeb and Parlee, 1967).

For the steady-state method (Figure 2), the solution is (Velho et al., 1969)

$$V = D \frac{\rho_m}{\rho_g} \frac{A_1 A_2}{A_1 L_2 + A_2 L_1} \frac{C_1 - C_0}{100} t \tag{33}$$

where A_1 is the internal cross-sectional area of the capillary tube, A_2 is the annular cross-sectional area between the capillary tube and the crucible wall, L_1 is the depth of immersion of the capillary tube, L_2 is the depth of stagnant liquid metal, C_1 is the saturation concentration of gas at the liquid metal surface inside the capillary tube, and C_o is the saturation concentration of gas at the liquid metal surface outside the capillary tube. A plot of V vs. t should be a straight line whose slope can be used to calculate D.

2.2. Electrochemical Methods

The electrochemical methods are either galvanostatic (Figure 3), or potentiostatic (Figure 4): both use a solid electrolye, either calcia-stabilized zirconia (ZrO_2 - CaO) or yttria-doped thoria (ThO_2 - Y_2O_3). As before, gas is dissolved in a sample of stagnant liquid metal, and the rate of gas absorption is measured as the change in electromotive force (emf) between the two electrodes for the galvanostatic method (Sano et al., 1970; Honma et al., 1971) or the change in cell current between the two electrodes for the potentiostatic method (Ramanarayanan and Rapp, 1972; Szwarc et al., 1972; Oberg et al., 1973; Klinedinst and Stevenson, 1973).

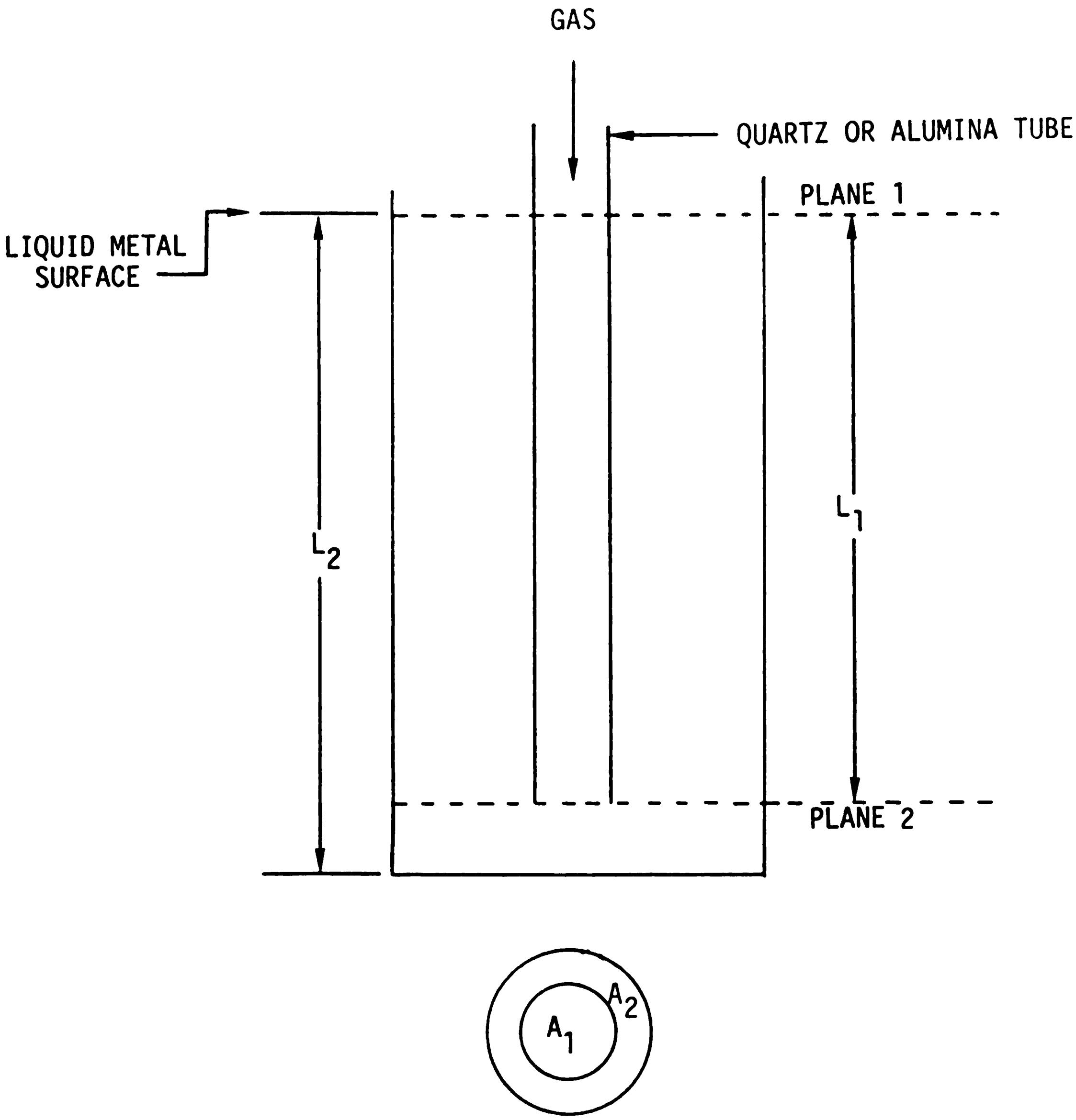

Figure 2. Schematic of the steady-state capillary-cell (after Velho et al., 1969).

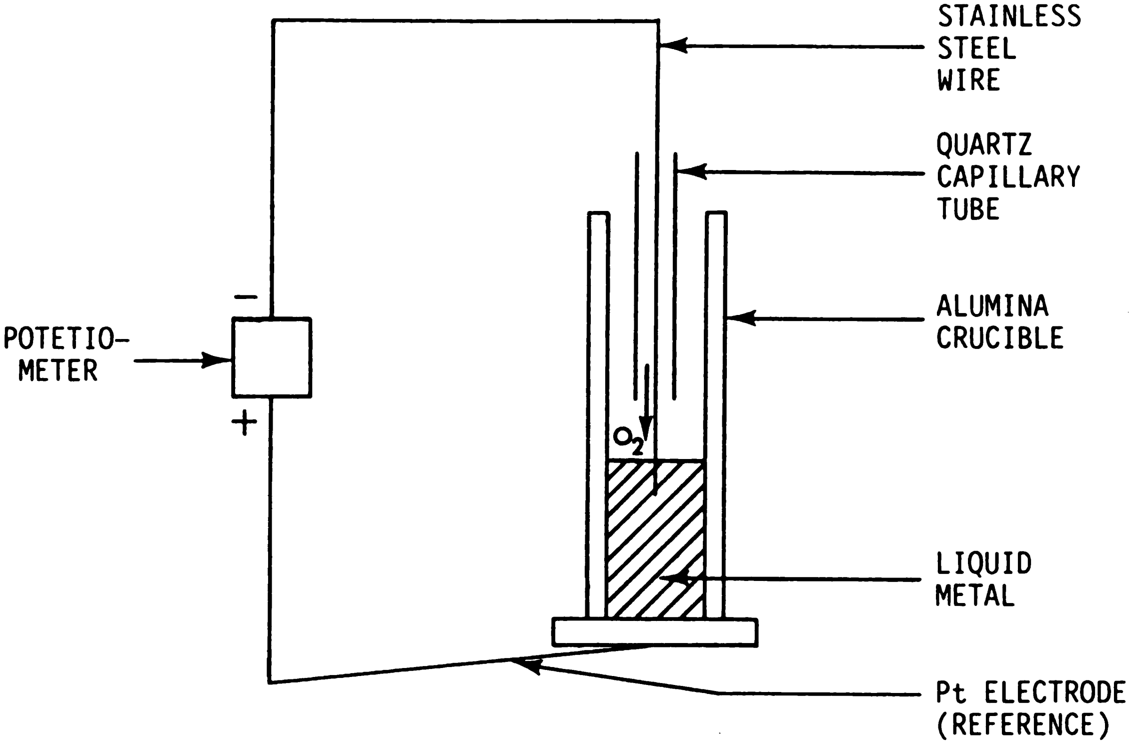

Figure 3. Schematic of electrochemcial cell for the galvanostatic method (after Sano etal., 1970).

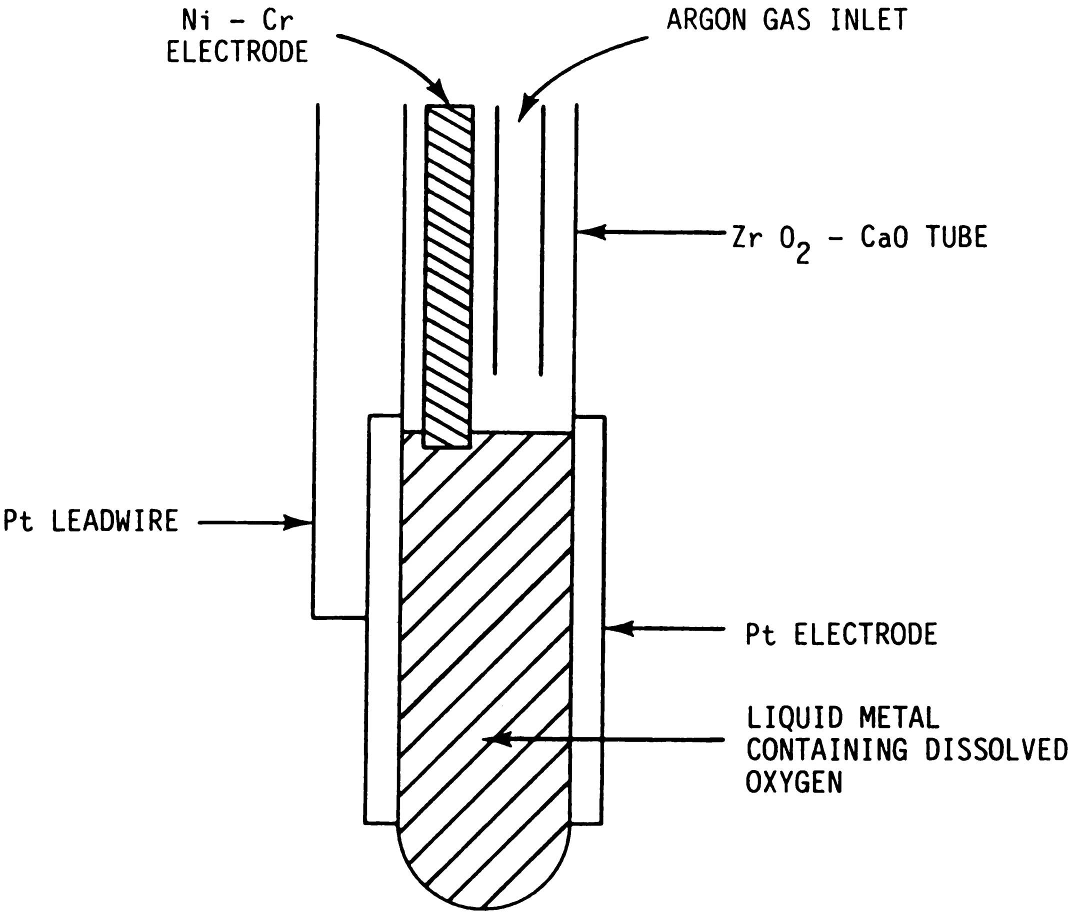

Figure 4. Schematic of electrochemcial cell for the potentiostatic method (after Oberg et al., 1973).

For both the capillary-cell methods and the electrochemical methods, the diffusion coefficient is assumed to be independent of gas concentration; this is equivalent to assuming that the activity coefficient is independent of gas concentration (Hahn and Stevenson, 1977).

Chapter 3:

RESULTS AND DISCUSSION

3.1. Experimentally-Determined Diffusion Coefficients of Gases in Pure Liquid Metals

Table 1 lists (alphabetically, by system) all the experimental diffusion data known to the author for hydrogen, nitrogen, and oxygen in pure liquid metals. Where available, D_o and Q are listed for a fit of the data to an Arrhenius relation:

$$D = D_o \exp(-Q/RT) \tag{34}$$

Equation (34) is used only for the convenience it gives, it is no less accurate than the data it correlates. D_o may be related to the structure and packing of particles in the liquid metal, but little is known about the independent character of D_o at this time (Wilson, 1965; Hahn and Stevenson, 1977). In the table, data marked with an asterisk is considered questionable.

Hydrogen-Liquid Metal Systems--For H-Al, the data of Byalik et al. are reasonable, but the temperature range is narrow. The data of Vashchenko et al. do not compare with those of other researchers. For H-Cu, the data of Sigrist et al. do not compare with those of other researchers. For H-Fe, the data of Arkharov et al. and Nyquist do not compare with those of other researchers. For H-Li, the data of Alire below 800°C are suspect because of an erroneous correlation used in determining experimental diffusivity values (Buxbaum and Johnson, 1982).

Nitrogen-Liquid Metal Systems--For N-Fe, the data of Chesnokov et al. and Lee and Parlee do not compare with those of other researchers.

TABLE 1

Diffusion data for gases in pure liquid metals.
An asterisk indicates questionable data.
Number in parentheses indicates reference.

System	D_0 (cm²/sec)	Q (cal/mol)	Temp. Range (°C)	Researcher(s)
H-Ag	4.54×10^{-2}	1359	985-1208	Sacris & Parlee (106)
*H-Al	7.47×10^{-1}	7900	900-1000	Byalik et al. (18)
H-Al	3.8×10^{-2}	4600	780-1000	Eichenauer & Markopoulos (33)
*H-Al	2.34×10^{-3}	15000	670-985	Vashchenko et al. (124)
H-Co	---	---	1600	Depuydt (26)
H-Cu	1.46×10^{-2}	4500	1100-1250	Chernega & Vashchenko
H-Cu	10.91×10^{-3}	2148	1103-1433	Sacris & Parlee (106)
*H-Cu	5.12×10^{-3}	5880	1103-1361	Sigrist et al. (111)
H-Cu	---	---	1101,1201	Wright & Hocking (130)
H-Cu	---	---	1090-1205	Yang & Lee (131)
*H-Fe	5.21×10^{-2}	10000	1560-1650	Arkharov et al. (6)
H-Fe	---	---	1600	Bester & Lange (12)
H-Fe	4.37×10^{-3}	4134	1550-1680	Depuydt & Parlee (27)
H-Fe	3.2×10^{-3}	3300	1547-1726	El-Tayeb & Parlee (37)
H-Fe	---	---	1550-1750	Ershov & Kasatkin (42)
H-Fe	---	---	1550-1650	Linchevskii & Shal'kevich (76)
*H-Fe	1.86×10^{-3}	9370	1550-1650	Nyquist (83)
H-Fe	2.57×10^{-3}	4100	1550-1720	Solar & Guthrie (112)
H-Li	13.0	25000	625-900	Alire (3)
H-Li	---	---	800-900	Buxbaum & Johnson (17)

TABLE 1 (Continued)

System	D_0 (cm^2/sec)	Q (cal/mol)	Temp. Range (°C)	Researcher(s)
H-Ni	---	---	1500-1750	Ershov & Kasatkin (42)
H-Ni	---	---	1450-1550	Linchevskii & Shal'kevich (76)
H-Ni	7.47×10^{-3}	8550	1478-1600	Sacris & Parlee (106)
*H-Ni	5×10^{-2}	9736	1468-1550	Wright & Hocking (130)
H-Sn	---	---	1081-1105	Ebro (31)
H-Sn	---	---	1000-1300	Sacris & Parlee (106)
N-Co	---	---	1600	Benner (10)
N-Fe	1.07×10^{-3}	11000	1600-1700	Arkharov et al. (5)
N-Fe	---	---	1600	Atarashiya (8)
N-Fe	2.86×10^{-3}	14600	1550-1700	Bogdanov et al. (14)
*N-Fe	0.4	15200	1550-1650	Chesnokov et al. (23)
N-Fe	---	---	1550-1750	Ershov & Kasatkin (42)
N-Fe	1.07×10^{-3}	11000	1560-1700	Ershov & Kovalenko (43)
N-Fe	---	---	1550-1680	Inouye et al. (61)
N-Fe	---	---	1550-1700	Kunze (68, 69)
*N-Fe	1.83×10^{-2}	23120	1560-1680	Lee & Parlee (74)
N-Fe	---	---	1600	Schwerdtfeger (108)
N-Fe	---	---	1600	Svjazin & El-Gammal (120)
N-Ni	---	---	1600	Benner (10)
N-Ni	6.602×10^{-2}	6580	1500-1700	Chesnokov & Linchevskii (22)

TABLE 1 (Continued)

System	D_0 (cm²/sec)	Q (cal/mol)	Temp. Range (°C)	Researcher(s)
N-Ni	---	---	1550-1750	Ershov & Kasatkin (42)
N-Zn	---	10100	466-574	Pechenyakov et al. (97)
O-Ag	---	---	950-1150	Besson et al. (11)
O-Ag	5.15×10^{-3}	9900	970-1200	Masson & Whiteway (77)
O-Ag	1.47×10^{-3}	7100	1000-1200	Mizikar et al. (79)
O-Ag	2.8×10^{-3}	8300	1000-1350	Oberg et al. (85)
O-Ag	1.85×10^{-3}	7500	980-1130	Otsuka et al. (88)
O-Ag	---	---	1000-1150	Otsuka & Kozuka (89)
O-Ag	1.47×10^{-3}	7100	950-1200	Parlee & Zeibel (94)
*O-Ag	2.63×10^{-3}	7300	990-1220	Rickert & El-Milligy (105)
O-Ag	3.0×10^{-3}	8700	1000-1200	Sano et al. (107)
O-Ag	2.2×10^{-3}	7900	1000-1200	Shah & Parlee (109)
O-Ag	---	---	1100	Suito et al. (117)
*O-Cu	7.25×10^{-3}	15722	1100-1300	El-Naggar & Parlee (36)
*O-Cu	2.61×10^{-2}	16700	1100-1400	Gerlach et al. (52)
O-Cu	---	---	1200	Kramss et al. (67)
O-Cu	6.9×10^{-3}	12900	1000-1350	Oberg et al. (85)
*O-Cu	2.63×10^{-3}	9370	1100-1350	Osterwald & Schwarzlose (87)
O-Cu	5.7×10^{-3}	11900	1128-1322	Otsuka & Kozuka (91)
*O-Cu	1.22×10^{-2}	14400	1100-1250	Rickert & El-Miligy (105)

TABLE 1 (Continued)

System	D_O (cm²/sec)	Q (cal/mol)	Temp. Range (°C)	Researcher(s)
*O-Cu	1.55×10^{-4}	8330	1100-1300	Shurygin & Kryuk (110)
*O-Fe	6.23×10^{-3}	10800	1550-1650	Ershov & Bychev (41)
O-Fe	---	---	1550-1750	Ershov & Kasatkin (42)
O-Fe	---	---	1550	Kawakami & Goto (64)
O-Fe	---	---	1560,1660	McCarron & Belton (78)
*O-Fe	3.34×10^{-3}	12000	1550-1680	Novokhatskii & Ershov (82)
O-Fe	---	---	1560	Otsuka & Kozuke (93)
O-Fe	---	---	1610	Schwerdtfeger (108)
O-Fe	---	---	1610	Shurygin & Kryuk (110)
*O-Fe	5.59×10^{-3}	19550	1560-1660	Suzuki & Mori (119)
O-Ga	3.68×10^{-3}	8370	750-950	Klinedinst & Stevenson (65)
O-In	8.22×10^{-4}	12600	750-950	Klinedinst & Stevenson (65)
O-Ni	---	---	1550-1750	Ershov & Kasatkin (42)
O-Ni	---	---	1500	Otsuka & Kozuka (93)
*O-Pb	6.32×10^{-5}	3580	700-900	Arcella (4)
O-Pb	---	---	750	Bandyopadhyay & Ray (9)
*O-Pb	9.65×10^{-5}	4800	800-1100	Honma et al. (60)
O-Pb	---	---	800	Kawakami * Goto (63)
O-Pb	1.90×10^{-3}	5000	800-1100	Osterwald & Charle (86)
O-Pb	1.48×10^{-3}	4660	800-1135	Otsuka & Kozuka (90)

TABLE 1 (Continued)

System	D_0 (cm^2/sec)	Q (cal/mol)	Temp. Range (°C)	Researcher(s)
O-Pb	1.44×10^{-3}	6200	740-1080	Szwarc et al. (123)
O-Sb	3.07×10^{-4}	938	697-857	Fitzner (50)
O-Sn	6.4×10^{-4}	4600	700-930	Otsuka & Kozuka (92)
O-Sn	9.9×10^{-4}	6300	784-1000	Ramanaryanan & Rapp (101)

Oxygen-Liquid Metal Systems--For O-Ag, the data are fairly uniform; moreover, D values calculated from Equation (34) are larger than those expected from any theory. Oberg et al. point out possible problems with the experiments of Rickert and El-Miligy. For O-Cu, the El-Naggar and Parlee and Gerlach et al. Q values are larger than the others. Oberg et al. point out possible problems with the experiments of Osterwald and Schwarzlose and Rickert and El-Miligy. The data of Shurygin and Kryuk do not compare with those of other researchers. For O-Fe, the data of Suzuki and Mori do not compare with those of other researchers. The data of Ershov and Bychev and Novokhatskii and Ershov are reasonable, but Ershov and Bychev used a narrow temperature range and Novokhatskii and Ershov took data at only three temperatures. For O-Ga and O-In, identical temperatures and temperature ranges were used, but the diffusion coefficient is much larger in gallium. Hahn and Stevenson noted that one possible explanation for this difference is the anomolous structure (from X-ray diffraction studies) of gallium. For most simple liquid metals with normal (i.e., random) structure (e.g., alkali metals, copper, gold, indium, and silver), the X-ray diffraction pattern gives a sharp symmetrical first peak. For some liquid metals (e.g., antimony, bismuth, gallium, germanium, lead, mercury, and zinc), the first peak is not symmetrical, but has a shoulder on the high-angle side (Wilson, 1965). This indicates some form of bonding or other interaction between particles, which lends credence to a cluster model for diffusion, such as that proposed by Egelstaff. For O-Pb, Otsuka and Kozuka point out possible problems with the experiments of Arcella and Honma et al.

3.2. Comparison of Experimental Data With Predictions From Hydrodynamical Theory

Table 2 compares experimental data with predictions from hydrodynamical theory. In this table, D_{EX} is the experimental value (from Equation (34)), D_H is given by Equation (6), and D_H' is given by Equation (7). Solvent viscosity values were taken from literature (Elliott and Gleiser, 1960) or calculated from a corresponding-states correlation (Chapman, 1966).

Hydrogen-Liquid Metal Systems--Except for H-Li, D_{EX} values are very much larger than D_H and D_H' values.

Nitrogen-Liquid Metal Systems--For N-Fe, D_{EX} values compare very favorably with D_H' values.

Oxygen-Liquid Metal Systems--As also seen in Figure 7 for O-Ag, D_{EX} values are about twice as large as D_H values and about four times as large as D_H' values. For O-Sn, D_{EX} values are smaller than D_H and D_H' values.

3.3. Comparison of Experimental Data With Predictions From Absolute Reaction-Rate Theory

Table 3 compares experimental data with predictions from absolute reaction-rate theory. In this table, D_{EX} is the experimental value (from Equation (34)); D_R, D_R', D_{MR}, D_{MR}', and D_{SS} (which will be referred to collectively as D_{ART}) are given by Equations (14), (17), (15), (18), and (16), respectively. The Goldschmidt radius is used for r_B because it is nearly identical with the covalent radius and is readily available in the literature (Chapman, 1966). Molar volumes were taken from the literature (Elliott and Gleiser, 1960; Protopapas and Parlee, 1974, 1976).

TABLE 2

Comparison of experimental data with the hydrodynamical theory.
An asterisk indicates quesitonable data.
Number in parentheses indicates reference.

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm^2/sec)	$D_H \times 10^{-5}$ (cm^2/sec)	$D'_H \times 10^{-5}$ (cm^2/sec)	Researcher(s)
H-Cu	1200	314	4.21	15.5	Chernega & Vashchenko (21)
		524			Sacris & Parlee (106)
*		68.7			Sigrist et al. (111)
H-Cu	1300	549	4.98	18.4	Sacris & Parlee (106)
*		78.0			Sigrist et al. (111)
*H-Fe	1600	355	2.81	10.3	Arkharov et al. (6)
		144			Depuydt & Parlee (27)
		132			El-Tayeb & Parlee (37)
*		15.0			Nyquist (83)
		85.4			Solar & Guthrie (112)
H-Fe	1700	138	3.22	11.8	El-Tayeb & Parlee (37)
		90.3			Solar & Guthrie (112)
*H-Li	700	3.15	38.7	150	Alire (3)
H-Li	800	10.5	46.8	183	Alire (3)
N-Fe	1600	5.57	2.25	5.14	Arkarov et al. (5)
		5.66			Bogdanov et al. (14)
*		673			Chesnokov et al. (23)
		5.57			Ershov & Kovalenko (43)
*		3.67			Lee & Parlee (74)
N-Fe	1700	6.47	2.58	5.93	Arkarov et al. (5)
		6.90			Bogdanov et al. (14)
		6.47			Ershov & Kovalenko (43)
N-Ni	1500	1020	2.44	5.58	Chesnokov & Linchevskii (22)
N-Ni	1600	1127	2.99	6.87	Chesnokov & Linchevskii (22)

TABLE 2 (Continued)

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm²/sec)	$D_H \times 10^{-5}$ (cm²/sec)	$D_H' \times 10^{-5}$ (cm²/sec)	Researcher(s)
O-Ag	1000	10.3	2.57	5.49	Masson & Whiteway (77)
		8.88			Mizikar et al. (79)
		10.5			Oberg et al. (85)
		9.54			Otsuka et al. (88)
		8.88			Parlee & Zeibel (94)
*		14.7			Rickert & El-Miligy (105)
		9.62			Sano et al. (107)
		9.68			Shah & Parlee (109)
O-Ag	1100	13.8	3.18	6.84	Masson & Whiteway (77)
		10.9			Mizikar et al. (79)
		13.7			Oberg et al. (85)
		11.8			Otsuka et al. (88)
		10.9			Parlee & Zeibel (94)
*		18.1			Rickert & El-Miligy (105)
		12.4			Sano et al. (107)
		12.2			Shah & Parlee (109)
O-Ag	1200	17.5	3.86	8.34	Masson & Whiteway (77)
		13.0			Mizikar et al. (79)
		16.4			Oberg et al. (85)
		13.0			Parlee & Zeibel (94)
*		21.7			Rickert & El-Miligy (105)
		15.4			Sano et al. (107)
		14.8			Shah & Parlee (109)
*O-Cu	1100	2.28	2.98	6.35	El-Naggar & Parlee (36)
*		5.73			Gerlach et al. (52)
		6.10			Oberg et al. (85)
*O-Cu		8.48			Osterwald & Schwarzlose (87)
*		6.22			Rickert & El-Miligy (105)
*		0.73			Shurygin & Kruyuk (110)

TABLE 2 (Continued)

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm^2/sec)	$D_H \times 10^{-5}$ (cm^2/sec)	$D_H' \times 10^{-5}$ (cm^2/sec)	Researcher(s)
*O-Cu	1200	3.37	3.61	7.72	El-Naggar & Parlee (36)
*		8.68			Gerlach et al. (52)
		8.41			Oberg et al. (85)
*		10.7			Osterwald & Schwarzlose (87)
		9.78			Otsuka & Kozuka (91)
*		8.91			Rickert & El-Miligy (105)
*		0.90			Shurygin & Kryuk (110)
*O-Cu	1300	4.74	4.27	9.18	El-Naggar & Parlee (36)
*		12.5			Gerlach et al. (52)
		11.1			Oberg et al. (85)
*		12.2			Osterwald & Schwarzlose (87)
		12.7			Otsuka & Kozuka (91)
*		1.08			Shurygin & Kryuk (110)
*O-Fe	1600	34.2	2.41	5.14	Ershov & Bychev (41)
*		13.3			Novokhatskii & Ershov (82)
*		2.96			Suzuki & Mori (119)
*O-Pb	900	1.36	8.22	18.4	Arcella (4)
*		1.23			Honma et al. (60)
		22.2			Osterwald & Charle (86)
		20.0			Otsuka & Kozuka (90)
		10.1			Szwarc et al. (123)
*O-Pb	1000	1.45	9.60	21.7	Honma et al. (60)
		26.3			Osterwald & Charle (86)
		23.5			Otsuka & Kozuka (90)
		12.4			Szwarc et al. (123)
O-Sn	800	7.40	9.67	21.9	Otsuka & Kozuka (92)
		5.16			Ramanaryanan & Rapp (101)
O-Sn	1000	8.20	12.5	28.7	Ramanaryanan & Rapp (101)

TABLE 3

Comparison of experimental data with the absolute reaction-rate theory.
An asterisk indicates questionable data.
Number in parentheses indicates reference.

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm^2/sec)	$D_R \times 10^{-5}$ (cm^2/sec)	$D_R' \times 10^{-5}$ (cm^2/sec)	$D_{MR} \times 10^{-5}$ (cm^2/sec)	$D_{MR}' \times 10^{-5}$ (cm^2/sec)	$D_{SS} \times 10^{-5}$ (cm^2/sec)	Researcher(s)
H-Cu	1200	314	24.8	27.9	4.14	4.64	3.98	Chernega & Vashchenko (21)
		524						Sacris & Parlee (106)
*		68.7						Sigrist et al. (111)
H-Cu	1300	549	29.3	32.9	4.89	5.52	4.71	Sacris & Parlee (106)
*		78.0						Sigrist et al. (111)
*H-Fe	1600	355	16.8	18.8	2.80	3.13	2.69	Arkharov et al. (6)
		144						Depuydt & Parlee (27)
		132						El-Tayeb & Parlee (37)
*		15.0						Nyquist (83)
		85.4						Solar & Guthrie (112)
H-Fe	1700	138	19.3	21.6	3.22	3.61	3.09	El-Tayeb & Parlee (37)
		90.3						Solar & Guthrie (112)
*H-Li	700	3.15	186	227	31.0	37.8	30.6	Alire (3)
H-Li	800	10.5	225	277	37.4	46.1	37.0	Alire (3)
N-Fe	1600	5.57	16.8	18.8	2.80	3.13	2.69	Arkharov et al. (5)
		5.66						Bogdanov et al. (14)
*		673						Chesnokov et al. (23)
		5.57						Ershov & Kovalenko (43)
*		3.67						Lee & Parlee (74)

TABLE 3 (Continued)

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm^2/sec)	$D_R \times 10^{-5}$ (cm^2/sec)	$D_R' \times 10^{-5}$ (cm^2/sec)	$D_{MR} \times 10^{-5}$ (cm^2/sec)	$D_{MR}' \times 10^{-5}$ (cm^2/sec)	$D_{SS} \times 10^{-5}$ (cm^2/sec)	Researcher(s)
N-Fe	1700	6.47	19.3	21.6	3.22	3.61	3.09	Arkhavov et al. (5)
		6.90						Bogdanov et al. (14)
		6.47						Ershov & Kovalenko (43)
N-Ni	1500	1020	18.4	20.6	3.07	3.43	2.95	Chesnokov & Linchevskii (22)
N-Ni	1600	1127	22.6	25.4	3.76	4.23	3.62	Chesnokov & Linchevskii (22)
O-Ag	1000	10.3	15.7	17.5	2.61	2.92	2.50	Masson & Whiteway (77)
		8.88						Mizikar et al. (79)
		10.5						Oberg et al. (85)
		9.54						Otsuka et al. (88)
		8.88						Parlee & Zeibel (94)
*		14.7						Rickert & El-Miligy (105)
		9.62						Sano et al. (107)
		9.68						Shah & Parlee (109)
O-Ag	1100	13.8	19.4	21.7	3.23	3.62	3.10	Masson & Whiteway (77)
		10.9						Mizikar et al. (79)
		13.7						Oberg et al. (79)
		11.8						Otsuka et al. (88)
		10.9						Parlee & Zeibel (94)
*		18.1						Rickert & El-Miligy (105)
		12.4						Sano et al. (107)
		12.2						Shah & Parlee (109)

TABLE 3 (Continued)

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm^2/sec)	$D_R \times 10^{-5}$ (cm^2/sec)	$D'_R \times 10^{-5}$ (cm^2/sec)	$D_{MR} \times 10^{-5}$ (cm^2/sec)	$D'_{MR} \times 10^{-5}$ (cm^2/sec)	$D_{SS} \times 10^{-5}$ (cm^2/sec)	Researcher(s)
O-Ag	1200	17.5	23.6	26.6	3.93	4.43	3.77	Masson & Whiteway (77)
		13.0						Mizikar et al. (79)
		16.4						Oberg et al. (85)
		13.0						Parlee & Zeibel (94)
*		21.7						Rickert & El-Miligy (105)
		15.4						Sano et al. (107)
		14.8						Shah & Parlee (109)
*O-Cu	1100	2.28	20.4	22.9	3.41	3.82	3.28	El-Naggar & Parlee (36)
*		5.73						Gerlach et al. (52)
		6.10						Oberg et al. (85)
*		8.48						Osterwald & Schwarzlose (87)
*		6.22						Rickert & El-Miligy (105)
*		0.73						Shurygin & Kryuk (110)
*O-Cu	1200	3.37	24.8	27.9	4.14	4.64	3.98	El-Naggar & Parlee (36)
*		8.68						Gerlach et al. (52)
		8.41						Oberg et al. (85)
*		10.7						Osterwald & Schwarzlose (87)
		9.78						Otsuka & Kozuka (91)
*		8.91						Rickert & El-Miligy (105)
*		0.90						Shurygin & Kryuk (110)

TABLE 3 (Continued)

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm^2/sec)	$D_R \times 10^{-5}$ (cm^2/sec)	$D_R' \times 10^{-5}$ (cm^2/sec)	$D_{MR} \times 10^{-5}$ (cm^2/sec)	$D_{MR}' \times 10^{-5}$ (cm^2/sec)	$D_{SS} \times 10^{-5}$ (cm^2/sec)	Researcher(s)
*O-Cu	1300	4.74	29.3	33.1	4.89	5.52	4.71	El-Naggar & Parlee (36)
*		12.5						Gerlach et al. (52)
		11.1						Oberg et al. (85)
*		12.2						Osterwald & Schwarzlose (87)
		12.7						Otsuka & Kozuka (91)
*		1.08						Shurygin & Kryuk (110)
*O-Fe	1600	34.2	16.8	18.8	2.80	3.13	2.69	Ershov & Bychev (41)
*		13.3						Novokhatskii & Ershov (82)
*		2.96						Suzuki & Mori (119)
*O-Pb	900	1.36	41.3	49.5	6.88	8.24	6.74	Arcella (4)
*		1.23						Honma et al. (60)
		22.2						Ostewald & Charle (86)
		20.0						Otsuka & Kozuka (90)
		10.1						Szwarc et al. (123)
*O-Pb	1000	1.45	48.3	58.2	8.04	9.71	7.87	Honma et al. (60)
		26.3						Ostewald & Charle (86)
		23.5						Otsuka & Kozuka (90)
		12.4						Szwarc et al. (123)
O-Sn	800	5.40	53.9	61.3	8.98	10.2	8.28	Otsuka & Kozuka (92)
		5.16						Ramanaryanan & Rapp (101)
O-Sn	1000	8.20	69.5	80.4	11.6	13.4	10.7	Ramanaryanan & Rapp (101)

Hydrogen-Liquid Metal Systems--Except for H-Li, D_{EX} values are very much larger than D_{ART} values.

Nitrogen-Liquid Metal Systems--For N-Fe, D_{EX} values are about one third as large as D_R and D_R' values. D_{EX} values are about twice as large as D_{MR}, D_{MR}', and D_{SS} values.

Oxygen-Liquid Metal Systems--For O-Ag, D_{EX} values are slightly larger than D_R and D_R' values. For O-Cu, D_{EX} values are about twice as large as D_{MR}, D_{MR}', and D_{SS} values. For O-Sn, D_{EX} values are slightly over half as large as D_{MR}, D_{MR}', and D_{SS} values.

3.4. Comparison of Experimental Data With Predictions From Vibrational Displacement Theory

Table 4 compares experimental data with predictions from vibrational displacement theory. In this table, D_{EX} is the experimental value (from Equation (34)) and D_V is given by Equation (24). Debye temperatures and force contants were taken from the literature (Gschneidner, 1964; Waser and Pauling, 1950).

Hydrogen-Liquid Metal Systems--Except for H-Li, D_{EX} values are very much larger than D_V values.

Nitrogen Liquid Metal Systems--For N-Fe, D_{EX} values are about twice as large as D_V values.

Oxygen-Liquid Metal Systems--For all systems, D_{EX} values are much larger than D_V values.

TABLE 4

Comparison of experimental data with the vibrational displacement theory.
An asterisk denotes questionable data.
Number in parentheses indicates reference.

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm^2/sec)	$D_V \times 10^{-5}$ (cm^2/sec)	Researcher(s)
H-Cu	1200	314	3.55	Chernega & Vashchenko (21)
		524		Sacris & Parlee (106)
*		68.7		Sigrist et al. (111)
H-Cu	1300	549	3.80	Sacirs & Parlee (106)
*		78.0		Sigrist et al. (111)
*H-Fe	1600	355	2.78	Arkharov et al. (6)
		144		Depuydt & Parlee (27)
		132		El-Tayeb & Parlee (37)
*		15.0		Nyquist (83)
		85.4		Solar & Guthrie (112)
*H-Li	700	3.15	21.1	Alire (3)
H-Li	800	10.5	23.2	Alire (3)
N-Fe	1600	5.57	2.78	Arkharov et al. (5)
		5.66		Bogdanov et al. (14)
*		6.73		Chesnokov et al. (23)
		5.57		Ershov & Kovalenko (43)
*		3.67		Lee & Parlee (74)
N-Fe	1700	6.47	2.93	Arkharov et al. (5)
		6.90		Bogdanov et al. (14)
		6.47		Ershov & Kovalenko (43)
N-Ni	1500	1020	3.59	Chesnokov & Linchevskii (22)
N-Ni	1600	1127	3.80	Chesnokov & Linchevskii (22)
O-Ag	1000	10.3	2.66	Masson & Whiteway (77)
		8.88		Mizikar et al. (79)
		10.5		Oberg et al. (85)
		9.54		Otsuka et al. (88)
		8.88		Parlee & Zeibel (94)
*		14.7		Rickert & El-Miligy (105)
		9.62		Sano et al. (107)
		9.68		Shah & Parlee (109)

TABLE 4 (Continued)

System	t(°C)	D_{EX} x 10_{-5} (cm^2/sec)	D_V x 10_{-5} (cm^2/sec)	Researcher(s)
O-Ag	1100	13.8	2.87	Masson & Whiteway (77)
		10.9		Mizikar et al. (79)
		13.7		Oberg et al. (85)
		11.8		Otsuka et al. (88)
		10.9		Parlee & Zeibel (94)
*		18.1		Rickert & El-Miligy (105)
		12.4		Sano et al. (107)
		12.2		Shah & Parlee (109)
O-Ag	1200	17.5	3.08	Masson & Whiteway (77)
		13.0		Mizikar et al. (79)
		16.4		Oberg et al. (85)
		13.0		Parlee & Zeibel (94)
*		21.7		Rickert & El-Miligy (105)
		15.4		Sano et al. (107)
		14.8		Shah & Parlee (109)
*O-Cu	1100	2.28	3.31	El-Naggar & Parlee (36)
*		5.73		Gerlach et al. (52)
		6.10		Oberg et al. (85)
*		8.48		Osterwald & Schwarzlose (87)
*		6.22		Rickert & El-Miligy (105)
*		0.73		Shurygin & Kryuk (110
*O-Cu	1200	3.37	3.55	El-Naggar & Parlee (36)
*		8.68		Gerlach et al. (52)
		8.41		Oberg et al. (85)
*		10.7		Osterwald & Schwarzlose (87)
		9.78		Otsuka & Kozuka (91)
*		8.91		Rickert & El-Miligy (105)
*		0.90		Shurygin & Kryuk (110)
*O-Cu	1300	4.74	3.80	El-Naggar & Parlee (36)
*		12.5		Gerlach et al. (52)
		11.1		Oberg et al. (85)
*		12.2		Osterwald & Schwarzlose (87)
		12.7		Otsuka & Kozuka (91)
*		1.08		Shurygin & Kryuk (110)
*O-Fe	1600	34.2	2.78	Ershov & Bychev (41)
*		13.3		Novokhatskii & Ershov (82)
*		2.96		Suzuki & Mori (119)

TABLE 4 (Continued)

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm²/sec)	$D_V \times 10^{-5}$ (cm²/sec)	Researcher(s)
*O-Pb	900	1.36	1.91	Arcella (4)
*		1.23		Honma et al. (60)
		22.2		Osterwald & Charle (86)
		20.0		Otsuka & Kozuka (90)
		10.0		Szwarc et al. (123)
*O-Pb	1000	1.45	2.08	Honma et al. (60)
		26.3		Osterwald & Charle (86)
		23.5		Otsuka & Kozuka (90)
		12.4		Szwarc et al. (123)
O-Sn	800	7.40	1.24	Otsuka & Kozuka (92)
		5.16		Ramanaryanan & Rapp (101)
O-Sn	1000	8.20	1.47	Ramanaryanan & Rapp (101)

3.5. Comparison of Experimental Data With Predictions From the Modified Enskog Theory

Table 5 compares experimental data with predictions from the modified Enskog theory. In this table, D_{EX} is the experimental value (from Equation (34)) and D_{MET} is given by Equation (31).

Hydrogen-Liquid Metal Systems--For H-Fe, D_{EX} values are slightly larger than D_{MET} values. For H-Ni, D_{EX} values are slightly smaller than D_{MET} values.

Nitrogen-Liquid Metal Systems--For N-Fe, D_{EX} values are about one-half as large as D_{MET} values.

Oxygen-Liquid Metal Systems--For O-Ag, D_{EX} values are slightly larger than D_{MET} values. For O-Cu, D_{EX} values are slightly smaller than D_{MET} values.

3.6. Conclusions Regarding the Overall Applicability of the Four Selected Theories

Hydrogen-Liquid Metal Systems--The modified Enskog theory is good for H-Fe and H-Ni (within a factor of two) and poor for the other systems (different by a factor of two or more). The other three theories (and modifications) are poor for all systems. There are two posible reasons for the failure of all these theories. First, quantum effects of hydrogen diffusion in liquids are not accounted for in any of these theories (Nakanishi et al., 1965; Ferrell and Himmelblau, 1967; Sridhar and Potter, 1977a). Second, delocalization of hydrogen particles in liquid metlas results in a large mobility for hydrogen (Oates et al., 1978). Both possiblities increase the diffusion coefficient. Examination of the experimental data indicates that a quantum

TABLE 5

Comparison of experimental data with the modified Enskog theory.
An asterisk denotes questionable data.
Number in parentheses indicates reference.

System	t(°C)	D_{EX} x 10^{-5} (cm^2/sec)	D_{MET} x 10^{-5} (cm^2/sec)	Researcher(s)
H-Al	800	436	87	Eichenauer & Markopoulos (33)
*		0.21		Vashchenko et al. (124)
*H-Al	900	2519	100	Byalik et al. (18)
		528		Eichenauer & Markopoulos (33)
*		0.38		Vashchenko et al. (124)
H-Cu	1200	314	81	Chernega & Vashchenko (21)
		524		Sacris & Parlee (106)
*		68.7		Sigrist et al. (111)
H-Cu	1300	549	89	Sacris & Parlee (106)
*		78.0		Sigrist et al. (111)
*H-Fe	1600	355	83	Arkharov et al. (6)
		144		Depuydt & Parlee (27)
		132		El-Tayeb & Parlee (37)
*		15.0		Nyquist (83)
		85.4		Solar & Guthrie (112)
H-Fe	1700	138	96	El-Tayeb & Parlee (37)
		90.3		Solar & Guthrie (112)
H-Ni	1500	66.0	80	Sacris & Parlee (106)
*		315		Wright & Hocking (130
H-Ni	1600	75.1	97	Sacirs & Parlee (106)
*		365		Wright & Hocking (130)
N-Fe	1600	5.57	11.5	Arkharov et al. (5)
		5.66		Bogdanov et a. (14)
*		6.73		Chesnokov et al. (23)
		5.57	11.5	Ershov & Kovalenko (43)
*		3.67		Lee & Parlee (74)
N-Fe	1700	6.47	13.5	Arkharov et al. (5)
		6.90		Bogdanov et al. (14)
		6.47		Ershov & Kovalenko (43)

TABLE 5 (Continued)

System	t(°C)	D_{EX} x 10_{-5} (cm^2/sec)	D_{MET} x 10^{-5} (cm^2/sec)	Researcher(s)
O-Ag	1000	10.3	8.0	Masson & Whiteway (77)
		8.88		Mizikar et al. (79)
		10.5		Oberg et al. (85)
		9.54		Otsuka et al. (88)
		8.88		Parlee & Zeibel (94)
*		14.7		Rickert & El-Miligy (105)
		9.62		Sano et al. (107)
		9.68		Shah & Parlee (109)
O-Ag	1100	13.8	10.8	Masson & Whiteway (77)
		10.9		Mizikar et al. (79)
		13.7		Oberg et al. (85)
		11.8		Otsuka et al. (88)
		10.9		Parlee & Zeibel (94)
*		18.1		Rickert & El-Miligy (105)
		12.4		Sano et al. (107)
		12.2		Shah & Parlee (109)
O-Ag	1200	17.5	12.3	Masson & Whiteway (77)
		13.0		Mizikar et al. (79)
		16.4		Oberg et al. (85)
		13.0		Parlee & Zeibel (94)
*		21.7		Rickert & El-Miligy (105)
		15.4		Sano et al. (107)
		14.8		Shah & Parlee (109)
*O-Cu	1100	2.28	9.50	El-Naggar & Parlee (36)
*		5.73		Gerlach et al. (52)
		6.10		Oberg et al. (85)
*		8.40		Osterwald & Schwarzlose (87)
*		6.22		Rickert & El-Miligy (105)
*		0.73		Shurygin & Kryuk (110)
*O-Cu	1200	3.37	11.2	El-Naggar & Parlee (36)
*		8.68		Gerlach et al. (52)
		8.41		Oberg et al. (85)
*		10.7		Osterwald & Schwarzlose (87)
		9.78		Otsuka & Kozula (91)
*		8.91		Rickert & El-Miligy (105)
*		0.90		Shurygin & Kryuk (110)

TABLE 5 (Continued)

System	t(°C)	$D_{EX} \times 10^{-5}$ (cm²/sec)	$D_{MET} \times 10^{-5}$ (cm²/sec)	Researcher(s)
*O-Cu	1300	4.74	12.9	El-Naggar & Parlee (36)
*		12.5		Gerlach et al. (52)
		11.1		Oberg et al. (85)
*		12.2		Osterwald & Schawzlose (87)
		12.7		Otsuka & Kozuka (91)
*		1.08		Shurygin & Kryuk (110)
*O-Fe	1600	34.2	11.8	Ershov & Bychev (41)
*		13.3		Novokhatskii & Ershov (82)
*		2.96		Suzuki & Mori (119)
O-Ga	800	7.26	40.0	Klinedinst & Stevenson (65)
O-Ga	900	10.1	46.7	Klinedinst & Stevenson (65)

correction of the modified Enskog theory may bring its predictions in line with experimental data.

Figure 5 shows hydrogen diameter (d_H) in liquid metals versus differences in Pauling electronegativity values (ΔE). The covalent diameter of hydrogen is shown for comparison. The hydrogen diameter, r_C in Equation (8), is calculated so that Equation (31) "predicts" a correct diffusion coefficient. It is expected that d_H should increase as ΔE increases because particles become larger as they acquire more negative charge. While there appears to be some correlation, this expectation fails dramatically in one central datum: hydrogen in aluminum. Thus, in hydrogen-liquid metal systems, no diffusion coefficient correlations can be recommended involving Pauling electronegativity differences.

<u>Nitrogen-Liquid Metal Systems</u>--The modified hydrodynamical theory (Equation (7)) is excellent for N-Fe (nearly idenical with experimental values). Modified absolute reaction-rate theory (Equations (15), (16), and (18)), vibrational displacement theory, and the modified Enskog theory are good for N-Fe. All theories are poor for N-Ni.

<u>Oxygen-Liquid Metal Systems</u>--The modified hydrodynamical theory is excellent for O-Cu and O-Pb, and good for O-Ag. The modified Enskog theory is excellent for O-Ag and O-Cu. The absolute reaction-rate theory (Equations (14) and (17)) is good for O-Ag, and modified absolute reaction-rate theory is good for O-Sn. The vibrational displacement theory is poor for all systems.

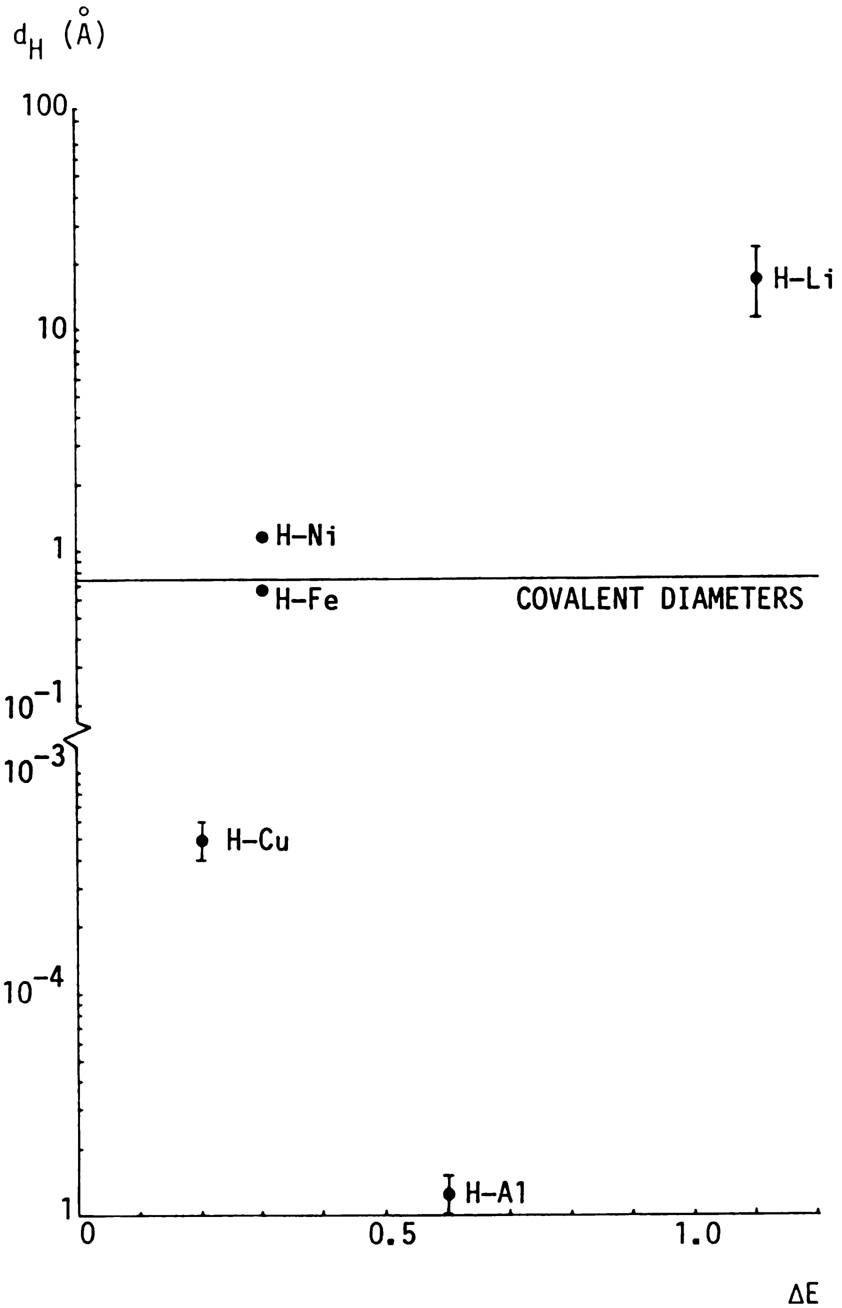

Figure 5. Hydrogen diameter versus differences in Pauling electronegativity values.

None of the four theories (and modifications) is applicable to all the systems examined, expecially the hydrogen-liquid metal systems. The modified hydrodynamical theory and the modified Enskog theory are good to excellent for the majority of the systems examined (excluding hydrogen). This is illustrated in Figures 6 and 7, which compare the modified hydrodynamical theory and the modified Enskog theory with the combined experimental data of two independent studies. The systems from which experimental data was taken were chosen on the basis of the following criteria:

1. At least five independent investigations done on the system.
2. Credibility of the studies done on the system (e.g., problems in performing the experiments).
3. Consistency of the studies done on the system.
4. Temperature range of at least 150°C.
5. Diffusion coefficient determined at no less than ten temperatures in the interval.
6. Availability of actual experimental values in the paper describing the study.

From Tables 2-5 and Figure 6 and 7, the modified hydrodynamical theory or the modified Enskog theory can be used to predict diffusion coefficients of nitrogen in liquid iron and oxygen in liquid metals.

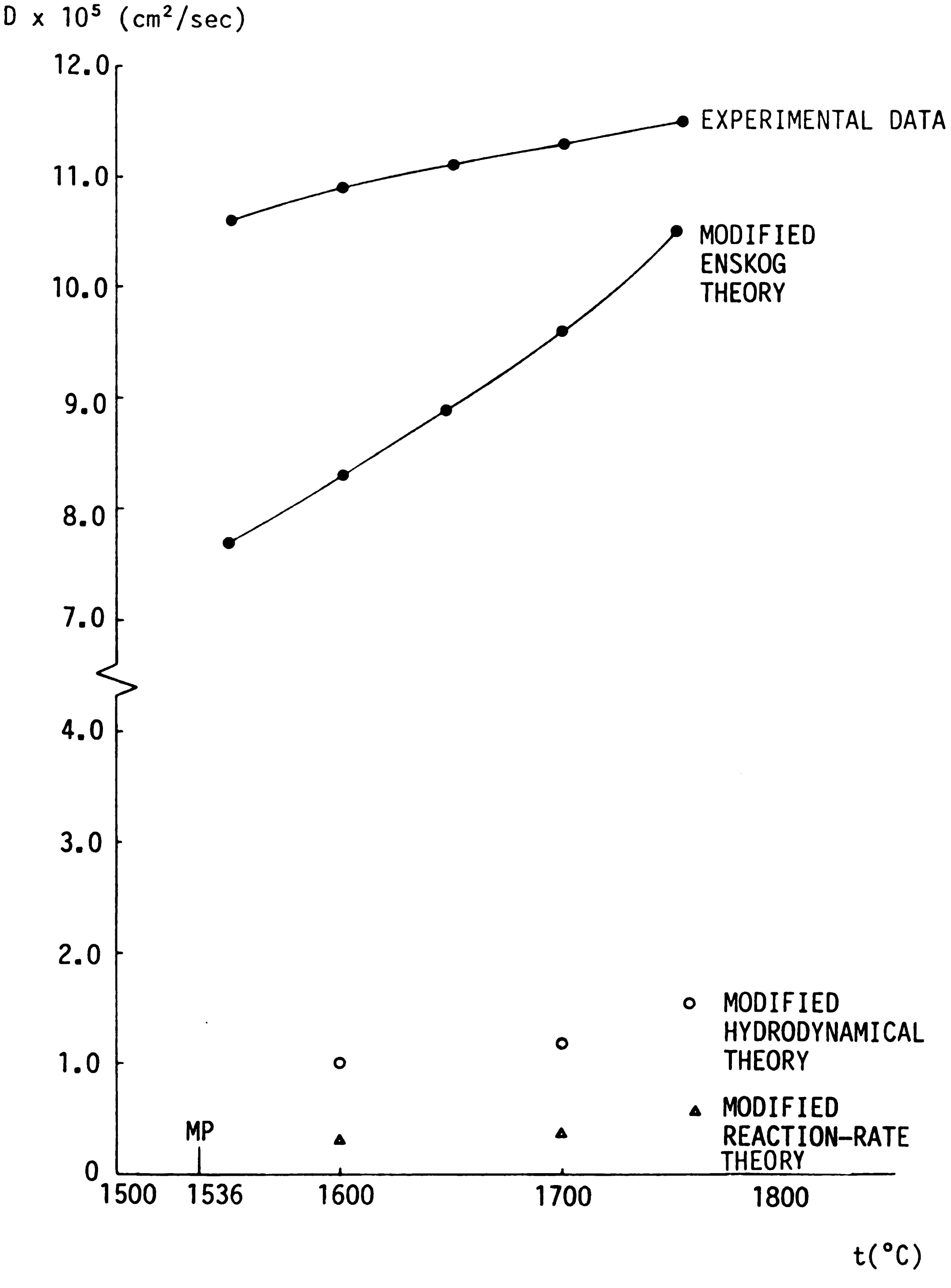

Figure 6. The H-Fe system.

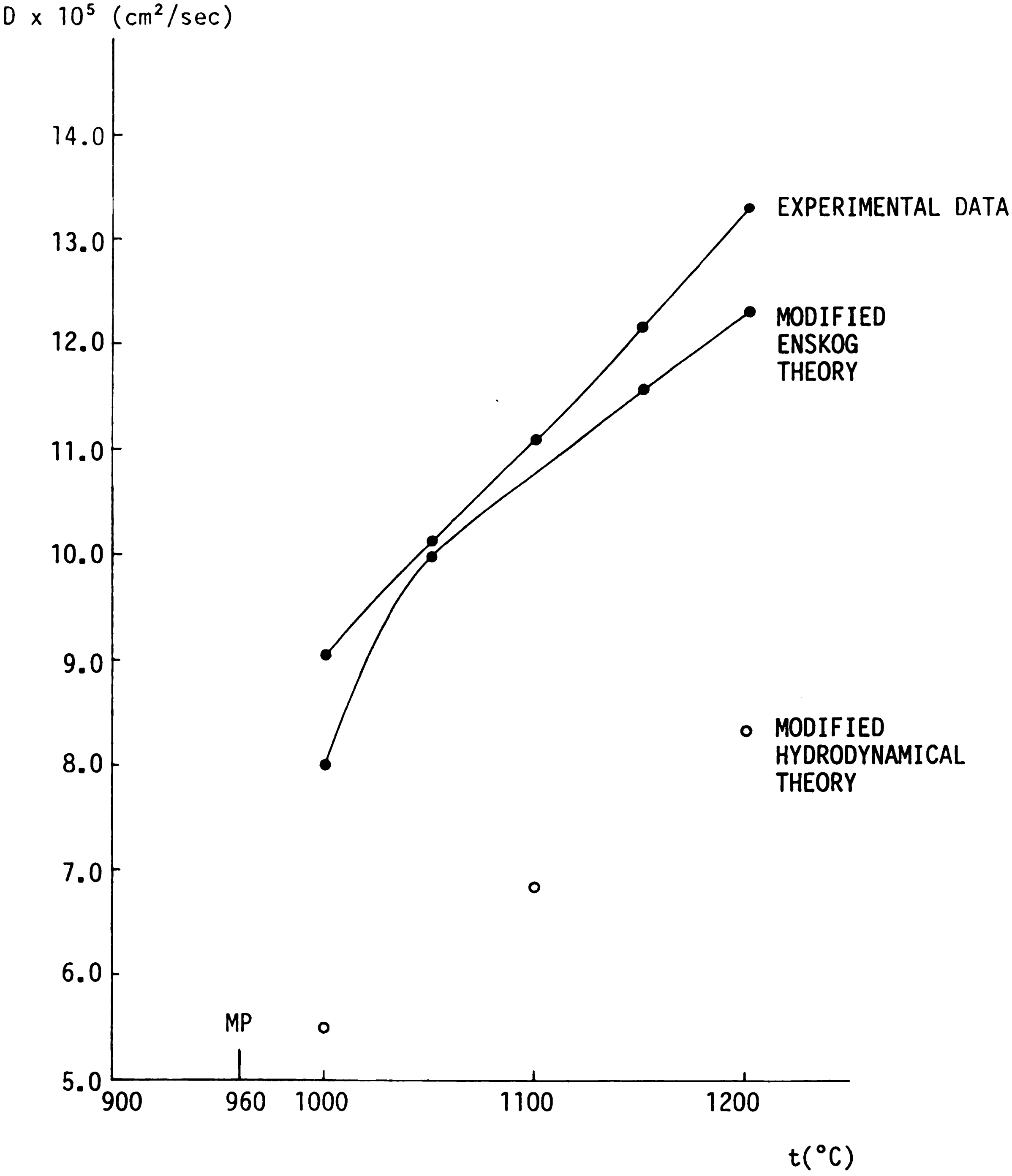

Figure 7. The O-Ag system.

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