THE UNIVERSITY OF OKLAHOMA

GRADUATE COLLEGE

SELF-DIFFUSION PHENOMENA IN CADMIUM AND ZINC AMALGAMS

A DISSERTATION

SUBMITTED TO THE GRADUATE FACULTY

in partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY

BY

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Norman, Oklahoma

1962

SELF-DIFFUSION PHENOMENA IN

CADMIUM AND ZINC AMALGAMS

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ABSTRACT

This dissertation presents the results of a theoretical and experimental investigation of diffusion phenomena in liquid metals. The relationship between diffusion and viscosity is of primary interest in this study.

The available theoretical models for liquid diffusion are critically reviewed. A new theoretical model is developed in which the activation energy for diffusion is precisely that for viscosity. This new model accurately predicts the self-diffusion behavior in seven liquid metals for which data are available. Further, it is shown that self-diffusion in pure liquid metals can be theoretically correlated to a constant geometrical parameter which is equal to the ratio of the radius of the diffusing particle to the interatomic spacing.

Densities, concentration cell potentials, and solute self-diffusion coefficients of binary mercury-rich amalgams of cadmium and zinc were measured over a range of temperatures from 50°C to 150°C. Compositions studied varied from 0.089 to 7.38 atomic percent cadmium and from 0.069 to 5.61 atomic percent zinc.

The new theoretical model adequately describes the diffusion behavior in these binary amalgams. The change in the geometrical parameter for the binary amalgams studied is in agreement with other data regarding the non-ideal behavior of these systems.

ACKNOWLEDGMENT

It is a pleasure for the author to express his sincere appreciation to the many individuals who have assisted in the laboratory work and in the preparation of this dissertation. Of these, the following persons deserve special mention for their assistance:

To Dr. William R. Upthegrove, Associate Professor of Metallurgical Engineering, for his direction of this research and the continual encouragement he offered.

To the Doctoral Advisory Committee who were, in addition to Dr. Upthegrove as chairman; Dr. Andrew Cosgarea, Jr., Associate Professor of Chemical Engineering and of Metallurgical Engineering; Mr. Earl LaFon, Associate Professor of Mathematics; Dr. George W. Murphy, Professor of Chemistry; Dr. Robert H. Perry, Associate Professor of Chemical Engineering; Dr. John E. Powers, Professor of Chemical Engineering; and Dr. William J. Viavant, Associate Professor of Computer Science. Each of these gentlemen has given unstintingly of his time and consideration to the author and this work.

To Messrs. James Hood, Robert Lawrence, and Norman Alexandre of the Physics Department Shops for their assistance in fabricating certain pieces of equipment and apparatus.

To the undergraduate students who served as laboratory assistants. In particular, the assistance rendered by Mr. Charles B. Childs and Mr. Brian D. Howard was outstanding.

To the author's fellow graduate students, Mr. William M. Lorkovic for his assistance in some phases of the preparation of this dissertation, and to Mr. Tom Wood who assisted with the computer evaluation of the mathematical analysis given in Appendix E. Also, the cooperation of Messrs. John Stark, Harvey Doering, and Morteza Mirshamsi who shared laboratory space with the author is to be acknowledged.

Finally, to the author's wife, Nancy, who has assisted in the preparation of this dissertaion and whose continual cooperation made possible the completion of this research program.

In conclusion, the author desires to acknowledge the financial support given by the Ohio Oil Company by means of a fellowship during the first year of this research, and to the United States Atomic Energy Commission which, under contract AT-(40-1)-2409, has sponsored the major portion of this research.

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CHAPTER I

INTRODUCTION

The objectives of this program of research represent an attempt to permit improved understanding of diffusion phenomena in liquid metals and the effect of associated system parameters upon these phenomena. One common aspect of most liquid metals research has been the attempt to correlate viscosity and diffusion data. There are several theoretical equations relating these variables, such as those of Eyring and Stokes-Einstein, which are customarily used in such analyses of diffusion. The success of these and other equations in accurately describing diffusion behavior could only be termed moderate. This is particularly the case when the same equation is used to predict the diffusion coefficients for all liquid metals for which diffusion data are available.

New theoretical ideas and interpretations of diffusion in liquids have been developed in this research and are presented in Chapter II. The success of this theoretical work in accurately describing pure component self-diffusion in

seven liquid metals has been most encouraging. These tests of the equation were made using self-diffusion data from the literature.

The prediction of diffusion behavior in solutions provides a more stringent test of theoretical equations than does pure component self-diffusion. Therefore, one of the major objectives in this research has been the measurement of self-diffusion coefficients in the cadmium-mercury and zinc-mercury systems as a function of concentration and temperature. Density and thermodynamic activity measurements were also made since these quantities are required in the theoretical analysis.

The cadmium and zinc amalgam systems were chosen for this study because certain binary amalgams of cadmium and of zinc comprise isoviscous solution pairs as reported by Golik and Karlikov (1). Isoviscous solutions are defined as possessing coincident kinematic viscosity-temperature curves. Golik and Karlikov also found that examination of the liquid alloy structures by X-ray diffraction showed coincident maxima of the radial distribution functions for these isoviscous solutions. Because the kinematic viscosity is the diffusivity for momentum transfer, the correlation of the kinematic viscosity behavior with the interatomic spacing as characterized by the radial distribution functions appears to be quite reasonable. Thus, in addition to studying the influence of viscosity upon diffusion phenomena, the choice of these amalgam systems provides opportunity to evaluate the relative

influence of structural effects to some extent under condition of equal kinematic viscosity.

CHAPTER II

THEORY OF LIQUID DIFFUSION

The understanding of diffusion phenomena in liquid metals is inferior as compared with either gaseous or solid state diffusion. The lack of accurate comprehension of these and other related transport phenomena in liquids is the consequence of a less complete knowledge of the liquid state. The situation regarding diffusion processes in liquids is further complicated by the experimental difficulties encountered in attempting to test the various theories proposed to describe diffusion and the effect of relevant system variables upon these phenomena.

Adolph Fick (2) first proposed a phenomenological model for diffusion in which the flux of the diffusing species is proportional to the concentration gradient of the species as is indicated by

$$J = - D \frac{\partial C}{\partial X} . \qquad (1)$$

In equation 1, known as Fick's first law, D is the diffusion coefficient and the gradient is measured normal to the unit area of the flux J.

Consideration of the accumulation rate of the diffusing species within an element of volume and use of equation 1 gives Fick's second law,

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial X^2} , \qquad (2)$$

assuming uniaxial diffusion in a homogeneous isotropic medium. It is further assumed that the diffusion coefficient is independent of the concentration of the diffusing substance. Employing vector notation, equation 2 for the three-dimensional case is written as

$$\frac{\partial C}{\partial t} = D \text{ div grad } C . \qquad (3)$$

If D cannot be considered as concentration independent, then D must be also treated as a variable, and instead of equation 3, the rate of accumulation is represented as

$$\frac{\partial C}{\partial t} = \text{div } (D \text{ grad } C) . \qquad (4)$$

A large number of experimental techniques for measuring diffusion coefficients have been based on the solutions to Fick's second law when applied to various diffusion geometries with the assumption of appropriate boundary and initial conditions. Such solutions have been described by several authors (3,4) and for the most part are intended to describe one-dimensional binary diffusion assuming the lack of concentration dependence by the diffusion coefficient and neglecting volume changes due to mixing.

Mechanistic Models

One of the best known equations relating diffusion and viscosity is that of Stokes-Einstein (5),

$$D = \frac{kT}{6\pi r\mu} \tag{5}$$

where

D = the self-diffusion coefficient,

k = Boltzmann's constant,

T = the absolute temperature,

r = radius of the diffusing particle, and

μ = the viscosity.

Stokes' law, which predicts the terminal velocity of relatively large, unattracting, hard, dense spheres through a liquid, is used in the derivation of equation 5 and is expressed as

$$F = 3\pi a\mu v_t. \tag{6}$$

F = the force of the sphere,

a = the sphere diameter, and

v_t = the terminal velocity.

Einstein's equation, expressing the self-diffusion coefficient as a function of the mobility, is

$$D = kTM \tag{7}$$

where the mobility, M, is the average velocity of the diffusing particle per unit force acting on that particle. To obtain the Stokes-Einstein equation, the mobility is determined from Stokes' law as v_t/F and the resulting expression is introduced into equation 7.

Although the Stokes-Einstein equation is derived on the assumption of large solute particles diffusing through a continuous medium, the radii of some liquid metal atoms calculated from this equation show comparatively close agreement with the values of crystallographic ionic radii. In view of the inconsistency between the Stokes-Einstein model and the supposed structure of liquid metals, this agreement is frequently described as "merely fortuitous."

The agreement between the crystallographic radii and those calculated using the Stokes-Einstein equation should probably not be accepted as a genuine verification of the supposition that the equation is applicable in describing diffusion except in an empirical manner. There are several reasons for this conclusion. First, the crystallographic radii are very much dependent upon the rather arbitrary assignment of a radius to one element either from experimental data, as for the Goldschmidt radii (6), or from theory in the case of the Pauling radii (7). In either case, the calculated radii are based on the assumed additive nature of ionic radii to give interionic distances in halide and oxide compounds. Secondly, examination of the radial wave functions from quantum mechanics indicates that no absolute significance should be given to the concept of ionic radii, since these probability functions tail off to zero for an infinite radial distance. In the third place, the significance of the agreement between the Stokes-Einstein radii and crystallographic radii, if it should be meaningful, would presume that the solid-state ionic

radii are equal to those in the liquid metals. Furthermore, the presumption would be that the diffusing atoms would be ions of a particular valence (the one which agrees most closely with the Stokes-Einstein radii). It may be that the atoms in liquid metals tend to resonate between various possible valence states, and a radius calculated from diffusion data would indicate an effective ionic dimension. At best, the concept of ionic radii gives a convenient and approximate indication of the atomic dimensions.

The fact that the Stokes-Einstein equation does predict approximately the diffusion behavior in liquid metals can be explained only by assuming that Stokes' law gives about the correct value of the mobility. Since the mobility is a ratio of the average velocity (taken to be the terminal value) to the force acting on the particle, both the velocity and the force could be subject to compensating errors and give an approximate value for the mobility. Lapple (8), in a discussion of particle dynamics, indicates that Stokes' law is subject to a lower limit where the Brownian motion of the particle begins to be effective. This effect would give an erroneous velocity as calculated from Stokes' law for a specified force.

In summary to this discussion of the Stokes-Einstein equation, it seems "merely fortuitous" that Stokes law predicts about the correct value of the mobility and, consequently,

the Stokes-Einstein equation describes approximately the diffusion behavior in liquid metals.

Sutherland (9) presented an empirical correction to the Stokes-Einstein equation in 1905 as an attempt to account for the size difference between the diffusing particles and those of the bulk liquid. In the extrapolation of diffusion data to determine the molecular mass of albumin, Sutherland found modification of the Stokes-Einstein model necessary in order to more accurately fit the available data. The empirically modified form of Stokes' law was given as

$$F = 3\pi a\mu v_t \left[\frac{\beta a + 2\mu}{\beta a + 3\mu} \right], \tag{8}$$

where β is the coefficient of sliding friction between the diffusing particle and the solution. It was postulated that no slipping should occur at the surface of a relatively large diffusing particle in a continuum; hence, β would become infinite in magnitude and the empirical correction term would be unity. This case would give the familiar form of Stokes' law. In the instance of diffusion in a particulate fluid, all particles being of a similar size, it was assumed that the diffusing sphere would move essentially in voids present in the array of solvent particles and, therefore, β would become zero. Under this condition equation 8 would become

$$F = 2\pi a\mu v_t \, . \tag{9}$$

Solving equation 8 for the mobility and substituting in equation 7 (Einstein's relation between the diffusion coefficient and mobility) gives

$$D = \frac{kT}{6\pi r\mu}\left[\frac{\beta a + 3\mu}{\beta a + 2\mu}\right] \qquad (10)$$

which is known as the Sutherland (or Sutherland-Einstein) equation. For liquid metals diffusion it is more likely that the conditions imposed on the model to give equation 9 should apply, and equation 10 becomes

$$D = \frac{kT}{4\pi r\mu} . \qquad (11)$$

Equation 11 fits some liquid metals diffusion data quite well, particularly if one chooses to use the Pauling univalent ionic radii for the appropriate term in the equation.

The Sutherland equation, being an empirical improvement of the Stokes-Einstein equation, is subject in the strictest sense to the same anomolous conditions as the original Stokes-Einstein equation. While the empirical modification seems to require that the diffusing and solution atoms be of the same size to give equation 11, this is not necessarily true. Lamb (10) has mathematically delineated the effect of slip at the surface of a sphere which moves in accordance with Stokes' law and gives the expression

$$F = 6\pi r\mu v_t\left[\frac{\beta r + 2\mu}{\beta r + 3\mu}\right] , \qquad (12)$$

which is equivalent to equation 8. This development imposes no size restrictions on the sphere except that it must be of sufficient size to obey Stokes' law. Thus, the assumption that similarly sized particles comprise the diffusion system for equation 11 is merely an attempt to define a physical

situation which is conducive to the occurrence of slip at the surface of the diffusing particle. This assumption tacitly implies that Stokes' law would be valid for a particulate system, and this conclusion would seem to be rather dubious.

Eyring (11) has presented a model for liquid diffusion based upon his theory of absolute reaction rates and using the concept that the liquid structure contains a number of holes or void spaces. The equation resulting from the model, which describes diffusion as an activated process, is usually given as

$$D = \frac{kT}{2r\mu} \quad . \tag{13}$$

The development of this equation is given in detail in the above mentioned reference and, therefore, will not be repeated here. It should be noted, however, that for liquid metals equation 13 gives about an order of magnitude poorer agreement with crystallographic radii than does the Stokes-Einstein equation.

More recently, the activation model for diffusion has been modified (12) to give

$$D = \frac{kT}{\xi\lambda\mu} = \frac{kT}{12r\mu} \quad . \tag{14}$$

The term ξ appearing in equation 14 is the number of nearest neighbors lying in the same plane as the diffusing atom and is equal to six with the assumption of a hexagonal or cubical close-packed liquid structure.

Figure 1 shows the structural model and mechanism of diffusion for the modification to Eyring's original equation. At the extreme left in Figure 1, the diffusing atom is shown in the center of the ring of its six nearest neighbors lying in the same plane. The diffusing atom is subsequently shown in the figure as it diffuses to the right. It is postulated that the diffusing atom shears independently with each neighbor and that simultaneous shearing with two or more atoms is not permitted since this would require additional activation energy. Further, it is assumed that the diffusing atom advances the distance between adjacent equilibrium positions, λ_d, every time a nearest neighbor atom jumps backward the distance λ. Thus,

$$\lambda_d = \frac{\lambda}{6} = \frac{\lambda}{\xi} \quad . \tag{15}$$

It appears that λ corresponds to the distance between adjacent lattice positions.

If k' is the frequency of a backward jump of each nearest neighbor atom, then the frequency of relative displacement with respect to all nearest neighbors is

$$k_d = 6k' = \xi k' \quad . \tag{16}$$

The substitution of equations 15 and 16 into the original expression derived by Eyring (11),

$$D = \lambda_d^2 k_d \quad , \tag{17}$$

gives

$$D = \frac{\lambda^2 k'}{\xi} \tag{18}$$

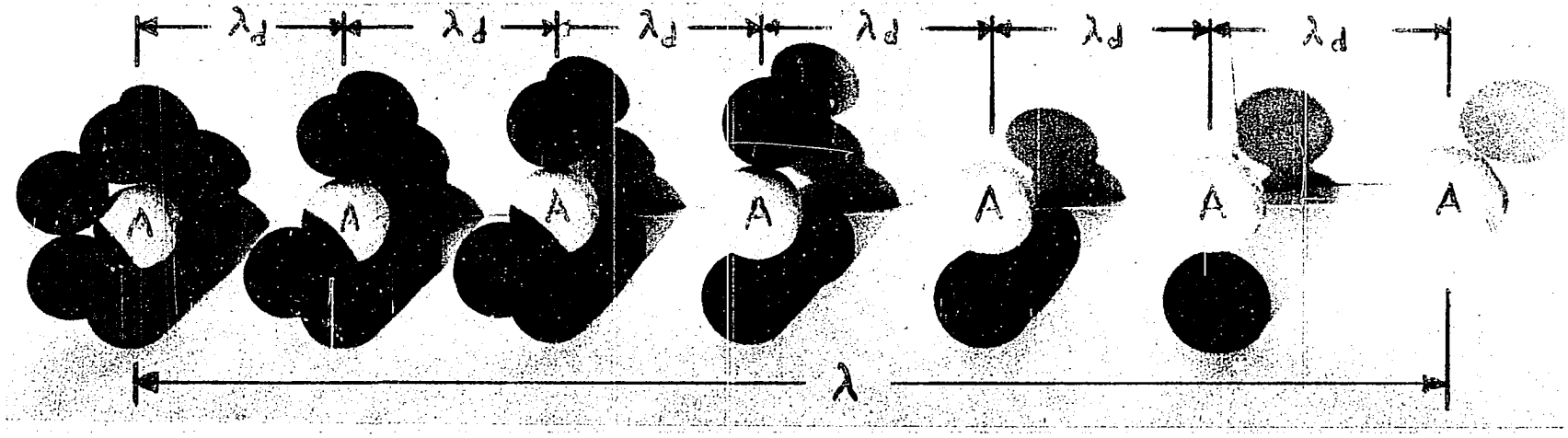


Figure 1. Eyring's Revised Model for Self-Diffusion

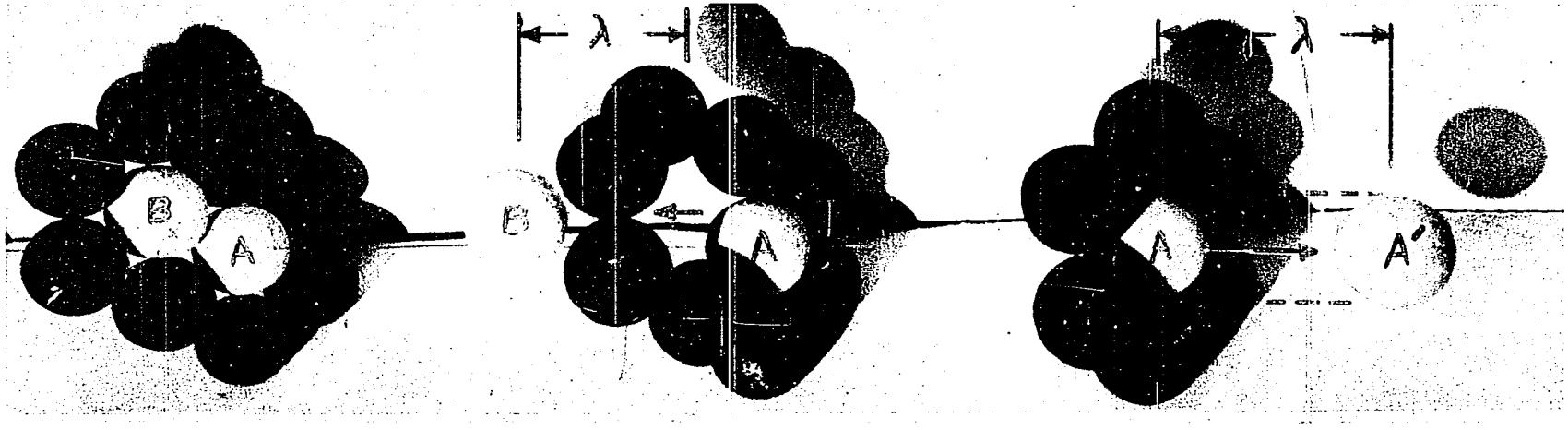


Figure 2. Models for Discussion of Eyring's Equation

The use of Eyring's equation for the viscosity to evaluate the reaction rate constant in equation 18 gives equation 14.

At the extreme left in Figure 2, the same atomic arrangement as in Figure 1 is shown except that additional atoms which surround one of the nearest neighbors to the original diffusing atoms are included. The second item from the left in Figure 2 shows the nearest neighbor atom after it has jumped backward the distance λ, as was described in the above discussion. Now, it seems there is no difference in this nearest neighbor jump and the net displacement of the diffusing atom shown at the right in Figure 2 when the center atom in the plane moves to the position indicated by the single atom. Thus, the modification of Eyring's theory seems to be valid only if the atoms surrounding the nearest neighbor atoms are neglected. If these additional atoms are accounted for, then the backward jump of a single nearest neighbor atom itself constitutes the diffusion process. This conclusion would, therefore, invalidate the premise for equation 14 and substantiate the original equation of Eyring. It should be remarked, however, that equation 14 does agree with the diffusion data for a great number of systems.

In 1959, Swalin (13) derived a theory for diffusion in liquids on the basis of a fluctuation model. The movement of atoms was postulated to result from local density fluctuations in the liquid. Using the Morse function to express the

fluctuation energy, Swalin presented the equation

$$D = 1.29 \times 10^{-8} \, T^2/\Delta H_v \alpha^2, \qquad (19)$$

where

D = the diffusion coefficient in cm^2/sec,

T = the absolute temperature in °K,

ΔH_v = the latent heat of vaporization in k cal/mole, and

α = a parameter related to the curvature of the potential energy versus distance curve, expressed in reciprocal angstroms.

Equation 19, without the numerical evaluation of certain terms, can be expressed as

$$D = 3Z^2 N_o k^2 T^2/96h\Delta H \, \alpha^2, \qquad (20)$$

where

Z = the number of nearest neighbors surrounding the diffusing atom (usually taken to be 10),

N_o = Avogadro's number,

k = Boltzmann's constant, and

h = Planck's constant.

Swalin further states that the evaluation of α is accomplished as

$$\alpha^2 = ZN_o K/4\Delta H_v, \qquad (21)$$

where K is the force constant obtained from the data of Waser and Pauling (14). If one substitutes equation 21 into 20, it is noticed that Swalin's equation reduces to

$$D = \frac{Zk^2T^2}{8hK} \qquad (22)$$

or

$$D = 3.60 \times 10^{-6} T^2/K \qquad (23)$$

where the force constant, K, is expressed in dynes per centimeter.

Swalin's theory predicts no activation energy for liquid diffusion, and it is stated (13) that the only basis for postulating an activated process for diffusion phenomena in liquids is that the Arrhenius equation,

$$D = D_o e^{-Q/RT}, \qquad (24)$$

seems to fit the data over small temperature intervals. In equation 24, D_o is a constant, Q is the activation energy, and R is the gas constant. Diffusion coefficients predicted by equation 19 agree very well with data for mercury and also tin. The agreement of the model with diffusion data in other liquid metals ranges from fair to poor as will be shown later in this chapter. Nevertheless, the work by Swalin gives an interesting approach to the understanding of liquid metals diffusion.

A New Theoretical Model for Liquid Diffusion

The basic scheme of the new theoretical interpretation of liquid diffusion developed in this research program was to evaluate the mobility in terms of a model consistent with the liquid state. Subsequently, Einstein's expression, equation 7, was used to determine the diffusion coefficient as a function of the mobility.

Figure 3 shows the model used in determining the mobility. Atom A is shown as it diffuses to the position indicated by A', the distance traversed being equal to the interatomic spacing, d.

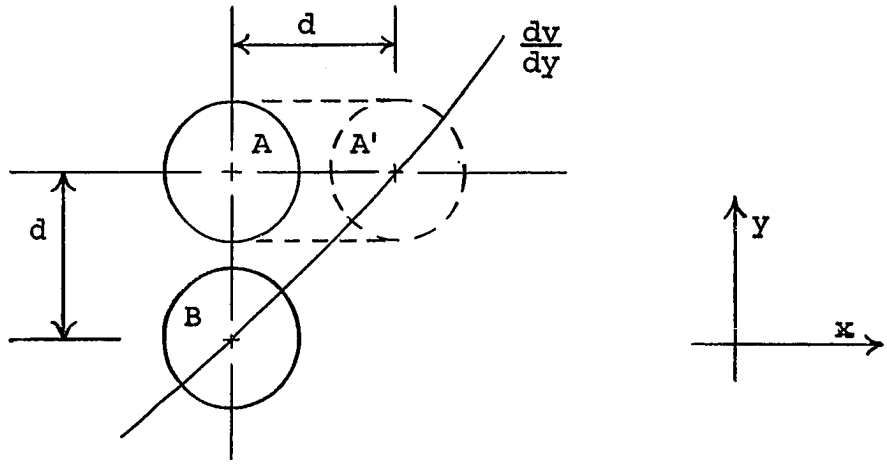


Figure 3. Model For New Diffusion Theory

The relative velocity of atom A with respect to any neighboring atom, B, is the product of the interatomic distance and a velocity gradient, which is shown superimposed upon the atoms. It should also be noticed in Figure 3 that the interatomic distance is indicated to be somewhat greater than the atomic diameter.

The defining equation for the viscosity,

$$\mu = \frac{F}{A} \frac{dy}{dv} , \tag{25}$$

is used in evaluating the mobility of atom A. Equation 25 may be rewritten as

$$F = A\mu \frac{dv}{dy} , \tag{26}$$

where F is the force acting upon the atom over an area A. First, in a general case, the area A is expressed as

$$A = \sigma d \ , \tag{27}$$

which is a definition of σ; that is, σ is the area divided by the interatomic spacing. Since d multiplied by the velocity gradient in equation 26 gives the relative velocity, v_A, of atom A with respect to its neighbors, the mobility is determined as follows:

$$F = \mu\sigma d \frac{dv}{dy} = \mu\sigma v_A \ ,$$

$$M = \frac{v_A}{F} = \frac{1}{\sigma\mu} \ . \tag{28}$$

To specifically evaluate the mobility, the assumption is made that the forces acting upon the atom as it diffuses are effective over the total surface area associated with the volume involved in the diffusion movement from position A to A' in Figure 3. This volume includes the volume of the atom plus the additional volume swept out during the displacement of the atom. In geometrical shape, the volume is then a cylinder of length d with hemispherical ends of radius r, the atomic radius. Thus,

$$A = 2\left[\frac{4\pi r^2}{2}\right] + \int_o^d 2\pi r dx$$

$$= 2\pi r(2r + d) \ . \tag{29}$$

Using equations 27 and 29, the geometrical parameter σ may be evaluated as

$$\sigma = \frac{A}{d} = 2\pi r(2b + 1) \ , \tag{30}$$

where the quantity b is the ratio of the atomic radius to the interatomic distance. Thus, from equations 28 and 30, the mobility is found to be

$$M = \frac{1}{2\pi r(2b + 1)\mu} . \tag{31}$$

Substitution of this expression for the mobility into equation 7 gives the diffusion coefficient as

$$D = \frac{kT}{2\pi r(2b + 1)\mu} . \tag{32}$$

Equation 32 applies to diffusion in single component systems; for the case of self-diffusion in binary systems, equation 32 becomes

$$D = \frac{kT}{2\pi r(2b + 1)\mu} \left[\frac{d\ln a}{d\ln N}\right] , \tag{33}$$

where a is the thermodynamic activity and N is the atomic fraction. The inclusion of the thermodynamic factor in equation 33 results from the same type of derivation employed by other investigators; for example, Eyring (11), Darken (15), and Onsager and Fouss (16). In the general case, equation 7 would contain the thermodynamic factor in order to account for non-ideal behavior of a given diffusion system.

Inspection of equation 32 will indicate that in the limiting case of an unexpanded liquid; that is, where the atomic diameter is equal to the interatomic distance, the equation gives the same geometrical factor of $4\pi r$ as given by the Sutherland empirical modification of the Stokes-Einstein equation.

The theoretical analysis of the diffusion behavior can be pursued beyond equation 33 by the introduction of an expression for the liquid viscosity. In this research, the decision was to use Eyring's model for the viscosity because it accurately predicts this quantity for a great number of liquids. Eyring's equation is (11)

$$\mu = \frac{N_o h}{V} e^{-\frac{\Delta S^*}{R}} e^{\frac{\Delta H^*}{RT}} , \qquad (34)$$

where V is the molar volume, ΔS^* is the activation entropy, and ΔH^* is the activation energy for viscosity. The other terms in the equation, although previously defined, are included in Appendix A for convenient reference. Substitution of equation 34 into 33 gives

$$D = \frac{kTV}{2\pi r(2b + 1)N_o h} e^{\frac{\Delta S^*}{R}} e^{-\frac{\Delta H^*}{RT}} \left[\frac{d\ln a}{d\ln N}\right] . \qquad (35)$$

The difficulties in correctly determining the atomic radius to be used in this equation and others make it desirable to modify this term in the equation if possible. To accomplish this revision of equation 35, the term σ from equation 30 is used instead of the geometrical factor which appears in equation 35. The method is to relate σ to the molar volume, V, and evaluate the proportionality constant involved in terms of the ratio, b, of the atomic radius to the interatomic spacing. It is postulated that the parameter b should be a constant for a given class of liquids, such as the liquid metals group.

The revision of equation 35 is as follows: First, σ is proportional to the radius, and the molal volume is proportional to the radius cubed; thus,

$$V = a_3\sigma^3,$$

or

$$\sigma = \left(\frac{V}{a_3} \right)^{1/3}, \tag{36}$$

where a_3 is a proportionality constant. Substitution of equation 36 into 35 gives

$$D = \frac{kT(a_3)^{1/3}\, V^{2/3}}{N_o h}\, e^{\frac{\Delta S^*}{R}}\, e^{-\frac{\Delta H^*}{RT}} \left[\frac{d\ln a}{d\ln N} \right]. \tag{37}$$

Secondly, the final task in revising equation 35 is to evaluate the constant a_3 only in terms of the parameter b. This is done with recourse to the spherical cage model for the free volume in liquids and using the relation between σ and r, equation 30.

The spherical cage model of the liquid structure, first used by Eyring and Hirschfelder (17), is discussed in some detail by Hildebrand and Scott (18). It is assumed that the center of each molecule moves in a circle whose radius is the difference between the interatomic spacing and the atomic diameter. This model is quite consistent with the model presented in Figure 3, in that the atomic diameter is assumed to be less than the interatomic spacing. The atoms are considered to be hard spheres.

The spherical model gives the free volume per gram molecular weight as

$$V^f = \frac{4\pi N_o}{3} (d - 2r)^3 \quad . \tag{38}$$

The molar volume of the liquid is

$$V = \frac{N_o d^3}{\gamma} \quad , \tag{39}$$

where γ is a constant dependent upon the geometry of the liquid structure. The unexpanded liquid, as previously defined, would have a molar volume, V_o, where

$$V_o = \frac{N_o (2r)^3}{\gamma} \quad . \tag{40}$$

The substitution of equations 39 and 40 into equation 38 gives the result,

$$V^f = \frac{4\pi\gamma}{3} (V^{1/3} - V_o^{1/3})^3 \quad . \tag{41}$$

Equation 41 can be rearranged to give the molar volume in terms of the free volume and the unexpanded liquid volume

$$V = \left[V_o^{1/3} + \left(\frac{3V^f}{4\pi\gamma} \right) 1/3 \right]^3 \quad . \tag{42}$$

By equating the expression for the molar volume from equation 36 and 39,

$$V = \frac{N_o d^3}{\gamma} = a_3 \sigma^3 \quad ,$$

and by substituting for σ from equation 30, the relation

$$\frac{N_o d^3}{\gamma} = a_3 \left[\frac{4\pi r^2}{d} + 2\pi r \right]^3 \tag{43}$$

is obtained. This equation is then solved for a_3 as follows:

$$a_3 = \frac{N_o d^6}{\gamma(4\pi r^2 + 2\pi r d)^3} = \frac{N_o d^6}{\gamma 8\pi^3 r^3 (2r + d)^3}$$

$$= \frac{N_o}{\gamma 8\pi^3 \left[8 \frac{r^6}{d^6} + 12 \frac{r^5}{d^5} + 6 \frac{r^4}{d^4} + \frac{r^3}{d^3} \right]}$$

$$= \frac{N_o}{\gamma 8\pi^3 \frac{r^3}{d^3} \left[2 \frac{r}{d} + 1 \right]^3} \cdot \qquad (44)$$

The constant γ is determined by the average number of nearest neighbors surrounding the atom. In the case of liquids, with a coordination number of 8 to 10, a reasonable choice is

$$\gamma = \frac{4}{3} \cdot \qquad (45)$$

Equation 45 and the definition of the parameter b are used with equation 44 to give the proportionality constant a_3 as a function of b:

$$a_3 = \frac{3N_o}{32\pi^3 b^3 (2b + 1)^3} \cdot \qquad (46)$$

To summarize briefly the results of the new theoretical model for liquid diffusion, the derivation gives the general equation

$$D = \frac{kT}{2\pi r(2b + 1)\mu} \left[\frac{d\ln a}{d\ln N} \right] \cdot \qquad (33)$$

The introduction of Eyring's expression for the viscosity into equation 33 yields

$$D = \frac{kT\,(a_3)^{1/3}\,V^{2/3}}{N_o h}\, e^{\frac{\Delta S^*}{R}}\, e^{-\frac{\Delta H^*}{RT}} \left[\frac{d\ln a}{d\ln N}\right], \qquad (37)$$

where the constant a_3 is determined as a function of the parameter b, equation 46. The term b, equal to the atomic radius divided by the interatomic distance, is postulated to be a constant for a given class of liquids.

Comparison of the New Theory with Diffusion Data from the Literature

Self-diffusion data for seven pure liquid metals were used to test the new theoretical expression, equation 37. These data are the result of research by other investigators and are available in the literature. Furthermore, a comparison was made between the new theory, which indicates that the activation energy for diffusion is precisely the same as for viscosity, and the theory of Swalin which gives no credence to the concept of activation energy.

The first problem in evaluating equation 37 is to determine the quantity b , which is used in equation 46 to specify the value of the constant a_3. Since the parameter b is postulated to be a constant for all the pure liquid metals, the decision was to use the mercury diffusion data of Meyer (19) to evaluate b and then test the other systems using this constant value of b. The mercury data were selected for the determination of b because of the very good precision of these measurements and also the fact that Swalin's equation predicts the diffusion behavior of this system quite well.

The value of b calculated from the mercury data was 0.419, which gave a_3 as 3.997×10^{21} per mole. As a matter of interest, the ratio b was also calculated using Pauling's univalent radius (7) for mercury and interatomic spacing data from Hendus (40). This method of calculation gave b equal to 0.416, which is very good agreement with the value calculated from data. This agreement is probably somewhat fortuitous, however, and is the consequence of having chosen mercury for the calculation. In general, the b values calculated from ionic radii and X-ray diffraction data for most liquid metals will be closer to 0.3 than to 0.4.

Values of ΔS^* and ΔH^* for use in equation 37 were developed from kinematic viscosity data. The kinematic viscosity is equal to the viscosity divided by the density. For purposes of calculation, these data were least square fitted to the expression

$$\nu = Be^{\frac{\Delta H^*}{RT}} \tag{47}$$

where ν is the kinematic viscosity and the constant B is defined using Eyring's equation for viscosity as

$$B = \frac{N_o h}{M} e^{-\frac{\Delta S^*}{R}} . \tag{48}$$

The term M in equation 48 is the molecular weight. The data were compared with values calculated from equation 47 to insure that the functional form of the equation used was reasonable for the data. It should also be mentioned that the use of the conventional Arrhenius correlation for the

absolute viscosity,

$$\mu = Ae^{\frac{E}{RT}}, \qquad (49)$$

will generally give erroneous values of the activation energy for viscosity even though the data will come close to the calculated least square fit line. For example, the use of equation 49 gives the activation energy for mercury equal to 603 calories per mole compared with 559 as calculated from equation 47. This discrepancy arises because the A term in equation 49 is temperature dependent due to a density factor, while the term B in equation 47 is essentially temperature independent. Because the density of many liquid metals systems is approximately a linear function of temperature, the term A in equation 49 appears to be a constant.

The molar volume term in equation 37 was evaluated from density data, and for the pure component systems the thermodynamic factor in the equation will be unity.

Figures 4 and 5 show the comparison of the new theory with the data and with Swalin's expression, equation 19. Equation 37 predicts the diffusion behavior in these liquid metals extremely well. It is also interesting to note in these plots of log D versus the reciprocal temperature that equation 37 gives a curved line although a constant activation energy has been used in calculating each line. The values of the activation energy and entropy used in these calculations are summarized in Table I; also, the literature references for density and viscosity data used in calculating

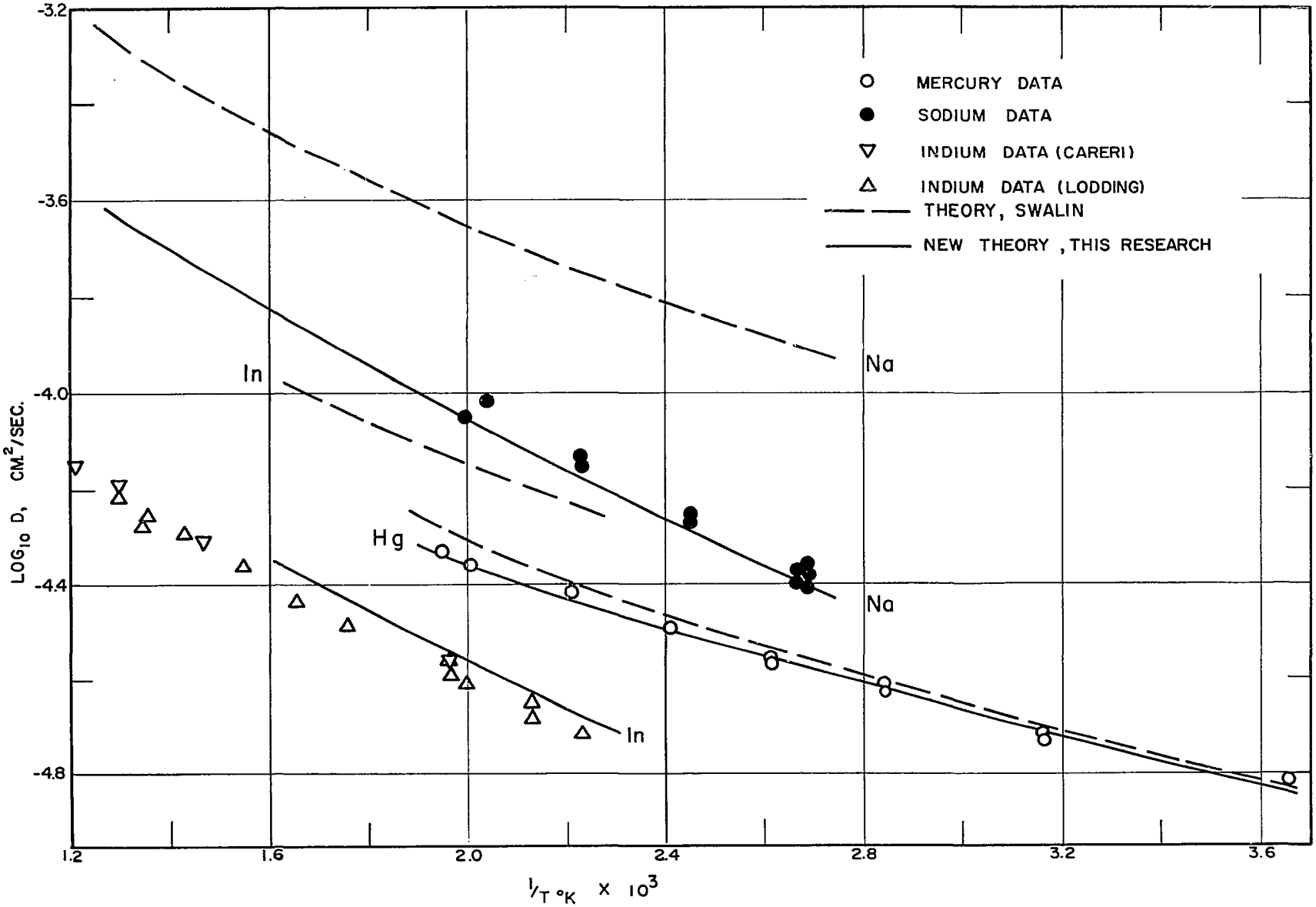


Figure 4. Comparison of Theory with Liquid Metals Self-Diffusion Data

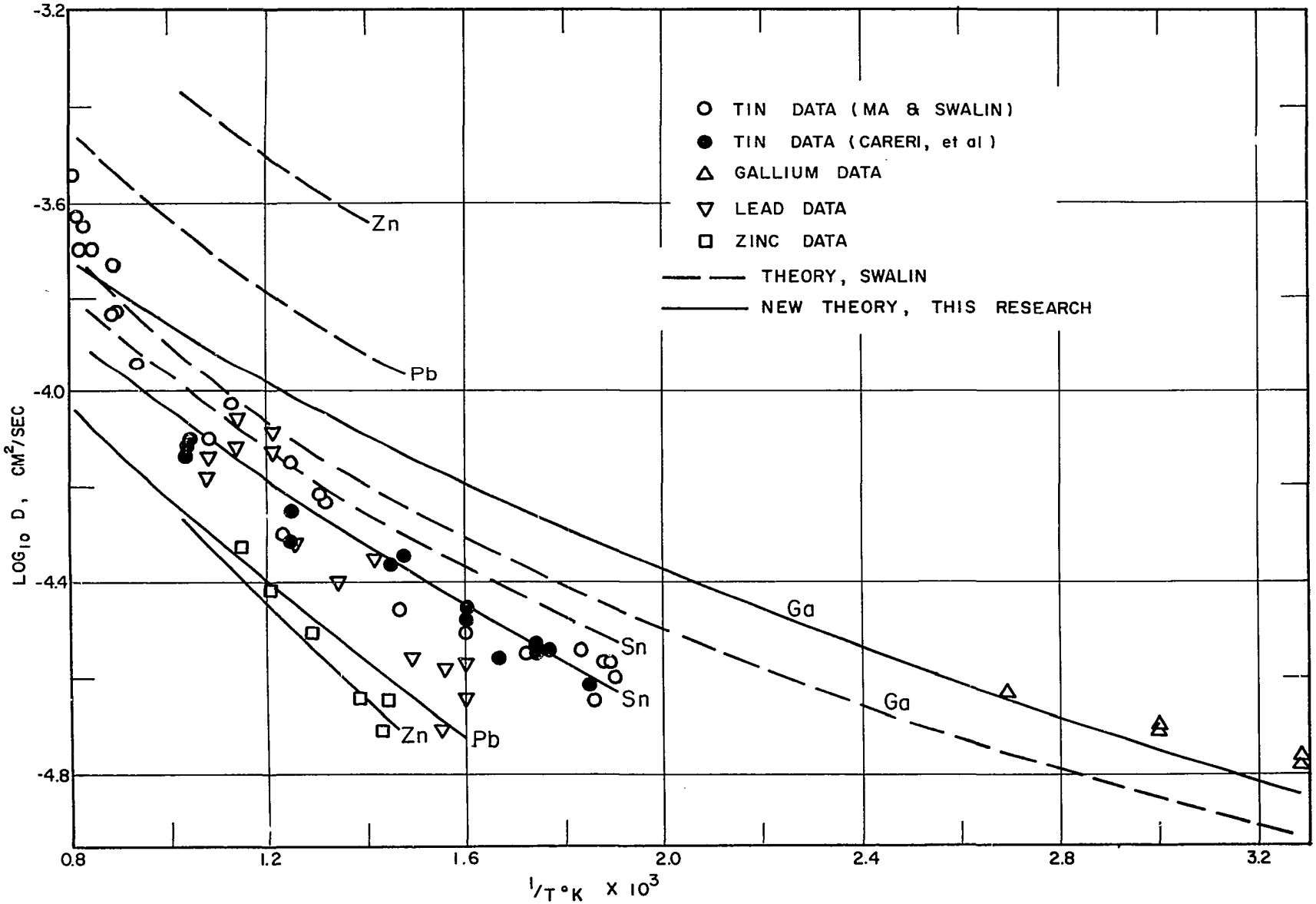


Figure 5. Comparison of Theory with Liquid Metals Self-Diffusion Data

TABLE I

ACTIVATION ENERGY AND ENTROPY
FOR VARIOUS LIQUID METALS

Element	Activation Energy, Q (Arrhenius)		Activation Energy, ΔH^*, For Viscosity		Activation Entropy, ΔS^*, For Viscosity
	cal/mole	Ref.	cal/mole	Ref.	cal/mole-°K
Mercury	1160 1005	20 21	559	28, 29, 30,	-6.16
Gallium	1122	23	904	31	-5.17
Tin	2768	25	1448	32, 33	-5.95
Indium	2554 2430	25 22	1236	34	-5.81
Zinc	5600	24	2768	35, 36	-4.78
Lead	4450	26	2130	37	-6.27
Sodium	2430	27	1392	38	-3.76

these quantities are indicated. The diffusion data shown in these figures were obtained from the references given in Table II.

Values for the activation energy calculated from the Arrhenius expression, equation 24, are given in Table I for comparison with the values used in equation 37. The activation energies determined by equation 24 are difficult to interpret in any theoretical sense. Therefore, the Arrhenius equation as applied to liquid metals diffusion should be

considered as an empirical representation of data over not too great a temperature range.

TABLE II

LITERATURE REFERENCES FOR LIQUID METALS SELF-DIFFUSION DATA

Element	Reference
Mercury	19
Gallium	23
Tin	25, 39
Indium	25, 22
Zinc	24
Lead	26
Sodium	27

The agreement between the theoretical prediction of equation 37 and the diffusion data for all the liquid metals used in this evaluation of the theory indicates that any one of the systems could have been used to determine the constant b. Since b is apparently a constant for the liquid metals and equal to 0.419, the free volume of these materials will vary only with the cube of the interatomic spacing (assuming the coordination number is a constant). Using equation 38 and the value for b gives the following expression for the free volume per mole for liquid metals:

$$V^f = 0.01072d^3, \tag{50}$$

where d is measured in angstroms. For mercury, the free volume calculated from equation 50 is 0.289 cm^3/mole. This calculated quantity is in good agreement with that for many other liquids (18).

In conclusion, it may be stated that the new theoretical model consistently and accurately predicts the diffusion behavior in the liquid metals tested to date. This model, which indicates the activation energy for diffusion to be equal to that for the viscosity from Eyring's equation, fits the data better than Swalin's equation, which is not based on an activation model. Finally, it should be mentioned that Eyring's latest equation for the diffusion coefficient, whether or not it is theoretically correct, would give results only three or four percent different from the equation developed in this work.

CHAPTER III

EXPERIMENTAL PROCEDURES AND EQUIPMENT

The experimental phases of this research involved the measurement of densities, concentration cell potentials to determine thermodynamic activities, and self-diffusion coefficients in the amalgams of cadmium and zinc. The necessary items of procedure and equipment for these measurements are described in this chapter.

Materials and Preparation of Amalgams

The materials and chemicals used in this work were all ACS Reagent Grade except the cadmium which was obtained from the Consolidated Mining and Smelting Company of Canada, Limited, and was their intermediate high purity grade (99.999% Cd).

The radioactive isotopes of zinc and cadmium used in the measurement of the self-diffusion coefficients are described in Appendix D.

The inert amalgams were prepared by dissolving the solute zinc or cadmium directly in the mercury. In some cases, the technique of Crenshaw (40) was employed, in which the mercury is made the cathode of a cell and a platinum anode is placed in distilled water over the mercury. The

solute material is placed on the mercury surface and a 12 volt potential applied to the cell in order to enhance the amalgam formation. The use of such a cell was also a convenient method for storing the alloys, once prepared, to avoid the preferential oxidation of the solute within the amalgam. These amalgams were in some instances stored under a deoxygenated glycerol layer.

The preparation of the radioactive amalgams proceeded in much the same fashion except that the radioisotope was either directly introduced in the mercury by electrolysis or first plated on inert material and then dissolved in the mercury. The active zinc amalgams were prepared by using a zinc chloride electrolyte solution, this being the chemical form of the isotope available from the supplier. In the case of active cadmium amalgams the plating was done from a cadmium sulfate electrolyte. The cadmium nitrate containing the isotope as purchased was treated with sodium carbonate to give a cadmium carbonate precipitate. The precipitate was subsequently dissolved in a sulfuric acid solution to give the desired cadmium sulfate solution. The change from the nitrate to sulfate electrolytes avoided the problems of cadmium oxide deposition associated with the nitrate bath. The radioactive amalgams were stored in the same manner as the inert amalgams.

These amalgams were all prepared by weight and the compositions verified using the concentration cell described in a later section.

The mercury was reclaimed from spent amalgams and, after purification, was reused in this work. The process of reclaiming the mercury began with acid washing of the spent amalgam. The contaminated mercury was next placed in a heated oxidizing cell with air introduced through a sparger below the amalgam surface, the effluent air being treated to remove any mercury vapor entrained from the cell. This oxidation of the impurities continued for four to eight hours. The contents of the cell became very viscous at the end of this process if there was a high concentration of impurity to begin with. After the oxidation step, the material in the cell was washed in beakers of acid and then sprayed through four-foot-long wash columns containing nitric acid or water. This process usually involved about six washings in the acid column and several times through the distilled water column. A sodium hydroxide solution was used to remove grease or oil contaminants. Finally, the mercury was, as a minimum, triple distilled, this operation being continued until no trace of impurity was visible on the mercury surface.

In all of these purification operations, special precautions were taken to segregate the radioactive amalgams from other amalgams, since the wash solutions used for the extractions of the active solute materials required special handling in compliance with safety regulations. These active waste solutions were concentrated by evaporation to facilitate handling and disposal.

Equipment

The Pycnometers

The pycnometers used in the density measurements were made using a T/S No. 19/38 pyrex ground joint as shown in Figure 6. The outer portion of the joint was used in fabricating the pycnometer cap shown at the bottom of Figure 7. The capillary section of the pycnometer was approximately 0.5 mm inside diameter. The apparatus shown at the top of Figure 7 was used in preparation of the amalgam under argon and for loading the pycnometers. The arrangement of stopcocks permitted the pycnometer attached to the bottom end (left side of the figure) to be evacuated before the filling operation.

The Concentration Cells

One of the concentration cells is shown in Figure 8 and consisted of three wells equipped with tungsten electrodes. Platinum electrodes were used on some of the cells. The arrangement of the connecting tube between the legs of the cell, as is shown in the figure, facilitated the filling of the cell in that possible mixing of amalgams could be avoided.

The Constant Temperature Baths

One of the constant temperature baths used in this work is shown in Figure 9. A pyrex jar 12 inches in diameter and 18 inches deep was equipped with a 1000 watt ballast heater and a 250 watt control heater. The control heat input was regulated by an off-on type mercurial thermoregulator or

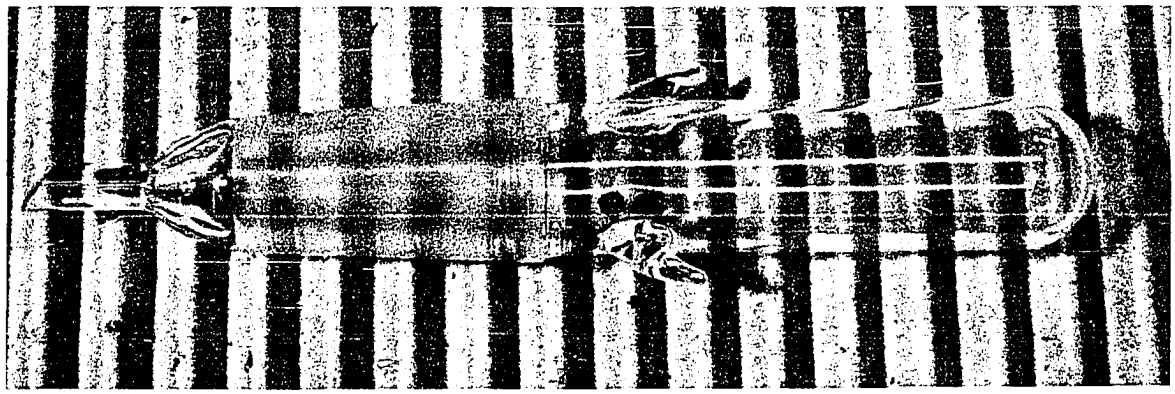


Figure 6. Pycnometer

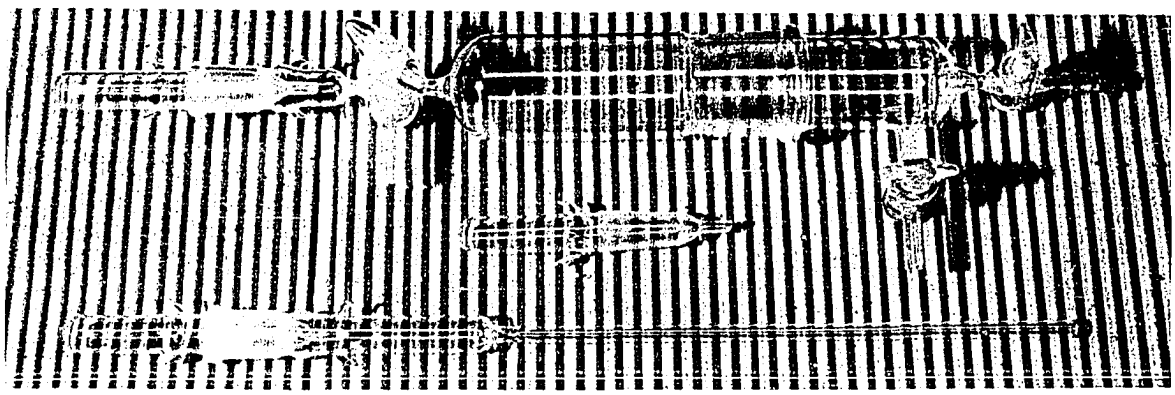


Figure 7. Pycnometer and Associated Apparatus

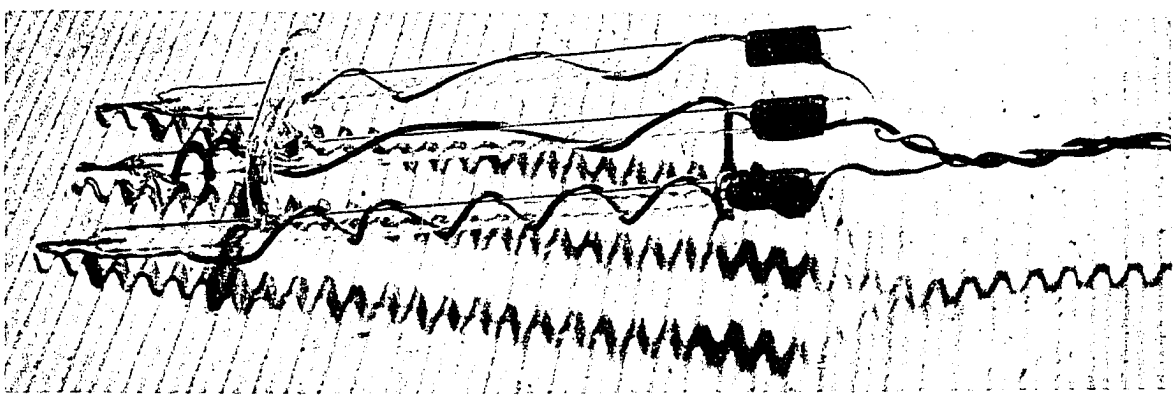


Figure 8. Concentration Cell

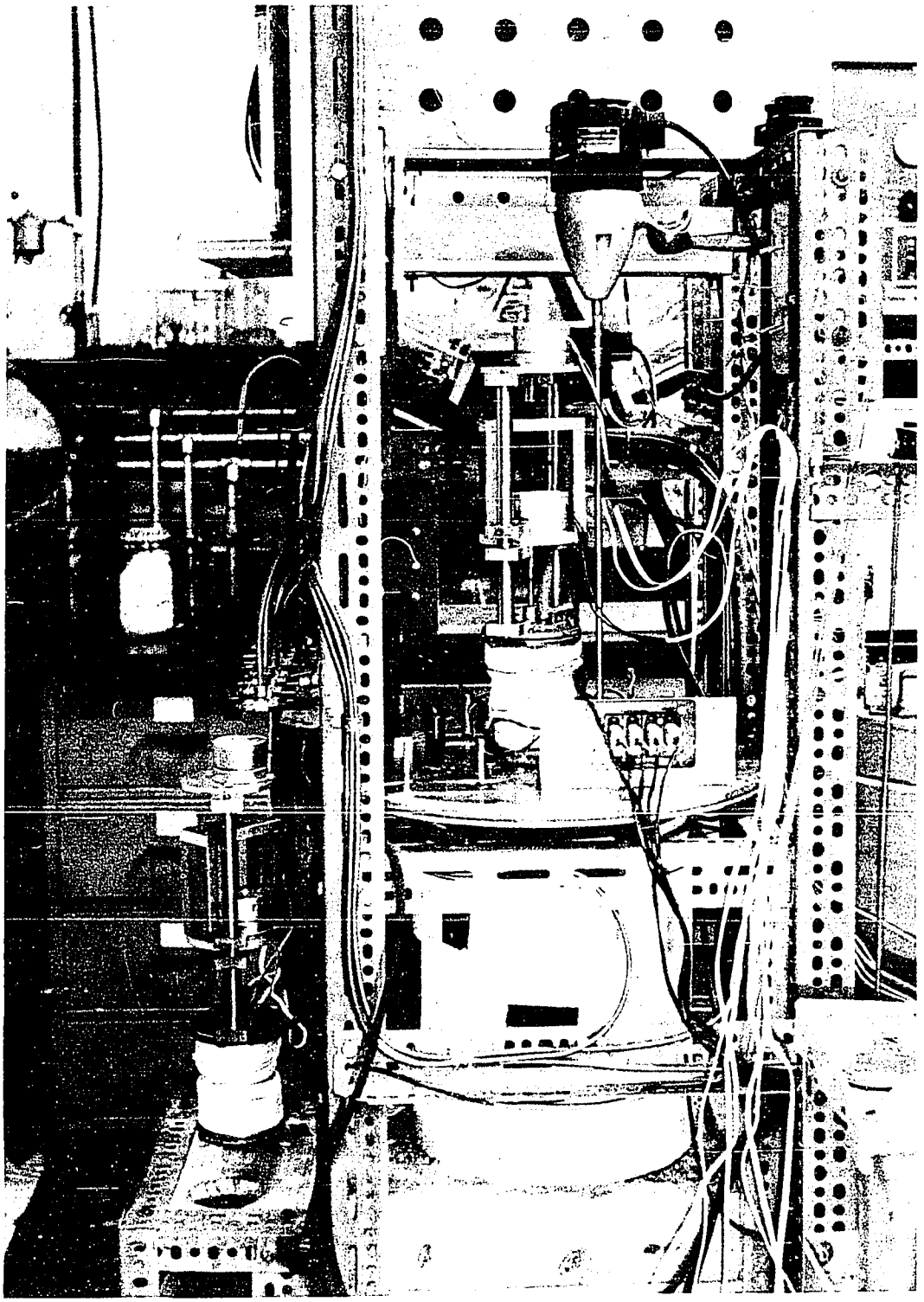


Figure 9. Constant Temperature Bath and Associated Equipment

a Bayley proportional controller. The temperature variation in the baths was no more than ± 0.01°C during data measurements or runs. As many as four calibrated thermocouples were located in the bath at various positions to measure temperature and check for gradients. The ballast heater input was adjusted at the individual temperature levels used by means of a variable autotransformer.

The liquid used in the constant temperature baths was Crisco. The recent changes in this material (an increase in unsaturated components) were noticed to have made the new product inferior to the former for this particular use from the standpoint of stability at temperatures up to 150°C. A constant speed mixer was used to stir the bath liquid, and it was observed that, of the various mixers used, those with sleeve bearings were superior in performance for this application.

The baths were shock mounted, and this was of particular importance during the diffusion runs.

The Diffusion Cells

The capillary-reservoir method (42) was used in measuring the diffusion coefficients. Figure 10 shows the diffusion cells and part of the associated equipment. The cells were made of 60 mm pyrex tubing about 18 inches long with a flattened bottom. Approximately 12 inches of the cell was immersed in the constant temperature bath liquid during a run. The upper portion of the cell was insulated to minimize

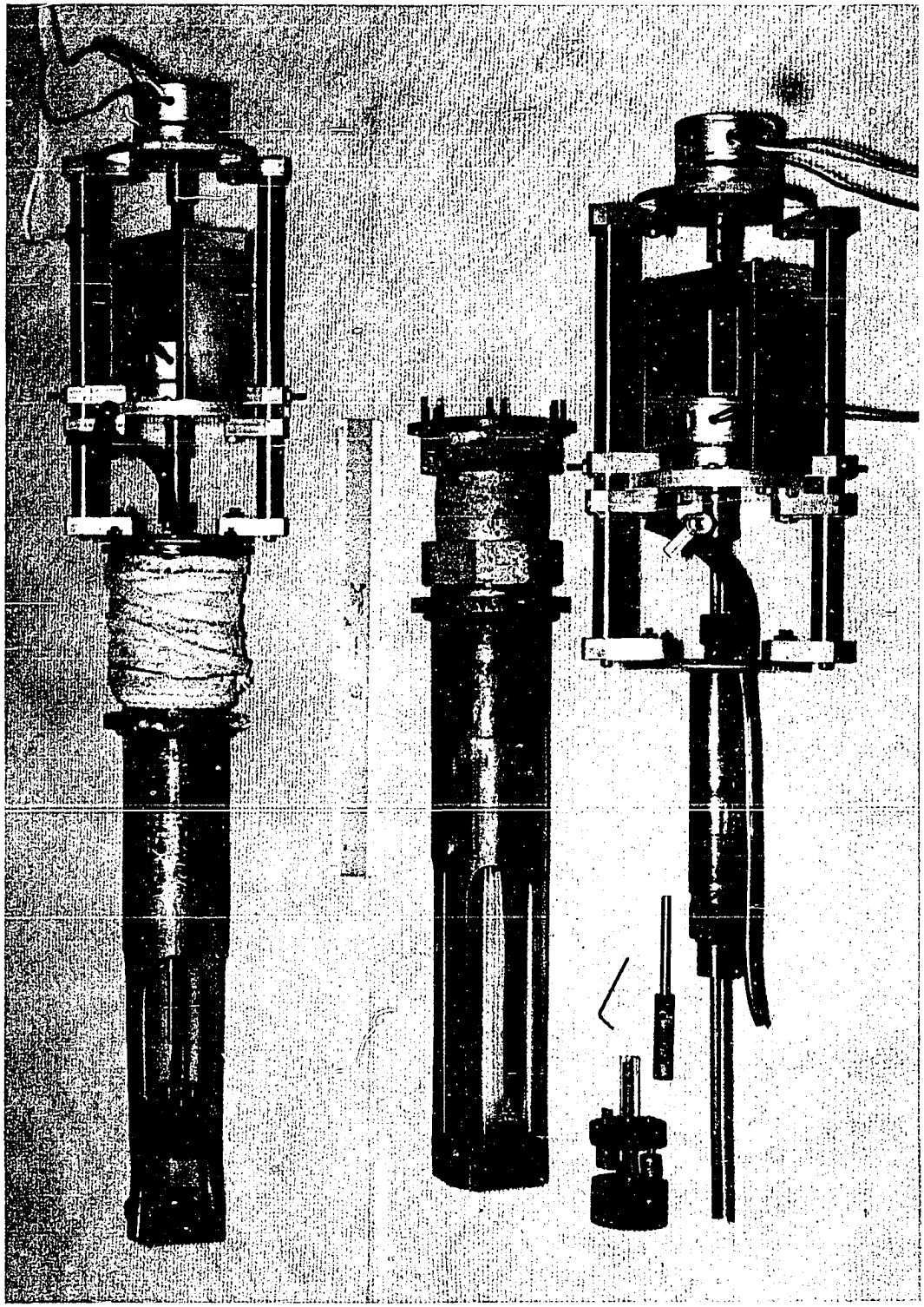


Figure 10. Diffusion Cells

thermal convection within the cell. As shown in the figure, the cell was held by a container made from three-inch brass tubing, part of which was cut out to permit circulation of bath liquid around the cell and to allow observation of the diffusion reservoir area. Near the upper end of the cell holder a flange was attached to support the whole assembly through the top of the constant temperature bath. This flange was insulated from the bath top with a rubber "O-ring" to reduce the transmission of vibrations to the diffusion cell from the rest of the equipment.

The top seal of the diffusion cell was made by brazing a flange to one end of a Dresser Coupling which was placed on the upper part of the cell. The top assembly was then bolted to the flange and sealed with an "O-ring." The top assembly consisted of the drive mechanisms for rotating and vertically positioning the capillary holder. The rotational drive was a one-half rpm synchronous motor, and a reversible 6 rpm synchronous motor was used for the vertical adjustment. The vertical motion was accomplished by a fixed screw drive from the reversible motor to the movable plate holding the rotational drive. The drive shaft for the capillary holder slipped through a seal in the flange plate of the top assembly. These capillary drive systems, although somewhat complex, performed very well. Earlier drive mechanisms used in this work had used a hand-operated device for vertical positioning and were equipped to rotate the capillaries at variable turning rates.

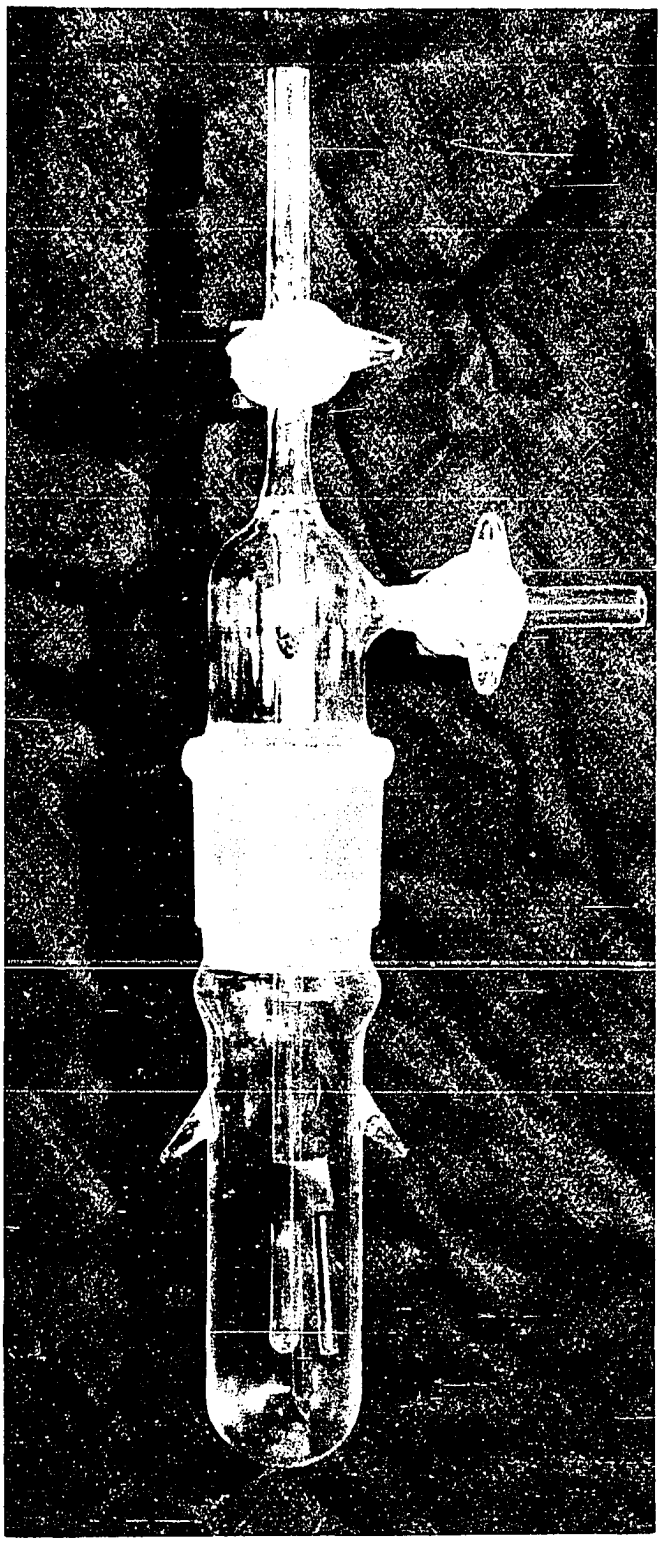


Figure 11. Capillary Filling Device

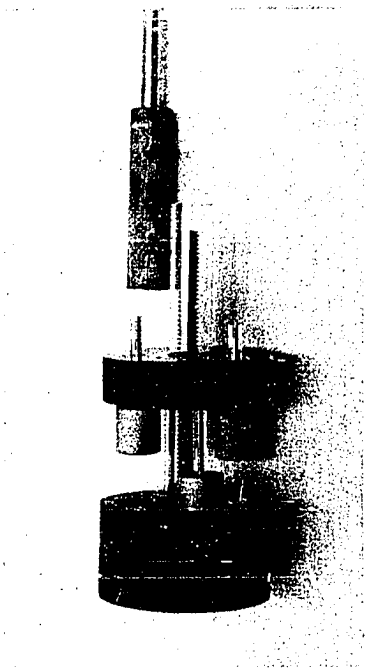


Figure 12.
Capillary Holder,
Disassembled

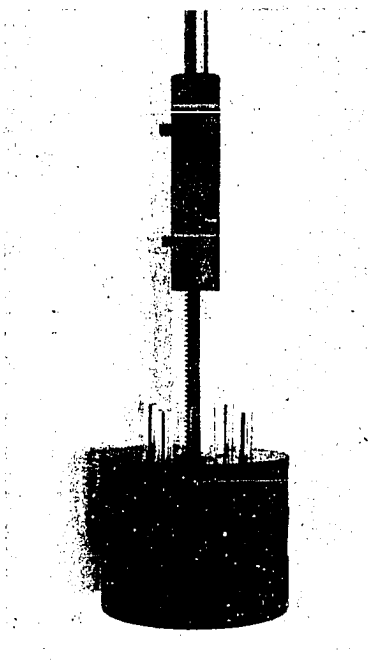


Figure 13.
Capillary Holder,
Assembled

The capillary holders are shown about half-size in Figures 12 and 13. These holders were made of lava and constructed to hold four capillaries. The capillaries were held in the holders by steel sleeves with setscrews. No steel parts of the holders contacted the reservoir amalgam. The complete holder assembly was attached to the capillary drive shaft by a steel coupling as indicated in the figures.

The Capillaries

The capillaries used in the diffusion measurements were fabricated from precision-bore pyrex tubing obtained from Corning Glass Works. The dimensions of the capillary tubing are presented in the following table. One end of the

TABLE III

CAPILLARY SPECIFICATIONS

Inside Diameter, millimeters	Outside Diameter, millimeters
1.000 ± 0.025	2.97
1.500 ± 0.025	3.20
2.000 ± 0.025	4.11

capillary was sealed so as to give a flat inside bottom and the open end was ground flat using 600 grit silicon carbide paper, the capillary being held in a jig to insure that the ground surface was normal to the capillary axis. The ground surface was finally polished on 3/0 metallographic paper to

yield a smooth surface without defects visible when inspected by a 20 power microscope.

The bore dimension of each capillary was checked using an optical comparator and was in every instance within the specified tolerance limits. The capillary inside lengths of approximately 3.5 cm were measured with a depth gauge to an accuracy of ± 0.0025 cm.

Approximately 12 dozen capillaries were used in the diffusion measurements and these were given no permanent designations. It was noted that the capillaries tended to become chipped at the open end during handling involving the use of hypodermic needles for washing the capillaries in spite of seemingly careful precautions. Consequently, each capillary was inspected under the microscope following the cleaning process and was reground and repolished if chipping had been observed. After the capillary was cleaned and without defects, the inside length was remeasured. A final inspection under the microscope followed the length determination and if the capillary showed no chips had developed during the insertion of the depth gauge, the capillary was then suitable for use in diffusion measurements.

Capillary Filling Device

The capillary filling device, Figure 11, permits the loading of amalgam into the capillaries by first evacuating the capillary and then, following the submergence of the open end below the amalgam surface, forcing the amalgam inside by

repressurizing the system with argon. The capillary amalgams were contained in the lower portion of the chamber and the capillary was held inverted over the amalgam surface. The use of an adjustable clamp to hold the entire device permitted immersing the end of the capillary in the amalgam as the device was tilted.

The Radioisotope Counting System

The isotope counting system was based on the use of a scintillation well-type detector and consisted of the following major components: a Nuclear-Chicago Corporation Model 183-B scaler, a Tracerlab Corporation Model RLI-4 pulse height analyzer, and a Tracerlab Corporation Type P-20CW scintillation detector. These system components were operated from a constant voltage transformer. Also, the detector shield and crystal were maintained at constant temperature by cooling water in an attempt to minimize the photopeak drift. Eventually, the system was enclosed and a forced convection cooling system with some control of the air temperature was used in attempt to reduce the instrument operating variations. Although this arrangement helped the situation, it would have been beneficial to have had some control over the humidity of the air used for cooling the system.

The performance of the counting system was satisfactory, as is shown in Appendix D, Figure D-1, which shows the radiation energy spectrums of various materials used in this research.

Procedures

Concentration Cell Measurements

The measurements of concentration cell potentials were made for two purposes. First, the thermodynamic activities and subsequently the thermodynamic factor needed for evaluating the diffusion equation were determined from these data. Secondly, the measurement of the cell potentials provided a convenient means for measuring or verifying the solute concentrations of various amalgams.

The reference amalgams in these measurements were slightly supersaturated at the temperatures used. A 0.1 molar solution of zinc sulfate and a 0.5 molar solution of cadmium sulfate were used as the cell electrolytes for the measurements of zinc and cadmium amalgam potentials, respectively. These electrolyte solutions were heated to about their boiling point and then cooled while bubbling argon through the solution in order to remove dissolved oxygen.

The procedure in measuring the cell emf was first to place the amalgams, one saturated and one dilute, into the concentration cell. The concentrated amalgam used as a reference was preheated to give a single phase solution before it was placed in the cell. After the electrolyte solution was added, the cell and contents were preheated in an auxiliary bath to a temperature five to ten degrees higher than that desired for the determination. This

procedure hastened the attainment of equilibrium conditions in the two-phase reference amalgam.

After sufficient time for thermal equilibration of the cell and contents with the constant temperature bath, the potential developed by the cell was measured with a potentiometer. Specifications of the potentiometer are given in Appendix C. These procedures gave highly reproducible measurements from several batches of constant-composition amalgams (prepared by weight). The emf measurements were checked also by taking repetitive readings on a single amalgam over a period of several days. No change in emf was observed.

The data for the concentration cell potentials of these amalgams at 50.0°C were used in the composition measurements (see Figure 15, Chapter IV). The errors associated with these composition determinations are discussed in Appendix C.

Density Measurements

Densities for cadmium and zinc amalgams were measured in pycnometers that were calibrated by using pure mercury. The procedure was to fill a pycnometer with amalgam and then place it in the constant temperature bath. As thermal equilibrium was reached, the excess amalgam (which was present because of the lower filling temperature) overflowed from the pycnometer and was caught in the cavity between the capillary section of the pycnometer and the top piece (see bottom

of Figure 7). After the pycnometers were removed from the bath and all oil cleaned from the outside, the pycnometer and contents were weighed on a standard analytical balance using an empty pycnometer as a counterpoise. The weight of the pycnometer when empty (measured against the same counterpoise) was subtracted from the weight of the pycnometer plus amalgam. This quantity was then divided by the pycnometer volume to give the amalgam density.

The pycnometer volumes were determined from the pure mercury calibration data and are given in Appendix B, Table B-I as a function of temperature. The calibration procedure was exactly as described above for the amalgam measurements to the point of calculating the density. The average for weights of mercury in the pycnometer was divided by the density of mercury (30) to obtain the volume. The volumes so determined were then used to calculate the densities of pure mercury presented in Table B-II. These calculations were made for the statistical purposes of testing for pycnometer bias and establishing the precision of this technique for measuring densities. The average of these data will obviously agree with the accepted mercury density data since these values were employed in the volume determinations. The statistical test showed that the pycnometer bias is not significant at a confidence level of 0.999. These statistical calculations are summarized in Table B-III.

Self-Diffusion Measurements

The measurement of self-diffusion coefficients was accomplished in the following manner. After the capillaries were prepared and filled with radioactive amalgam as previously described in this chapter, they were counted to determine the initial activity level of each capillary. The number of counts taken for any measurement of initial or final capillary activities varied from 6400 to 128,000 counts depending on the isotope and the specific activity of the amalgam. Next, the capillaries were placed in the capillary holder and this was coupled to the capillary drive shaft. The glycerol layer covering the reservoir in the diffusion cell was then removed, the diffusion cell being already at temperature in the constant temperature bath. After the capillary drive assembly with the capillary holder in a raised position was secured to the cell, the vertical drive lowered the holder into the reservoir at a rate of one-third inch per minute. As the capillary mouths were submerged, the starting time of the run was read from a stopwatch which had been synchronized with an electric clock. When the capillaries were well beneath the reservoir surface, the vertical drive was stopped and the rotational drive started. The reservoir surface was then again covered with the glycerol solution, which was preheated as it flowed down the side of the cell to the reservoir so as not to create turbulence within the reservoir.

At the termination of the diffusion run, the rotational drive was stopped and the capillaries were raised out of the reservoir by the vertical drive. The time was measured (as described above) when the capillaries broke through the reservoir surface. After removal from the cell, the capillaries were left with glycerol covering the open end to prevent oxidation of the amalgam solute during the time period which elapsed before the final activity count was obtained. It was found necessary to let these capillaries stand about a day before the final counts were measured because of the concentration gradient of radioisotope which developed within the capillary during diffusion. This gradient, which must be present in the diffusion process, was such that the geometry of the counting system used could not give immediately the final average count. Thus, the waiting period permitted this gradient to be diminished and the final average activity level of the capillary to be correctly measured.

Background activity measurements were frequently made and all count rates used in calculating the diffusion coefficient were corrected for the background level. Also, a reference sample of the isotope was counted before and after each capillary counting to obtain a gross correction factor for both radioactive decay and variations in the counting system performance.

The problem of convection within the capillary-reservoir system has been a major consideration. Convection

could occur because of density gradients resulting either from thermal or concentration gradients. The existence of thermal variations within the reservoirs of these amalgam systems was not measurable in the constant temperature bath, probably because of the high thermal conductivity of the amalgams. Severe convection effects were observed, however, with capillary diameters as small as one millimeter in situations where the reservoir density was only slightly greater than the density of the capillary amalgam. Therefore, in the measurement of self-diffusion coefficients, the composition match between capillary and reservoir solutions was very critical. In this investigation the concentrations of these materials were checked by the use of the emf cell shown in Figure 8, with the reservoir amalgam as one electrode, the capillary amalgam as a second electrode, and a saturated reference amalgam as the third electrode. This arrangement permitted independent readings of the cell potential between the capillary or reservoir amalgam and the reference amalgam. Also, a check could be made directly between the capillary and reservoir amalgams. Compositions of the solutions were adjusted if necessary until the emf developed during the verification procedure was precisely zero to the limit of the potentiometer sensitivity. This technique appears to have eliminated convection as a significant source of error.

A brief description of the mathematical analysis of diffusion in the capillary-reservoir system, based on the

use of Fick's law, is given in Appendix E. The resulting solution to the analysis is presented in tabular form and this was used in calculating the diffusion coefficients from the measured data. Appendix F contains a sample data sheet used for the diffusion runs and also gives sample calculations for the evaluation of the diffusion coefficient.

The possible effect of capillary turning rate on the diffusion coefficient was checked from zero to 1.27 rpm and these data are given in Appendix G. The data indicate no significant effect of the turning rate on the coefficient. Thus, the selection of the one-half rpm drive mentioned earlier was arbitrary.

Summary of Experimental Work

The experimental objectives of this program included the following items.

A. The measurement of densities for cadmium and zinc amalgams as a function of composition and temperature.

B. The measurement of solute thermodynamic activities in various cadmium and zinc amalgams as a function of temperature.

C. The measurement of solute self-diffusion coefficients in mercury-rich amalgams of cadmium and zinc at various compositions and temperatures.

The temperature levels selected for measurements were 50.0, 70.2, 93.2, 119.1, and 150.0°C, although not all of

the above items were necessarily measured at each temperature. The choice of concentrations was generally arbitrary for the measurements of densities and thermodynamic activities. For the case of the diffusion measurements, however, it was decided to specify the ratio of cadmium amalgam concentrations to those of the zinc amalgams in order to use the isoviscous relationship between certain of these amalgams, as was mentioned in Chapter I. Using the data of Golik and co-workers (43,44) to determine this concentration ratio, the cadmium amalgam concentrations versus those of zinc amalgams for isoviscous solution behavior are plotted in Figure 14. From the figure, it is observed that the ratio of atomic percent cadmium to zinc in these amalgams is about, 1.32 and this relation was used as a guide in choosing the amalgam compositions for the diffusion measurements. The curve in Figure 14 must pass through the origin on the graph since pure mercury is isoviscous with itself. The data used to prepare Figure 14 are given below.

TABLE IV

REPORTED ISOVISCOUS CADMIUM AND ZINC AMALGAM PAIRS

Atomic Percent Cadmium	Atomic Percent Zinc	Reference
13.9	10.6	43
18.1	12.1	43
30.9	21.1	43
31.9	24.4	44
37.3	28.2	44
43.3	32.6	44

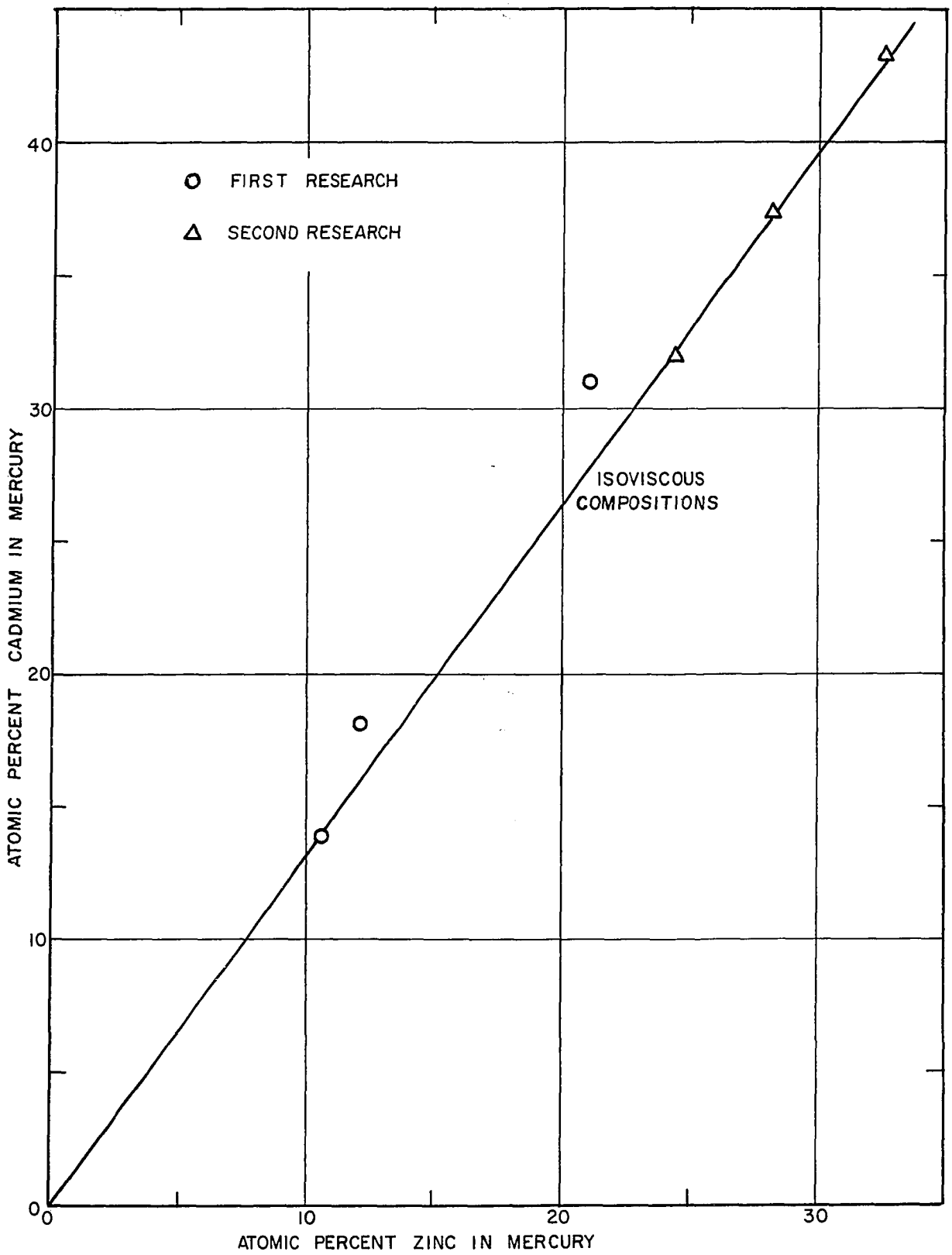


Figure 14.--Reported Solute Compositions for Isoviscous Amalgams of Cadmium and Zinc (Data of Golik and Co-workers)

CHAPTER IV

EXPERIMENTAL RESULTS

The results of the concentration cell potential measurements are summarized in Figure 15 which shows the logarithm of the weight percent solute versus the emf developed by the cell. The data for these zinc and cadmium amalgam potentials are given in Appendix K, Tables K-I and K-II, respectively. These measurements were made at 50.0, 70.2, and 93.2°C. During the determination of the amalgam emf values, the equilibrium solubilities of cadmium in mercury and of zinc in mercury at these temperatures were evaluated to check various data available in the literature. These data, given in Figure 16 and Table V, show good agreement with the values presented by Hansen (45), but the equilibrium solubilities for zinc amalgams deviate from data given by Iggena (46). The data of Benjamin and Strickland-Constable (47) for zinc amalgams measured at lower temperatures than in the present work show a tendency to agree with this research.

Values for the thermodynamic factor in the diffusion equation are given in Figure 17. These quantities

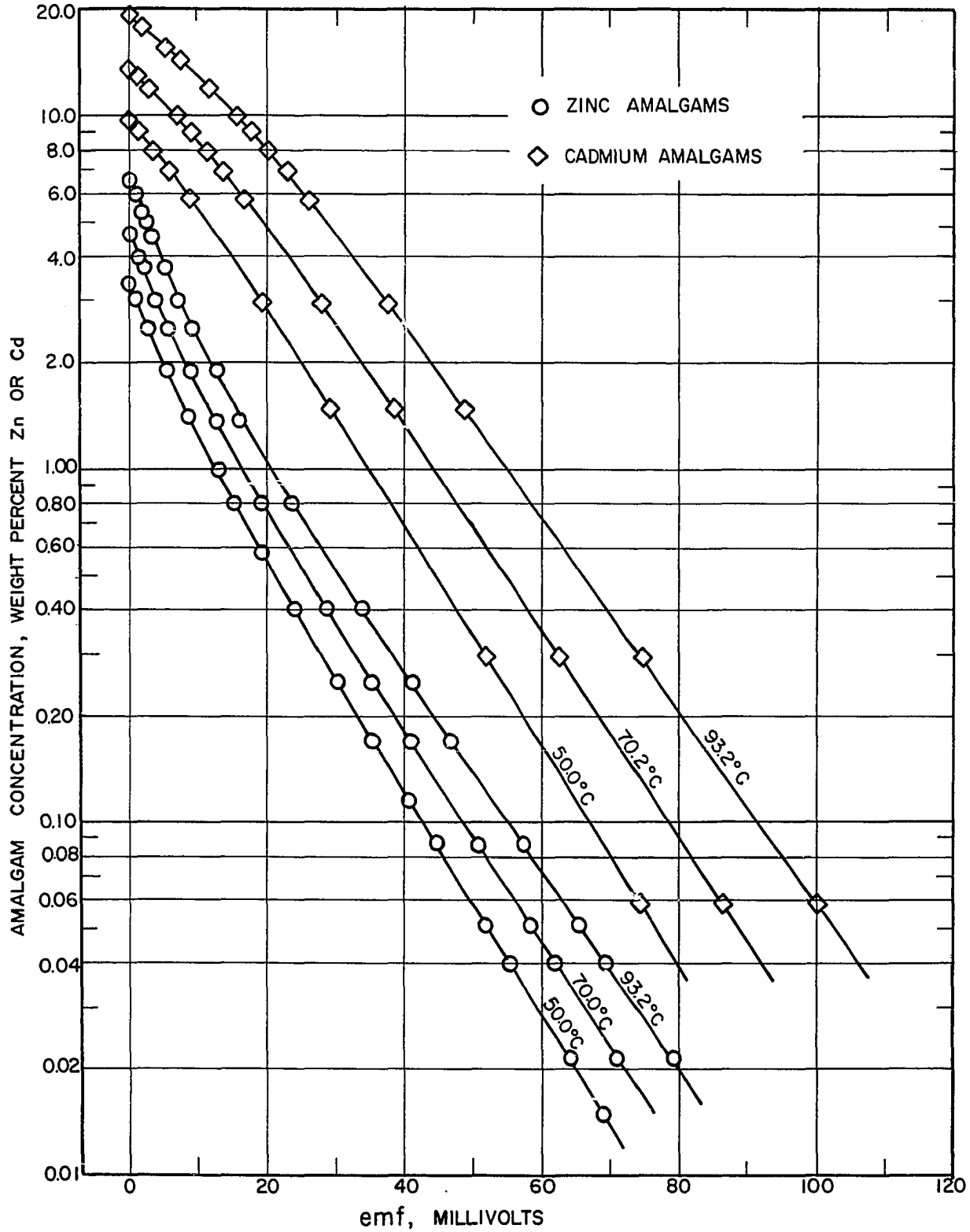


Figure 15. Concentration Cell Potential for Various Amalgams
Cells: (1) Cd-Hg (sat) | 0.5M Cd^{++} | Cd-Hg (dilute)
(2) Zn-Hg (sat) | 0.1M Zn^{++} | Zn-Hg (dilute)

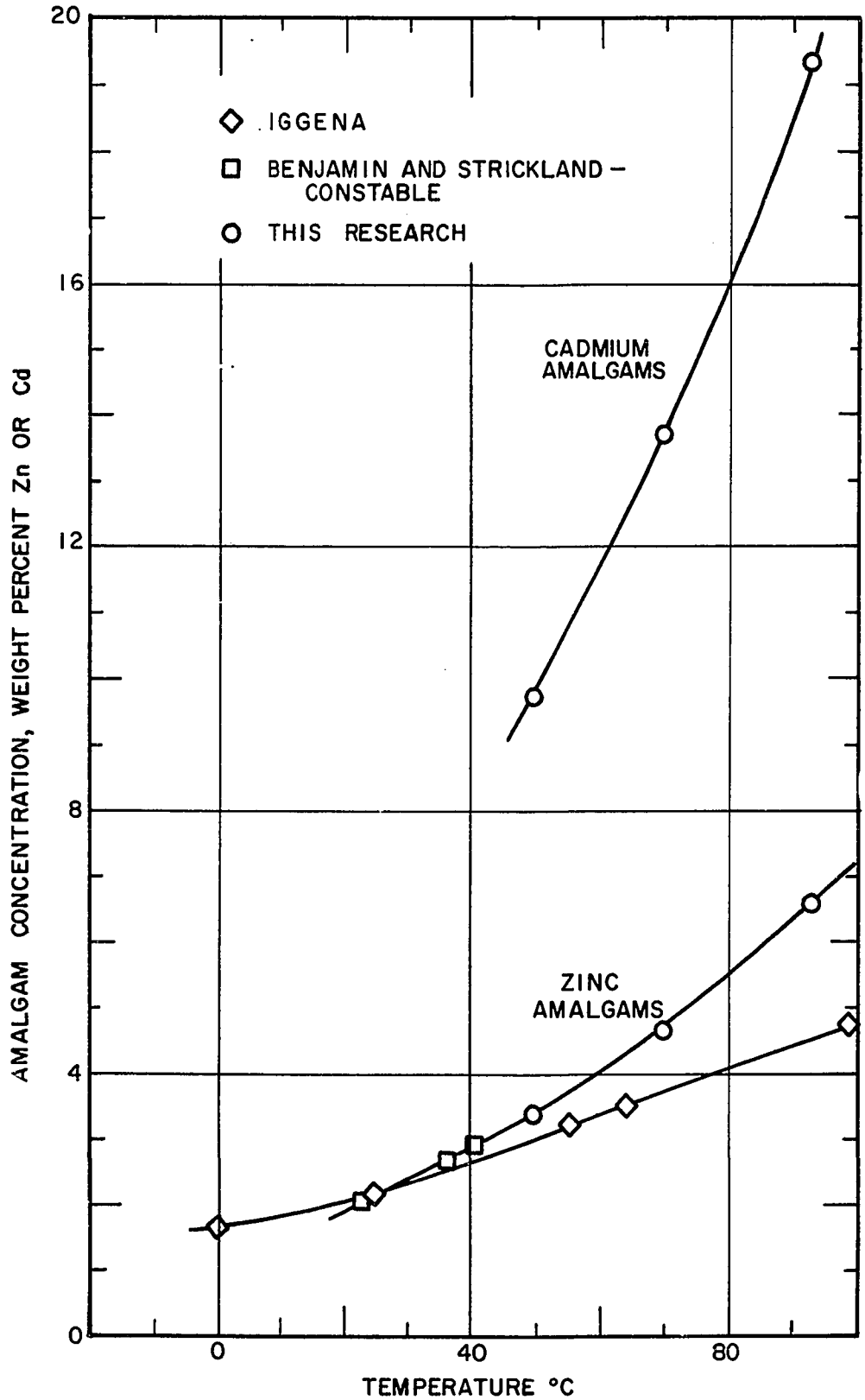


Figure 16. Equilibrium Solubilities of Various Amalgams

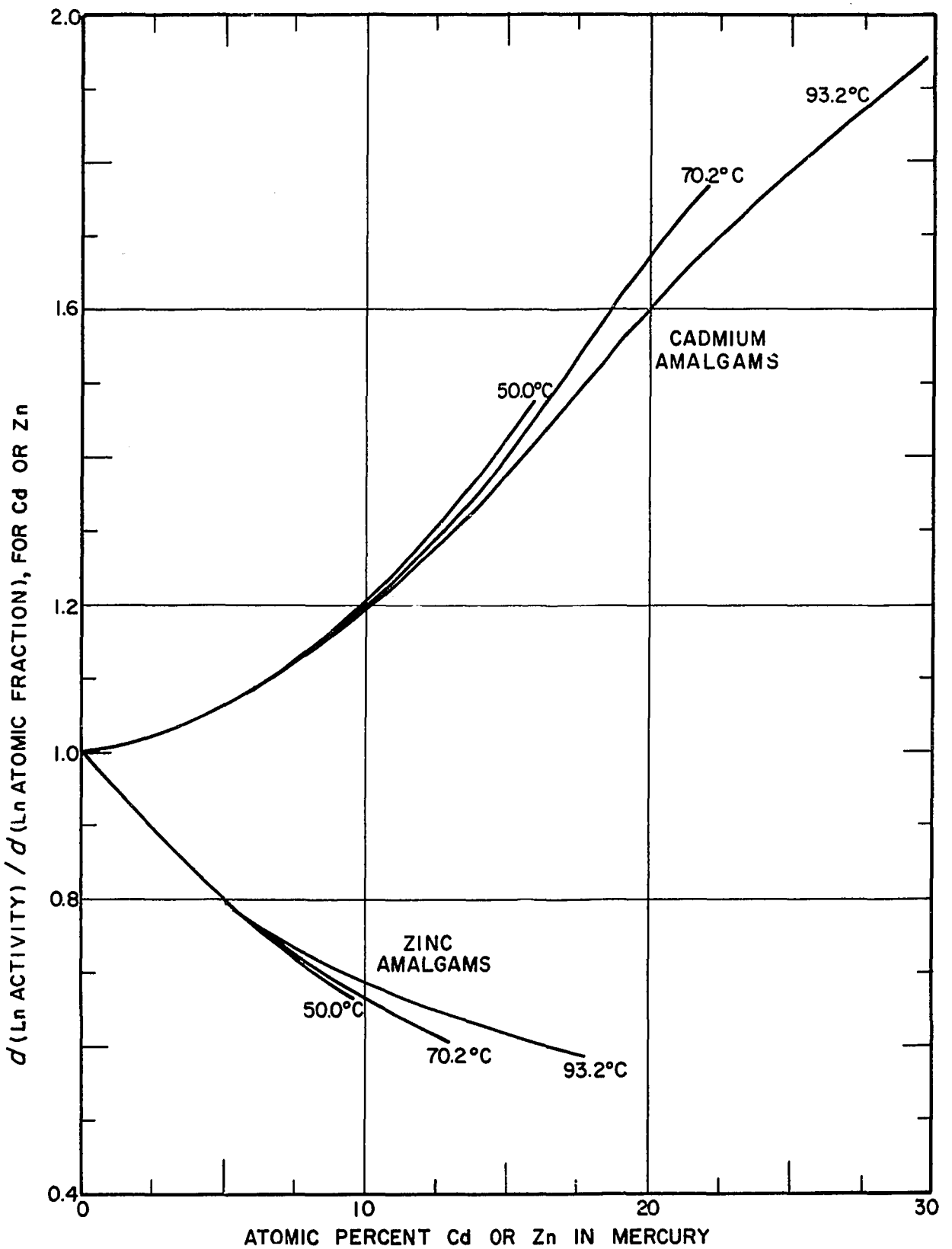


Figure 17.--Concentration Dependence of the Thermodynamic Factor for the Diffusion Equation, Cadmium and Zinc Amalgams

were calculated from the concentration cell potentials measured for the various amalgams of cadmium and zinc. The method of calculation is given in Appendix K and the calculated values are in Table K-III. This term does not show any appreciable temperature dependence at the lower concentration levels used in the diffusion measurements which are discussed later in this chapter.

TABLE V

EQUILIBRIUM SOLUBILITIES OF ZINC OR CADMIUM IN MERCURY, THIS RESEARCH

Temperature °C	Weight Percent Zinc	Weight Percent Cadmium
50.0	3.348	9.710
70.2	4.645	13.758
93.2	6.540	19.310

Amalgam densities for solutions of up to 10 atomic percent cadmium in mercury and up to 5.6 atomic percent zinc in mercury were obtained. These data, obtained in the temperature interval from 50.0 to 150.0°C, are shown as Figures 18 and 19. The average of values from the data which are given in Appendix B, Tables B-IV and B-V, are indicated below in Table VI. From Table B-III the standard deviation of these measurements is found to be about ± 0.0006 gm/cm^3, and the standard deviation of the average would be ± 0.0002 gm/cm^3.

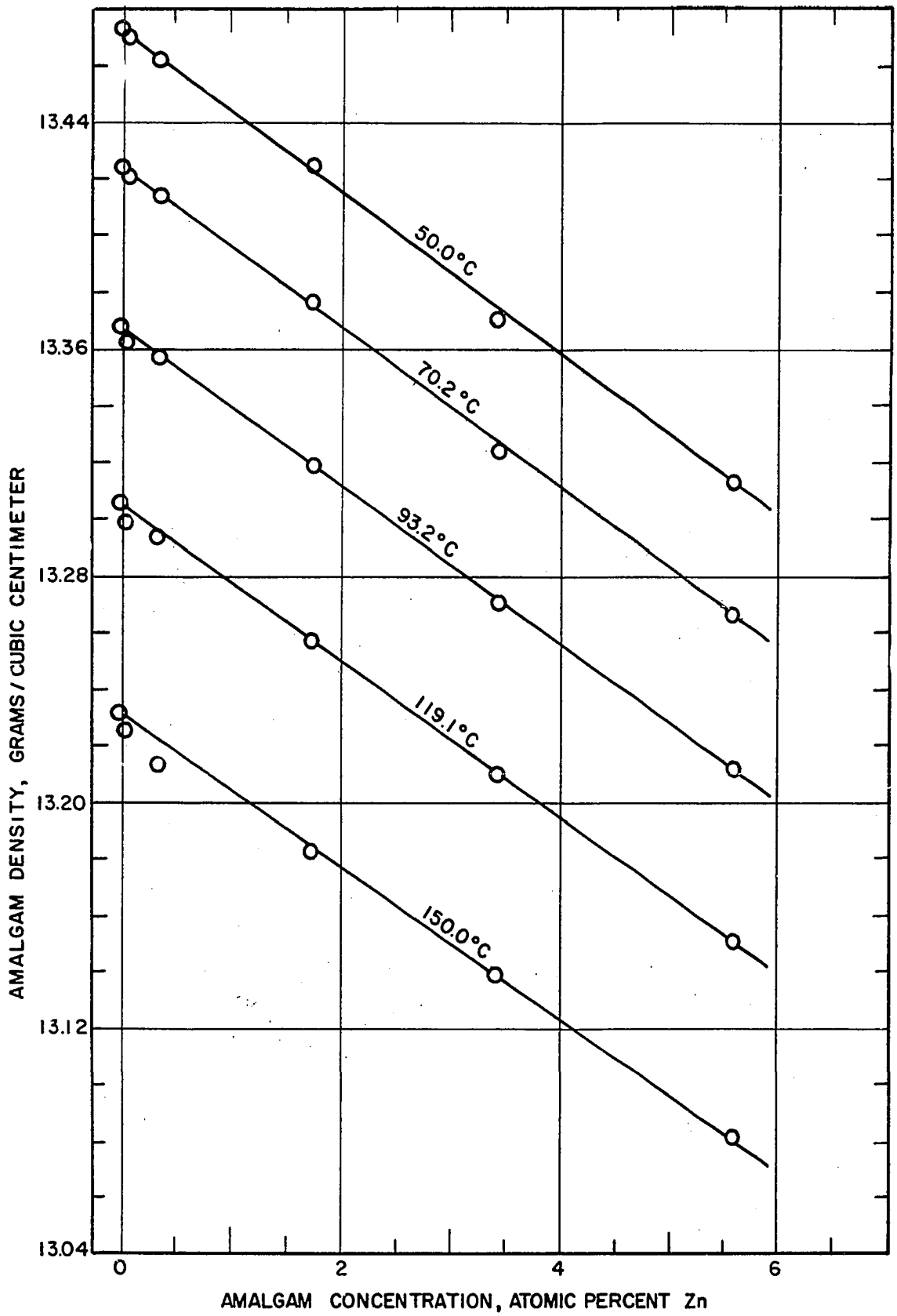


Figure 18. Density of Zinc Amalgams

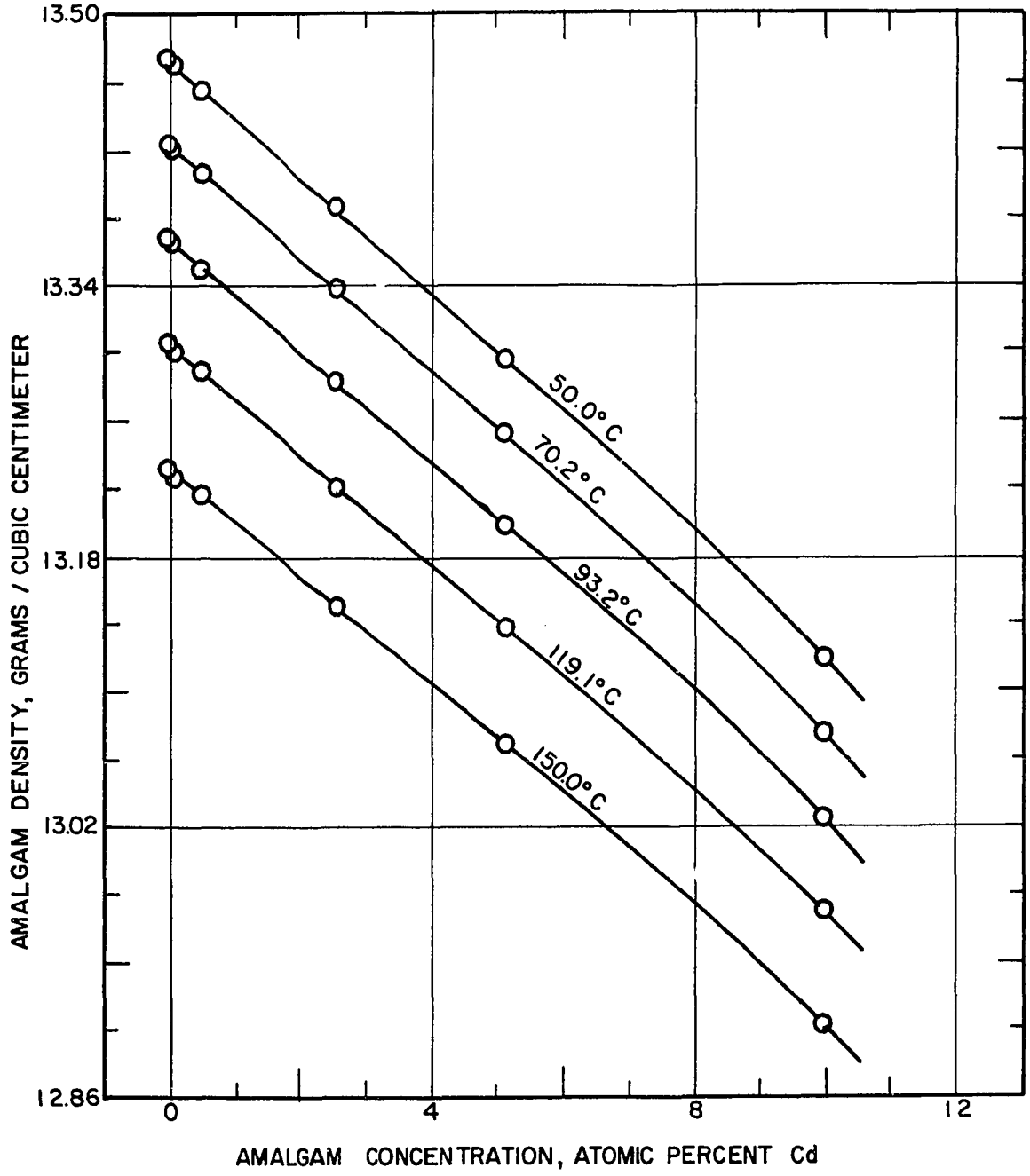


Figure 19. Density of Cadmium Amalgams

TABLE VI

DENSITIES OF VARIOUS CADMIUM OR ZINC AMALGAMS VERSUS TEMPERATURE, THIS RESEARCH

Atomic Percent Solute	Amalgam Density, gm/cm^3, at Temperature				
	50.0°C	70.2°C	93.2°C	119.1°C	150.0°C
Cadmium Amalgams					
0.1047	13.4689	13.4199	13.3626	13.3001	13.2269
0.5250	13.4550	13.4066	13.3511	13.2885	13.2162
2.625	13.3852	13.3382	13.2832	13.2206	13.1500
5.169	13.2979	13.2515	13.1983	13.1375	13.0688
9.998	13.1206	13.0760	13.0253	12.9695	12.9035
Zinc Amalgams					
0.0583	13.4698	13.4211	13.3630	13.2987	13.2247
0.350	13.4625	13.4140	13.3581	13.2943	13.2140
1.750	13.4245	13.3765	13.3194	13.2572	13.1831
3.446	13.3705	13.3246	13.2701	13.2102	13.1399
5.609	13.3136	13.2672	13.2137	13.1517	13.0828

The zinc amalgam density data were least square fitted to the linear relationship

$$\rho_a = A\, N_{Zn} + \rho_T \,, \tag{51}$$

where

ρ_a = density of amalgam,

A = a constant,

N_{Zn} = atomic fraction of zinc, and

ρ_T = density of pure mercury at temperature.

Since the intercept was fixed as the value for pure mercury, only the slopes were determined by the method of least

squares. The values for A as a function of temperature are summarized in Table VII.

A quadratic equation was selected for least square fitting the density data for cadmium amalgams:

$$\rho_a = A\, N_{Cd}^2 + B\, N_{Cd} + \rho_T \,. \qquad (52)$$

The values of the constants A and B are also given in Table VII. The curves shown in Figures 18 and 19 are those calculated from equations 51 and 52, respectively.

TABLE VII

LEAST SQUARE FIT CONSTANTS FOR CADMIUM AND ZINC AMALGAM DENSITIES

Temperature	Zinc Amalgams	Cadmium Amalgams	
°C	A	A	B
50.0	-2.866	-2.658	-3.257
70.2	-2.806	-2.949	-3.184
93.2	-2.786	-2.807	-3.149
119.1	-2.758	-1.877	-3.176
150.0	-2.681	-2.495	-3.035
over-all	-2.779	-2.557	-3.160

The self-diffusion data for zinc in zinc amalgams and cadmium in cadmium amalgams are given as Appendices H and I. Average values of these coefficients are presented in Table VIII. An analysis of the errors expected and those calculated from the data is presented in Appendix J. The calculated over-all standard deviation for the self-diffusion coefficient of zinc in binary amalgams was $\pm$ 0.180 x 10^{-5} cm^2/sec, and

that for the cadmium coefficients in binary amalgams was $\pm$ 0.204 x 10^{-5} cm^2/sec.

TABLE VIII

AVERAGE VALUES OF SOLUTE SELF-DIFFUSION COEFFICIENTS IN VARIOUS CADMIUM OR ZINC AMALGAMS, VERSUS TEMPERATURE

Atomic Percent Solute	Diffusion Coefficient x 10^5, cm^2/sec, at Temp			
	50.0°C	70.2°C	93.2°C	119.1°C
Cadmium Amalgams				
0.0889	2.692	2.762	2.378	2.254
0.4741	1.914	1.932	3.394	3.256
2.309	1.705	2.591	2.182	2.801
4.626	1.657	2.785	1.880	3.597
7.382	- -	1.873	1.685	2.119
Zinc Amalgams				
0.0694	2.125	2.763	2.848	3.676
0.3597	2.171	2.361	2.930	3.981
1.750	1.816	2.452	2.646	3.137
3.473	1.645	2.096	2.450	2.780
5.609	1.646	1.880	1.928	2.148

CHAPTER V

DISCUSSION OF RESULTS

The results of the concentration cell measurements and the density determinations presented in Chapter IV are sufficiently accurate for the purpose of evaluating the molar volume and the thermodynamic factor required in equation 37. Therefore, no further elaboration on these results will be given.

Comparison of Diffusion Data with Theory

To evaluate the activation energy and entropy terms in equation 37, the data of Golik and co-workers (43,44) for the kinematic viscosity of certain cadmium and zinc amalgams were used. These data were least square fitted to equation 47 to determine the activation energy and the constant B, which is related to the activation entropy by an equation similar to equation 48.

$$B = \frac{N_o\ h\ e^{-\frac{\Delta S^*}{R}}}{N_1\ M_1 + N_2\ M_2} \quad . \tag{53}$$

In equation 53, the subscripts 1 and 2 refer to the components of the binary solution; all other terms are as in equation 48.

Figure 20 shows the activation energies of cadmium and zinc amalgams versus the volume fraction of solute in the amalgam. The data points indicated in the figure were calculated from Golik's data and in some cases differ from the reported values (43,44). Although Golik has indicated that the activation energy can be correlated to the weight fraction, this choice would not seem to be realistic in terms of the supposed physical situation. Consequently, the correlation presented here is one involving the volume fraction. The lines shown on the graph represent a linear equation through the intercept points. This gives the following expressions:

for zinc amalgams,

$$\Delta H^*_{Zn\text{-}Hg} = 2210\ \varphi_{Zn} + 559\ ; \qquad (54)$$

for cadmium amalgams,

$$\Delta H^*_{Cd\text{-}Hg} = 1150\ \varphi_{Cd} + 559\ . \qquad (55)$$

The term φ in the above equations is the volume fraction.

Since one of the necessary conditions for isoviscous solution behavior is that the activation energies be equal, one can predict the relationship between isoviscous cadmium amalgam and zinc amalgam compositions from equations 54 and 55. This gives the ratio of the volume fraction of cadmium in mercury to that of zinc in mercury as 1.92, or in terms of atomic fractions,

$$\frac{N_{Cd}}{N_{Zn}} = 1.92 \left[\frac{9.45 + 5.20\ \varphi_{Zn}}{14.03 + 1.19\ \varphi_{Zn}} \right] . \qquad (56)$$

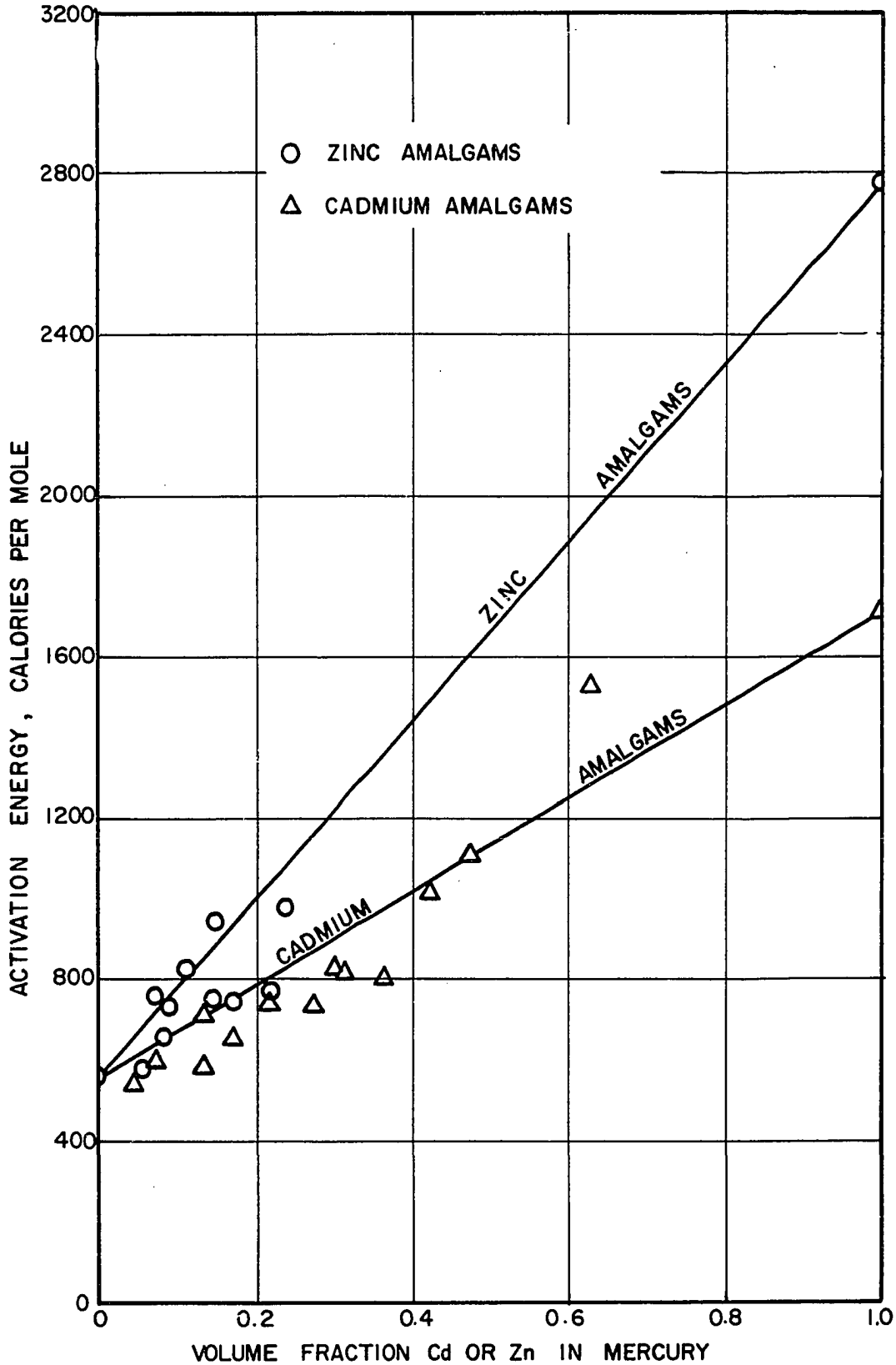


Figure 20.--Activation Energy for Viscosity Of Various Cadmium and Zinc Amalgams (Calculated from Data of Golik and Co-workers)

Equation 56 is in good agreement with the earlier prediction of isoviscous amalgam compositions based on the experimentally determined values shown in Figure 14.

The other necessary condition for isoviscous behavior, assuming equation 47 to correctly represent the situation, is that the values for B must be equal. Therefore, one can determine the concentration ratio of the solute components based on the B values and use this as a check against that obtained from the activation energies. The B quantities for various cadmium and zinc amalgams were calculated from Golik's data and are shown in Figure 21. These B values were least square fitted as a linear function of the volume fraction for a fixed intercept equal to the B for pure mercury. The resulting expressions were:

for zinc amalgams,

$$B_{Zn-Hg} \times 10^4 = 4.396 + 3.736\ \varphi_{Zn}\ ; \qquad (57)$$

for cadmium amalgams,

$$B_{Cd-Hg} \times 10^4 = 4.396 + 2.013\ \varphi_{Cd}\ . \qquad (58)$$

From equations 57 and 58, the ratio of φ_{Cd} to φ_{Zn} is determined to be 1.86. This calculation of the concentration ratio is in good agreement with the value of 1.92 calculated from the activation energies.

As an attempt to evaluate the effect of solute composition on the parameter b in equation 46, the diffusion data were least square fitted to a linear relation between the

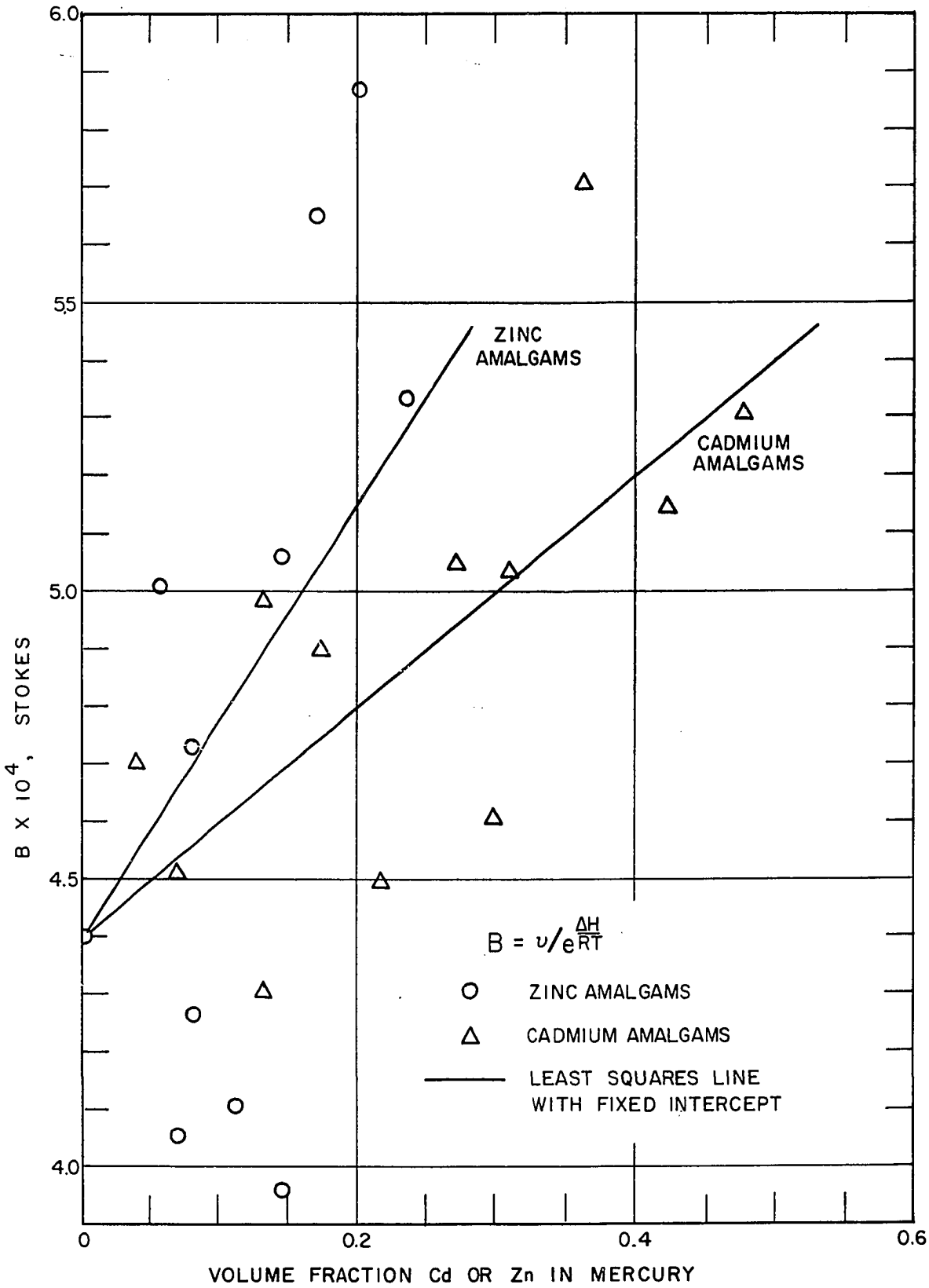


Figure 21.--Constant from Kinematic Viscosity Equation for Cadmium and Zinc Amalgams (Calculated from Data of Golik and Co-workers)

logarithm of the coefficient and the reciprocal of the absolute temperature in order to smooth the data for purposes of the calculation. Using this empirical expression of the data and equation 37, values of a_3 were determined. Subsequently, from equation 46, the values of b were calculated and these are given in Table IX.

TABLE IX

VALUES OF THE PARAMETER b CALCULATED FROM AMALGAM DIFFUSION DATA

Atomic Percent Solute	b
Zinc Amalgams	
0.0694	0.405
0.360	0.406
1.750	0.406
3.473	0.409
5.609	0.377
Cadmium Amalgams	
0.0889	0.345
0.474	0.454
2.309	0.450
4.626	0.444
7.382	0.471

From the results presented in Table IX, it can be observed that the experimental data are not sufficiently accurate to delineate the influence of solute composition on the parameter b. However, an average value for b may be

determined for each of the amalgam systems. Neglecting the two values in Table IX which deviate so greatly (that is, the values at 5.609 atomic percent zinc and at 0.0889 atomic percent cadmium), the average b value is 0.406 for the zinc amalgams and is 0.455 for the cadmium amalgams. The magnitude of the parameter b for the pure liquid metals systems was given as 0.419 in Chapter II.

Figures 22 through 31 show the comparison between the experimentally determined solute self-diffusion coefficients for these amalgam systems and the theoretical prediction from equation 37. The average value of the measured coefficients is indicated by the use of circles, and the spread of the data is shown by the vertical bars. The number of data points is indicated beside each data group.

The consistent errors in the measured diffusion coefficients are more difficult to estimate than the random errors discussed in Appendix J. The components of the consistent error are generally subject to fluctuations and these changes may or may not be random. The consistent error is a measure of the accuracy of the measurements while the random errors affect the precision of such measurements. The magnitude of consistent errors is usually a great problem in the experimental determination of diffusion coefficients in liquids.

Convection and vibration effects probably comprise a large portion of the consistent error for most liquid diffusion systems. The vibration present in the diffusion

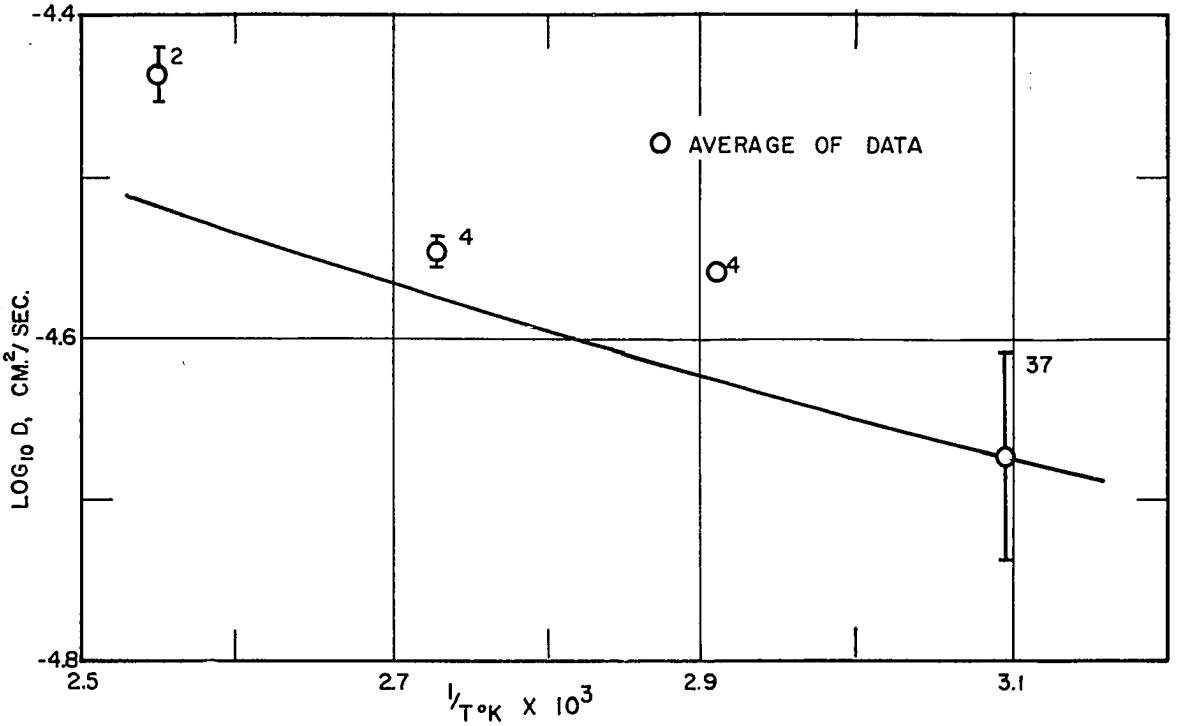
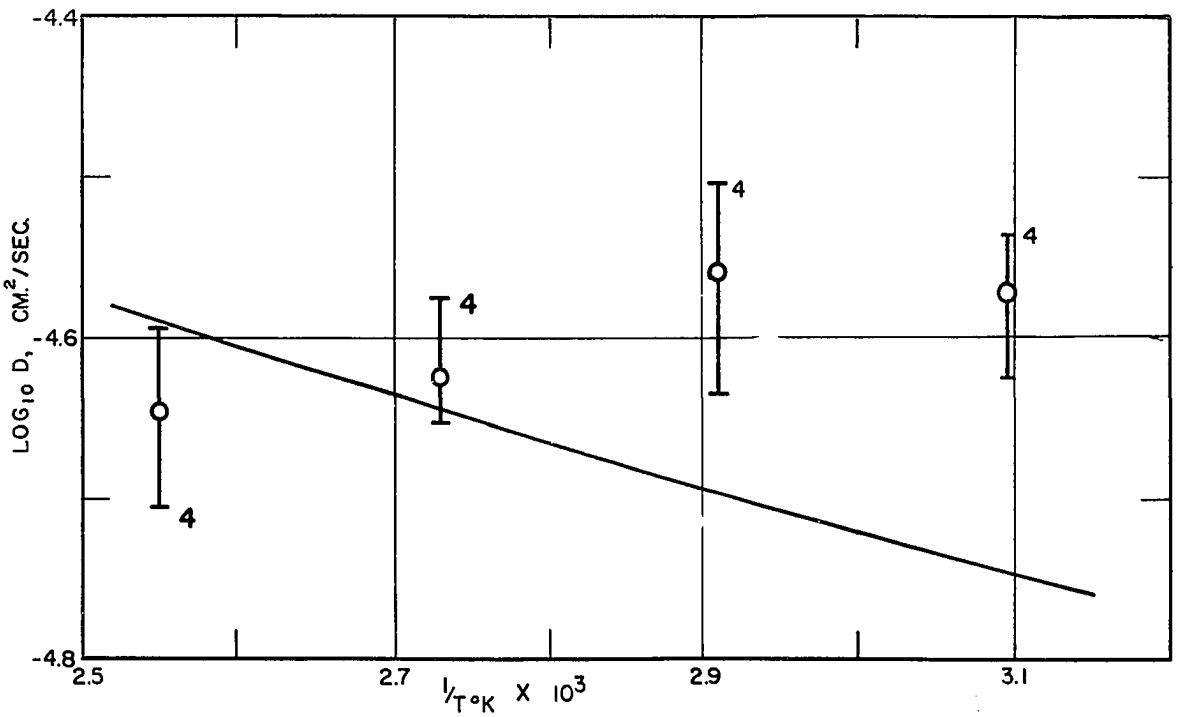


Figure 22.--Temperature Dependence of the Self-Diffusion Coefficient for Zinc in 0.0694 Atomic Percent Zinc Amalgam, Curve Calculated from Theory

Figure 23.--Temperature Dependence of the Self-Diffusion Coefficient for Cadmium in 0.0889 Atomic Percent Cadmium Amalgam, Curve Calculated from Theory



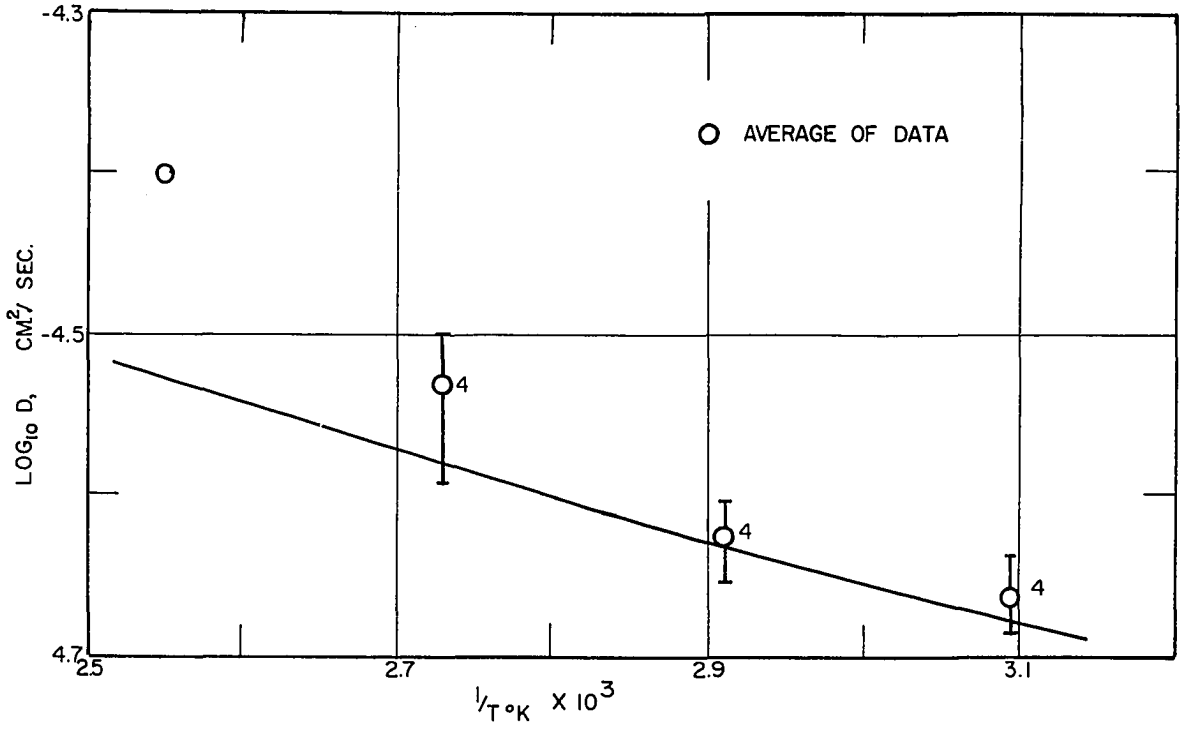
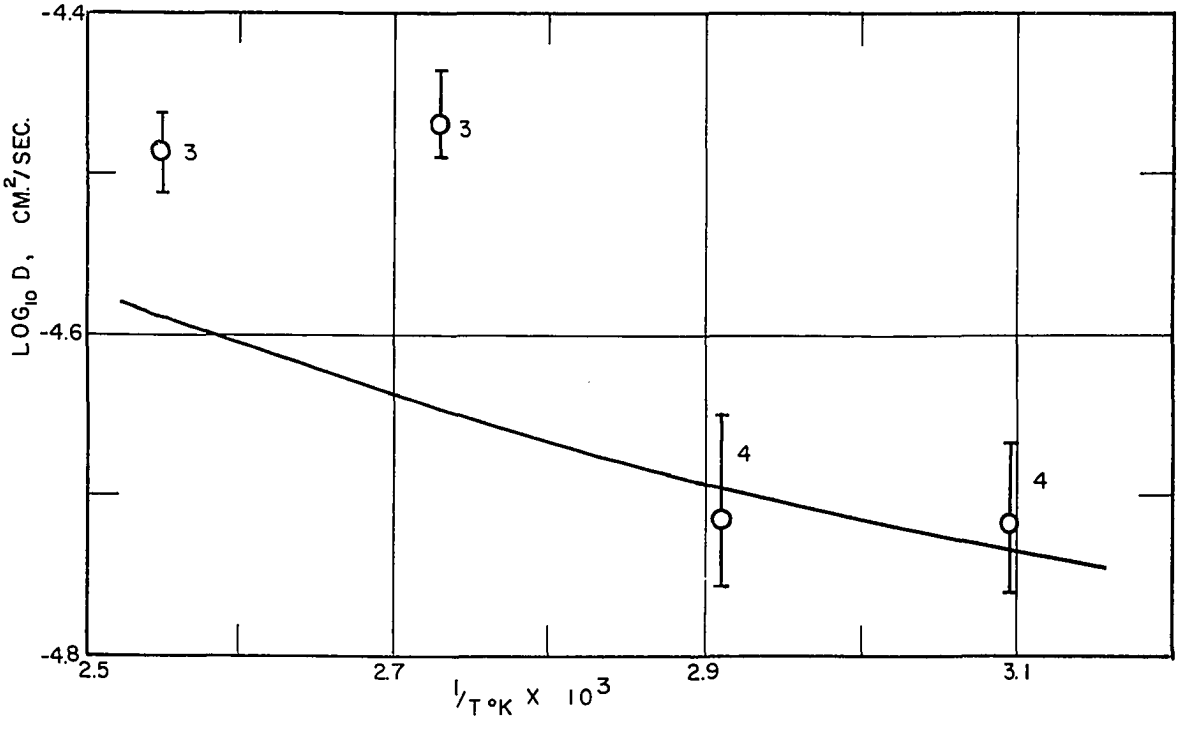


Figure 24.--Temperature Dependence of the Self-Diffusion Coefficient for Zinc in 0.360 Atomic Percent Zinc Amalgam, Curve Calculated from Theory

Figure 25.--Temperature Dependence of the Self-Diffusion Coefficient for Cadmium in 0.474 Atomic Percent Cadmium Amalgam, Curve Calculated from Theory



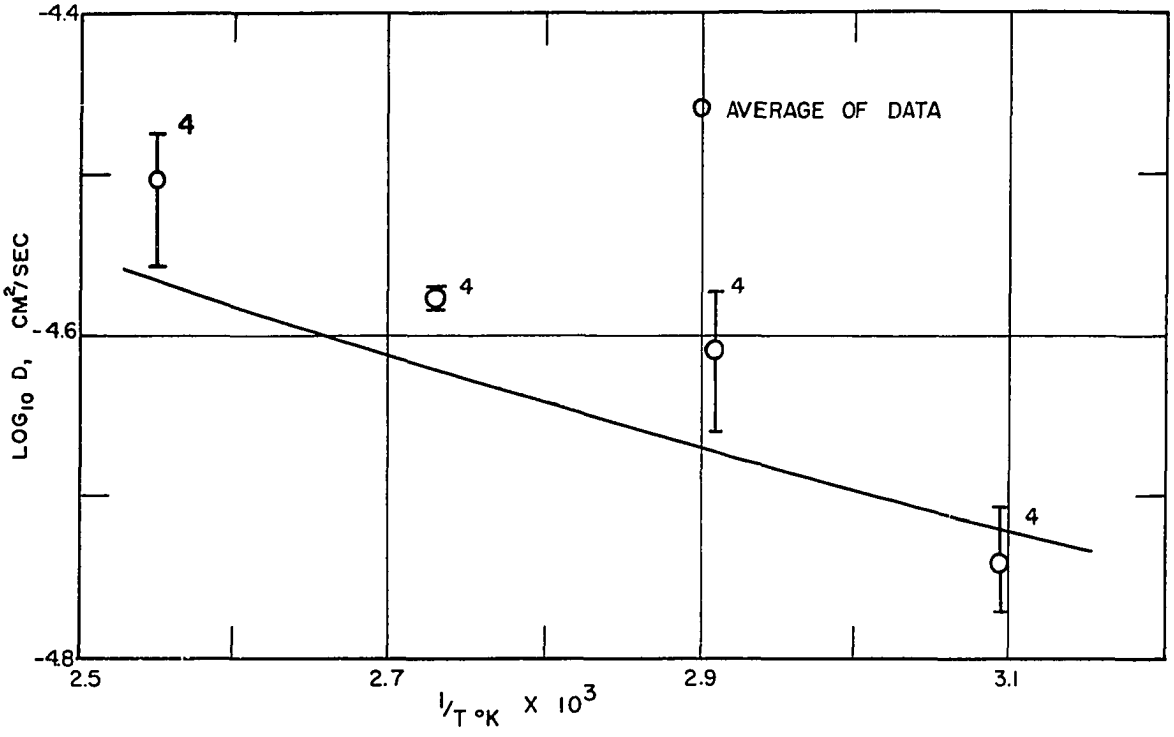
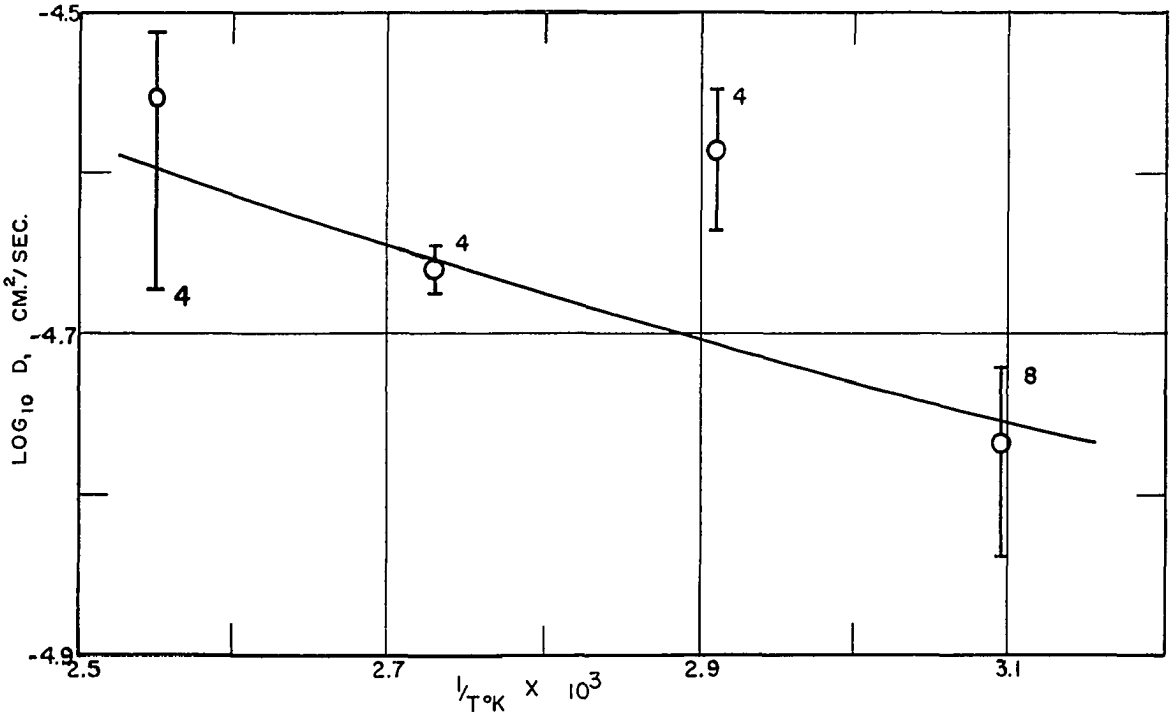


Figure 26.--Temperature Dependence of the Self-Diffusion Coefficient for Zinc in 1.750 Atomic Percent Zinc Amalgam, Curve Calculated from Theory

Figure 27.--Temperature Dependence of the Self-Diffusion Coefficient for Cadmium in 2.309 Atomic Percent Cadmium Amalgam, Curve Calculated from Theory



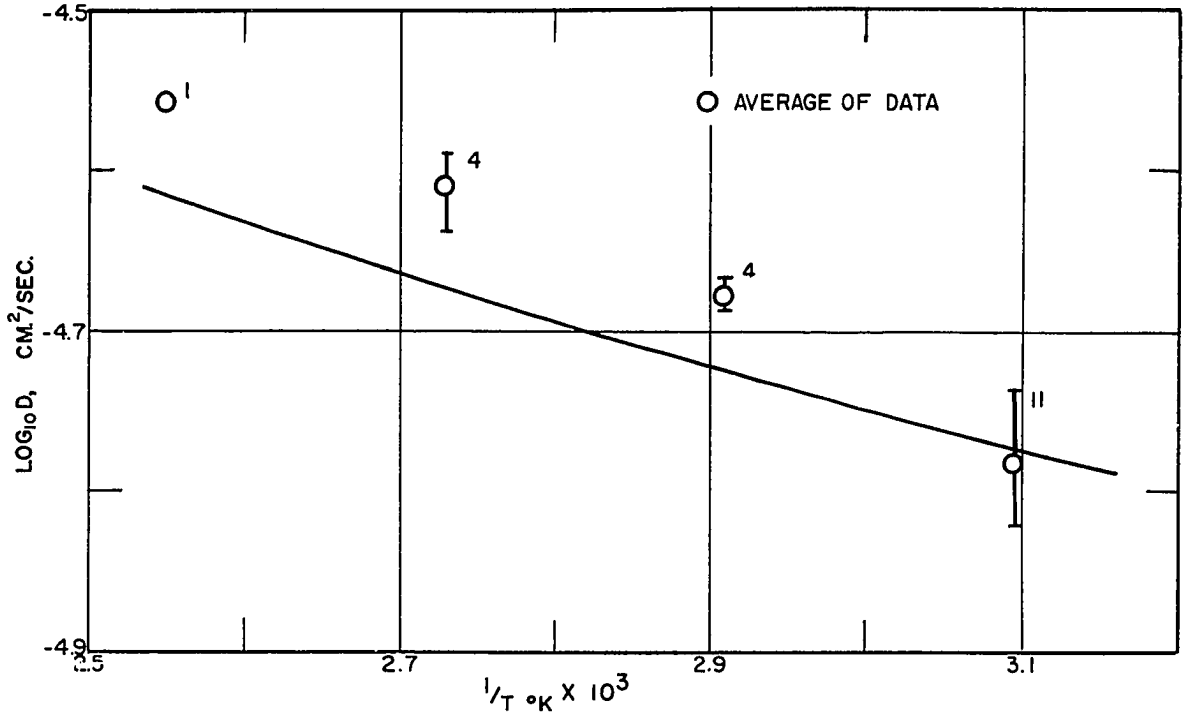
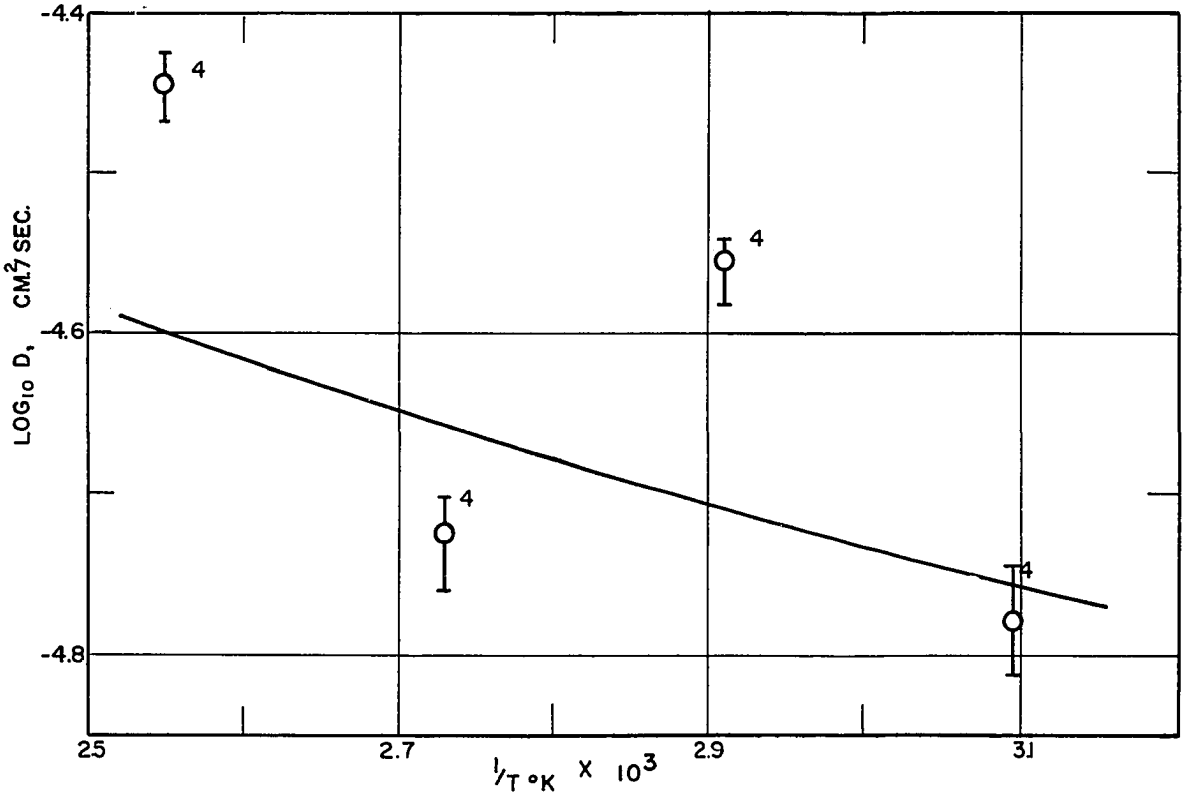


Figure 28.--Temperature Dependence of the Self-Diffusion Coefficient for Zinc in 3.473 Atomic Percent Zinc Amalgam, Curve Calculated from Theory

Figure 29.--Temperature Dependence of the Self-Diffusion Coefficient for Cadmium in 4.626 Atomic Percent Cadmium Amalgam, Curve Calculated from Theory



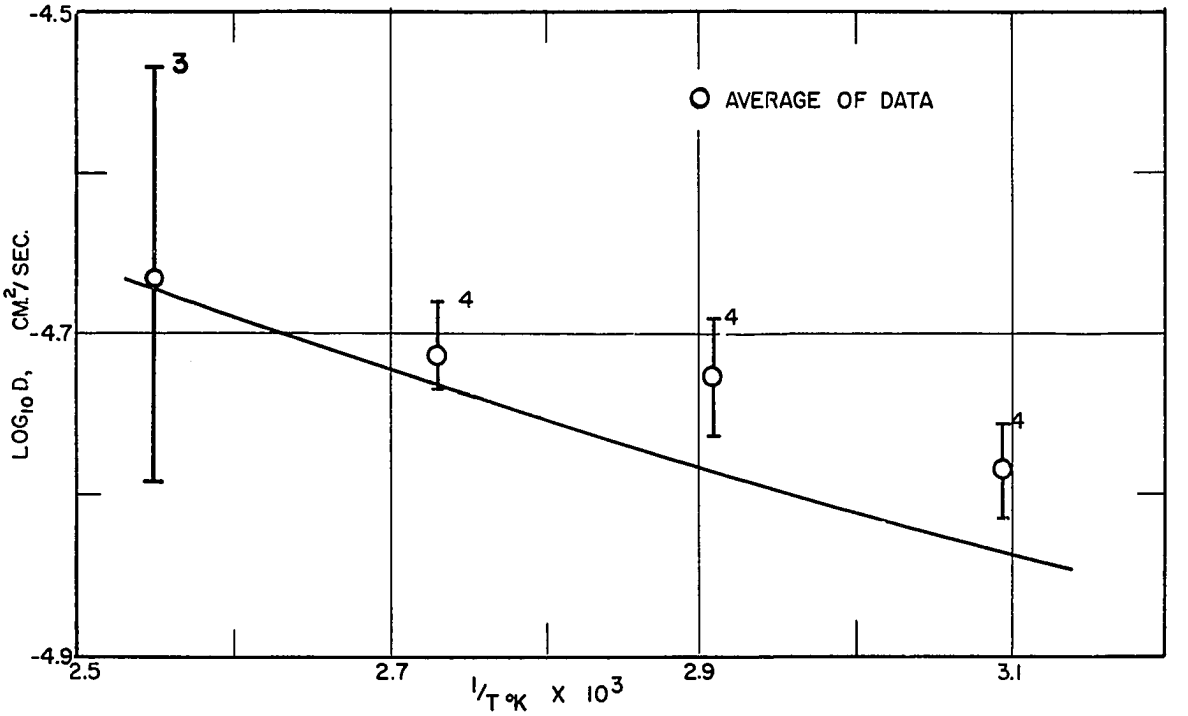
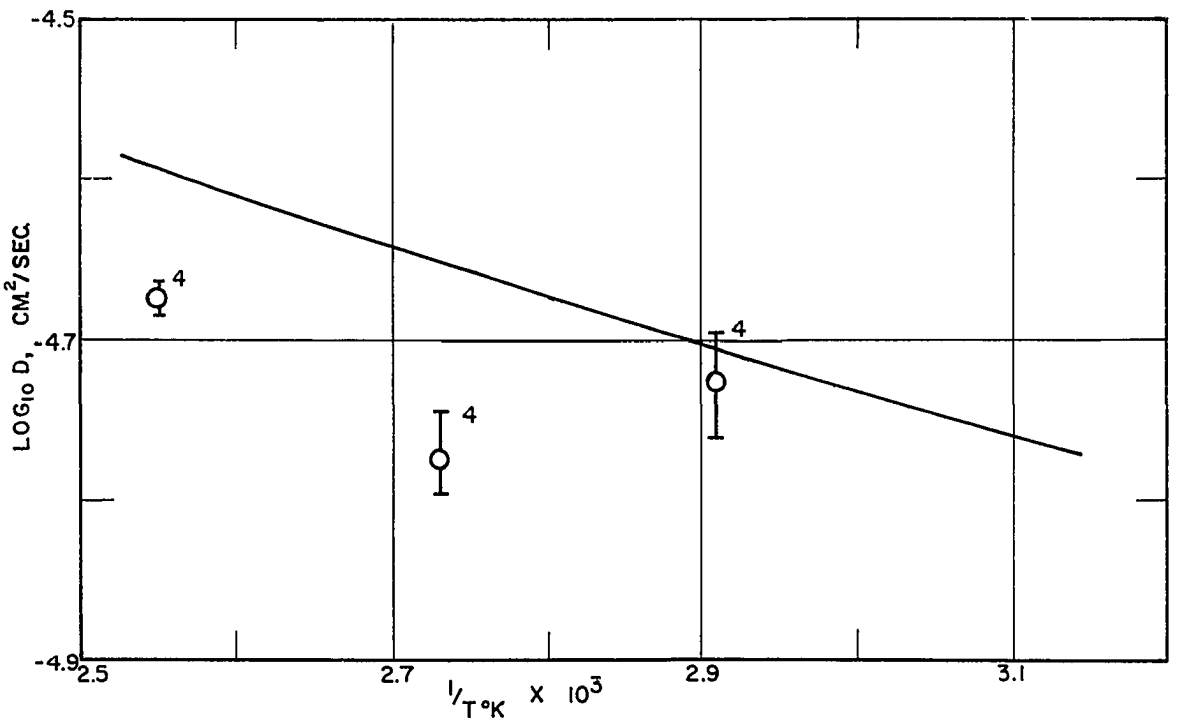


Figure 30.--Temperature Dependence of the Self-Diffusion Coefficient for Zinc in 5.609 Atomic Percent Zinc Amalgam, Curve Calculated from Theory

Figure 31.--Temperature Dependence of the Self-Diffusion Coefficient for Cadmium in 7.382 Atomic Percent Cadmium Amalgam, Curve Calculated from Theory



apparatus may, for example, arise from mixers, drive motors and shafts, and other equipment or operations within the laboratory and building. The possible influence of convection effects on the diffusion data was previously discussed in Chapter III.

The structural parameter b was found to be constant for pure liquid metals as postulated in this work. The change in the parameter calculated for the solute in amalgams studied is in agreement with other data regarding the non-ideal behavior of these systems, such as the investigations discussed in Chapters 11 and 19 of reference 18. The fact that the value of b is less than that for pure components in the case of the zinc amalgams is consistent with the postulate that the zinc in these amalgams shows a tendency to form diatomic molecules. In other words, the interatomic forces between the zinc atoms are somewhat greater than those between the zinc and mercury atoms.

In the case of the cadmium amalgams, however, the parameter b is larger in value than for the pure components. This observation is in accord with the postulate that the cadmium and mercury atoms show some indication of compound formation in these amalgams. Again, in terms of interatomic forces, this would indicate the attraction between cadmium and mercury atoms is greater than the cadmium to cadmium or mercury to mercury forces of attraction.

CHAPTER VI

CONCLUSIONS

Concentration cell potentials for various cadmium and zinc amalgams were measured at temperatures of 50.0, 70.2, and 93.2°C. The amalgam compositions used ranged from 0.066 atomic percent zinc and from 0.105 atomic percent cadmium up to the equilibrium solubility at each temperature. The equilibrium solubilities determined in this investigation are in good agreement with the majority of previous measurements.

The amalgam densities were evaluated over a range of temperatures from 50.0 to 150.0°C. Amalgams of 0.058 to 5.61 atomic percent zinc and of 0.105 to 10.00 atomic percent cadmium were used for these determinations.

Measurements of solute self-diffusion coefficients in various binary mercury-rich amalgams of cadmium and zinc were made at temperatures from 50.0 to 119.1°C. These amalgam systems varied in composition from 0.089 to 7.38 atomic percent cadmium and from 0.069 to 5.61 atomic percent zinc.

A new theoretical model for diffusion was developed in this research which indicates the activation energy for

diffusion is precisely that for viscosity. This equation was shown to describe diffusion phenomena very well in the case of pure liquid metals. Further, it was shown that the self-diffusion in these liquid metals could be correlated to a constant geometrical parameter defined as the ratio of the diffusing particle radius to the interatomic spacing.

The new theoretical prediction adequately describes the solute self-diffusion behavior in the various cadmium and zinc amalgams studied. It was observed that the geometrical parameter for the solute in these amalgam systems relative to that for pure liquid metals is consistent with the deviations from ideal behavior shown by these amalgams.

It is hoped that the results of this research of diffusion phenomena in liquid metals will in some means contribute to the understanding of these and related phenomena in liquids.

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APPENDIX A

Nomenclature

NOMENCLATURE

English Letters

a = sphere diameter

a = thermodynamic activity

a_3 = proportionality constant in the diffusion equation

A = area

b = r/d, ratio of atomic radius to interatomic distance

B = constant in the kinematic viscosity equation

c = concentration of the diffusing species at any point and time

c_f = final average concentration in capillary after diffusion

c_o = initial uniform concentration in capillary

c_r = concentration in reservoir

C = concentration

C_f' = final capillary concentration, cpm, uncorrected for isotope decay and instrument variation

C_o' = initial capillary concentration, cpm, uncorrected for volume expansion

d = interatomic spacing, angstroms

D = self-diffusion coefficient, cm^2/sec

D_o = constant in Arrhenius equation, cm^2/sec

E = concentration cell potential, volts or millivolts

F = correction factor for isotope decay and instrument variation

F = force acting on a diffusing atom

F = force acting on the sphere (Stokes' law)

F = 23,074 calories/volt-equivalent, used in calculating thermodynamic activities from emf data

G = factor correcting for volume expansion of amalgam between capillary filling temperature of 25°C and the diffusion temperature

h = Plank's constant, 6.62×10^{-27} erg-sec

ΔH^* = activation energy for viscosity, cal/mole

ΔH_V = latent heat of vaporization, kcal/mole

J = flux of the diffusing species

k = Boltzmann's constant, 1.38×10^{-16} erg/°K

k' = frequency of backward jump of each nearest neighbor atom

k_d = frequency of relative displacement with respect to all nearest neighbors

K = force constant from data of Waser and Pauling, dynes/cm

L = inside length of capillary, cm

M = mobility, average velocity of the diffusing particle/unit force acting on the particle

M = molar concentration, moles of component per liter of solution

M = molecular weight

n = number of determinations

n = number of equivalents per mole (2 for both zinc and cadmium)

N = atomic fraction of zinc or cadmium in amalgam

No = Avogadro's number, 6.02×10^{23} atoms/gram-molecular weight

Q = activation energy, Arrhenius equation

r = radius of the diffusing particle

R = gas constant, 1.987 cal/mole-°K

ΔS^* = activation entropy for viscosity, cal/mole-°K

t = time duration of diffusion

T = absolute temperature, °K

v_A = relative velocity of atom A with respect to its nearest neighbor atoms

v_t = terminal velocity

V = molar volume, cm^3/mole

V^f = molar free volume, cm^3/mole

V_o = molar volume of unexpanded liquid, cm^3/mole

x = distance

Z = number of nearest neighbors surrounding the diffusing atom

Greek Letters

α = parameter related to curvature of potential energy versus distance curve; reciprocal Angstroms (1/Å)

β = coefficient of sliding friction between diffusing particle and the solution (Sutherland Equation)

$\beta = x/L$

γ = activity coefficient, a/N

γ = constant dependent on the geometry of the liquid structure, determined by average number of nearest neighbors surrounding the atom

$\theta = Dt/L^2$

λ = distance between adjacent lattice positions

λ_d = distance between adjacent equilibrium positions

μ = absolute viscosity

ν = kinematic viscosity

$\xi = c/c_o$

ξ = number of nearest neighbors lying in same plane as diffusing atom

$\sigma = A/d$

σ = standard deviation

$\bar{\sigma}$ = standard deviation of the average

σ_j = joint standard deviation

ψ = $(c_f-c_r)/(c_o-c_r)$

Abbreviations

Ave Ref Cnt Rate = average reference count rate from initial and final reference count rates

AZn-10 = active zinc capillary amalgam #10

BG Count = background radiation level determination

C_f', CPM-BG = final concentration, in counts/minute, minus background

C_o', CPM-BG = initial concentration, in counts/minute, minus background

Corr. Factor, F = correction factor, F, used in determining the concentration ratio

cpm = counts per minute

CPM-BG = activity in counts per minute, corrected for background count rate

CTB = constant-temperature bath

Dia = inside diameter of capillary, mm

emf = electromotive force, concentration cell potential

mc = millicuries

mv = millivolts

Ref Cnts = number of reference counts taken

Ref Cnt Time = time required to count 128,000 reference counts

RPM = revolutions per minute, capillary turning rate

Temp = temperature

APPENDIX B

Density Data

TABLE B-I

VOLUME OF PYCNOMETERS, CUBIC CENTIMETERS, AT VARIOUS TEMPERATURES

Pycnometer Number	Temperature				
	50.0°C	70.2°C	93.2°C	119.1°C	150.0°C
0	14.3863	14.3888	14.3934	14.3971	14.4001
1	14.2197	14.2220	14.2254	14.2294	14.2319
2	14.6753	14.6771	14.6810	14.6852	14.6872
4	14.6314	14.6339	14.6375	14.6426	14.6452

TABLE B-II

DENSITY MEASUREMENTS, PURE MERCURY

Data Point Number	Weight of Mercury in Pycnometer grams	Mercury Density gm/cm^3
	50.0°C	
201-0*	193.8224	13.4727
202-0	193.8261	13.4730
203-0	193.8311	13.4733
204-0	193.8260	13.4730
205-0	193.8209	13.4726
201-1	191.5834	13.4731
202-1	191.5741	13.4724
203-1	191.5738	13.4724
204-1	191.5800	13.4728
205-1	191.5887	13.4735

*Number following hyphen is the pycnometer number.

TABLE B-II--Continued

Data Point Number	Weight of Mercury in Pycnometer grams	Mercury Density gm/cm^3
201-2	197.7153	13.4726
202-2	197.7140	13.4726
203-2	197.7132	13.4725
204-2	197.7282	13.4735
205-2	197.7218	13.4731
201-4	197.1177	13.4722
202-4	197.1272	13.4729
203-4	197.1239	13.4727
204-4	197.1326	13.4732
205-4	197.1334	13.4733
	70.2°C	
206-0	193.1443	13.4232
207-0	193.1480	13.4235
208-0	193.1493	13.4236
209-0	193.1585	13.4242
210-0	193.1597	13.4243
206-1	190.9157	13.4240
207-1	190.9095	13.4235
208-1	190.9163	13.4240
209-1	190.9070	13.4234
210-1	190.9174	13.4241
206-2	197.0139	13.4232
207-2	197.0251	13.4240
208-2	197.0282	13.4242
209-2	197.0253	13.4240
210-2	197.0195	13.4236
206-4	196.4370	13.4234
207-4	196.4345	13.4232
208-4	196.4485	13.4242
209-4	196.4448	13.4240
210-4	196.4502	13.4243

TABLE B-II--Continued

Data Point Number	Weight of Mercury in Pycnometer grams	Mercury Density gm/cm^3
	93.2°C	
211-0	192.4170	13.3684
212-0	192.4186	13.3685
213-0	192.4186	13.3685
214-0	192.4153	13.3683
215-0	192.4020	13.3674
211-1	190.1629	13.3678
212-1	190.1642	13.3679
213-1	190.1659	13.3680
214-1	190.1653	13.3680
215-1	190.1804	13.3691
211-2	196.2543	13.3679
212-2	196.2598	13.3683
213-2	196.2562	13.3680
214-2	196.2641	13.3686
215-2	196.2602	13.3683
211-4	195.6707	13.3678
212-4	195.6796	13.3684
213-4	195.6817	13.3685
214-4	195.6776	13.3682
215-4	195.6751	13.3681
	119.1°C	
216-0	191.5660	13.3059
217-0	191.5671	13.3060
218-0	191.5678	13.3060
219-0	191.5640	13.3057
220-0	191.5662	13.3059
216-1	189.3315	13.3056
217-1	189.3364	13.3060
218-1	189.3373	13.3061
219-1	189.3346	13.3059
220-1	189.3363	13.3060

TABLE B-II--Continued

Data Point Number	Weight of Mercury in Pycnometer grams	Mercury Density gm/cm^3
216-2	195.3959	13.3056
217-2	195.4014	13.3060
218-2	195.3948	13.3056
219-2	195.4039	13.3062
220-2	195.4004	13.3059
216-4	194.8333	13.3059
217-4	194.8222	13.3052
218-4	194.8236	13.3052
219-4	194.8594	13.3077
220-4	194.8272	13.3055
	150.0°C	
221-0	190.5347	13.2315
222-0	190.5320	13.2313
223-0	190.5555	13.2329
224-0	190.5509	13.2326
225-0	190.5281	13.2310
221-1	188.3154	13.2319
222-1	188.2967	13.2306
223-1	188.3053	13.2312
224-1	188.3319	13.2331
225-1	188.3250	13.2326
221-2	194.3397	13.2319
222-2	194.3389	13.2318
223-2	194.3397	13.2319
224-2	194.3350	13.2316
225-2	194.3453	13.2323
221-4	193.8131	13.2339
222-4	193.7820	13.2318
223-4	193.7810	13.2317
224-4	193.7681	13.2308
225-4	193.7737	13.2312

TABLE B-III

ANALYSIS OF VARIANCE FOR PYCNOMETER CALIBRATION,
FROM MERCURY DENSITY DATA

	Sum of Squares	Degrees of Freedom	Mean Square	F Test Ratio
Column Means, Temperature	0.7243402056	4	0.18108505	603,616.83
Row Means, Pycnometer	-0.000000047	3	-0.000000016	0.0667
Interaction, Temp.×Pycno.	0.00000498	12	0.0000004	1.333
Subtotal	0.7243452433	19	0.0381234	
Error	0.00002491	80	0.0000003	
Total	0.72437015	99	0.0073168	

Conclusions: (1) At a confidence level of 0.999, the effect of temperature is significant.
(2) At a confidence level of 0.999, there is no effect of pycnometer bias.
(3) The temperature × pycnometer interaction is not significant.

TABLE B-IV

DENSITY MEASUREMENTS, ZINC AMALGAMS

Data Point Number	Atomic Percent Zinc	Weight of Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		50.0°C	
226-0*	0.0583	193.7780	13.4696
227-0	0.0583	193.7817	13.4699
228-0	0.0583	193.7748	13.4694
229-0	0.0583	193.7855	13.4701
230-0	0.0583	193.7801	13.4698
226-1	0.350	191.4240	13.4619
227-1	0.350	191.4316	13.4624
228-1	0.350	191.4308	13.4624
229-1	0.350	191.4499	13.4637
230-1	0.350	191.4283	13.4622
226-2	1.750	196.9958	13.4236
227-2	1.750	196.9929	13.4234
228-2	1.750	197.0236	13.4255
229-2	1.750	197.0108	13.4247
230-2	1.750	197.0225	13.4255
226-4	3.446	195.6192	13.3698
227-4	3.446	195.6273	13.3704
228-4	3.446	195.6275	13.3704
229-4	3.446	195.6289	13.3705
230-4	3.446	195.6447	13.3716
276-0	5.609	191.5383	13.3139
277-0	5.609	191.5407	13.3141
278-0	5.609	191.5319	13.3135
276-1	5.609	189.3119	13.3134
277-1	5.609	189.3102	13.3132
278-1	5.609	189.3106	13.3133

*Number following hyphen is the pycnometer number.

TABLE B-IV--Continued

Data Point Number	Atomic Percent Zinc	Weight of Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		70.2°C	
231-0	0.0583	193.1150	13.4212
232-0	0.0583	193.1214	13.4216
233-0	0.0583	193.1098	13.4208
234-0	0.0583	193.1094	13.4208
235-0	0.0583	193.1171	13.4213
231-1	0.350	190.7824	13.4146
232-1	0.350	190.7708	13.4138
233-1	0.350	190.7674	13.4135
234-1	0.350	190.7806	13.4145
235-1	0.350	190.7675	13.4137
231-2	1.750	196.3332	13.3768
232-2	1.750	196.3224	13.3761
233-2	1.750	196.3219	13.3761
234-2	1.750	196.3432	13.3775
235-2	1.750	196.3214	13.3760
231-4	3.446	194.9912	13.3246
232-4	3.446	194.9914	13.3246
233-4	3.446	194.9924	13.3247
234-4	3.446	194.9915	13.3246
235-4	3.446	194.9902	13.3246
279-0	5.609	190.9025	13.2674
280-0	5.609	190.8991	13.2672
281-0	5.609	190.9001	13.2673
279-1	5.609	188.6879	13.2673
280-1	5.609	188.6858	13.2672
281-1	5.609	188.6851	13.2671

TABLE B-IV--Continued

Data Point Number	Atomic Percent Zinc	Weight of Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		93.2°C	
236-0	0.0583	192.3417	13.3632
237-0	0.0583	192.3414	13.3632
238-0	0.0583	192.3516	13.3639
239-0	0.0583	192.3322	13.3625
240-0	0.0583	192.3311	13.3624
236-1	0.350	190.0413	13.3593
237-1	0.350	190.0199	13.3578
238-1	0.350	190.0193	13.3577
239-1	0.350	190.0213	13.3579
240-1	0.350	190.0186	13.3577
236-2	1.750	195.5507	13.3200
237-2	1.750	195.5330	13.3188
238-2	1.750	195.5297	13.3186
239-2	1.750	195.5452	13.3196
240-2	1.750	195.5500	13.3199
236-4	3.446	194.2621	13.2715
237-4	3.446	194.2444	13.2703
238-4	3.446	194.2236	13.2689
239-4	3.446	194.2389	13.2700
240-4	3.446	194.2398	13.2700
282-0	5.609	190.1796	13.2130
283-0	5.609	190.1753	13.2127
284-0	5.609	190.1630	13.2118
282-1	5.609	187.9748	13.2140
283-1	5.609	187.9755	13.2141
284-1	5.609	187.9573	13.2128

TABLE B-IV--Continued

Data Point Number	Atomic Percent Zinc	Weight of Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		119.1°C	
241-0	0.0583	191.4568	13.2983
242-0	0.0583	191.4688	13.2991
243-0	0.0583	191.4625	13.2987
244-0	0.0583	191.4640	13.2988
245-0	0.0583	191.4594	13.2985
241-1	0.350	189.1767	13.2948
242-1	0.350	189.1748	13.2946
243-1	0.350	189.1677	13.2941
244-1	0.350	189.1682	13.2942
245-1	0.350	189.1657	13.2940
241-2	1.750	194.6772	13.2567
242-2	1.750	194.6808	13.2569
243-2	1.750	194.6975	13.2581
244-2	1.750	194.6895	13.2575
245-2	1.750	194.6812	13.2570
241-4	3.446	193.4236	13.2096
242-4	3.446	193.4257	13.2098
243-4	3.446	193.4455	13.2111
244-4	3.446	193.4292	13.2100
245-4	3.446	193.4366	13.2105
285-0	5.609	189.3470	13.1517
286-0	5.609	189.3479	13.1518
287-0	5.609	189.3473	13.1518
285-1	5.609	187.1563	13.1528
286-1	5.609	187.1290	13.1509
287-1	5.609	187.1334	13.1512

TABLE B-IV--Continued

Data Point Number	Atomic Percent Zinc	Weight of Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		150.0°C	
246-0	0.0583	190.4424	13.2251
247-0	0.0583	190.4382	13.2248
248-0	0.0583	190.4354	13.2246
249-0	0.0583	190.4348	13.2245
250-0	0.0583	190.4310	13.2243
246-1	0.350	188.0706	13.2147
247-1	0.350	188.0459	13.2130
248-1	0.350	188.0527	13.2135
249-1	0.350	188.0606	13.2140
250-1	0.350	188.0714	13.2148
246-2	1.750	193.6229	13.1831
247-2	1.750	193.6095	13.1822
248-2	1.750	193.6251	13.1832
249-2	1.750	193.6204	13.1829
250-2	1.750	193.6410	13.1843
246-4	3.446	192.4330	13.1397
247-4	3.446	192.4285	13.1394
248-4	3.446	192.4632	13.1417
249-4	3.446	192.4362	13.1399
250-4	3.446	192.4174	13.1386
288-0	5.609	188.3837	13.0821
289-0	5.609	188.3738	13.0814
290-0	5.609	188.3710	13.0812
288-1	5.609	186.1963	13.0830
289-1	5.609	186.2013	13.0834
290-1	5.609	186.1940	13.0829

TABLE B-V

DENSITY MEASUREMENTS, CADMIUM AMALGAMS

Data Point Number	Atomic Percent Cadmium	Weight of Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		50.0°C	
251-0*	0.1047	193.7680	13.4689
252-0	0.1047	193.7783	13.4696
253-0	0.1047	193.7652	13.4687
254-0	0.1047	193.7610	13.4684
255-0	0.1047	193.7641	13.4687
251-1	0.5250	191.3284	13.4551
252-1	0.5250	191.3239	13.4548
253-1	0.5250	191.3229	13.4548
254-1	0.5250	191.3237	13.4548
255-1	0.5250	191.3303	13.4553
251-2	2.625	196.4321	13.3852
252-2	2.625	196.4323	13.3852
253-2	2.625	196.4314	13.3852
254-2	2.625	196.4348	13.3854
255-2	2.625	196.4301	13.3851
251-4	5.169	194.5668	13.2979
252-4	5.169	194.5706	13.2982
253-4	5.169	194.5657	13.2978
254-4	5.169	194.5641	13.2977
255-4	5.169	194.5649	13.2978
276-2	9.998	192.5441	13.1203
277-2	9.998	192.5387	13.1199
278-2	9.998	192.5550	13.1210
276-4	9.998	191.9794	13.1210
277-4	9.998	191.9690	13.1203
278-4	9.998	191.9765	13.1208

*Number following hyphen is the pycnometer number.

TABLE B-V--Continued

Data Point Number	Atomic Percent Cadmium	Weight of Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		70.2°C	
256-0	0.1047	193.1050	13.4205
257-0	0.1047	193.0943	13.4198
258-0	0.1047	193.0913	13.4196
259-0	0.1047	193.0980	13.4200
260-0	0.1047	193.0910	13.4195
256-1	0.5250	190.6694	13.4066
257-1	0.5250	190.6672	13.4065
258-1	0.5250	190.6693	13.4066
259-1	0.5250	190.6620	13.4061
260-1	0.5250	190.6740	13.4070
256-2	2.625	195.7670	13.3383
257-2	2.625	195.7627	13.3380
258-2	2.625	195.7682	13.3383
259-2	2.625	195.7645	13.3381
260-2	2.625	195.7652	13.3381
256-4	5.169	193.9127	13.2509
257-4	5.169	193.9277	13.2519
258-4	5.169	193.9188	13.2513
259-4	5.169	193.9136	13.2510
260-4	5.169	193.9314	13.2522
279-2	9.998	191.9200	13.0762
280-2	9.998	191.9134	13.0757
281-2	9.998	191.9208	13.0762
279-4	9.998	191.3440	13.0754
280-4	9.998	191.3510	13.0759
281-4	9.998	191.3591	13.0764

TABLE B-V--Continued

Data Point Number	Atom Percent Cadmium	Weight in Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		93.2°C	
261-0	0.1047	192.3358	13.3628
262-0	0.1047	192.3396	13.3630
263-0	0.1047	192.3298	13.3624
264-0	0.1047	192.3291	13.3623
265-0	0.1047	192.3347	13.3627
261-1	0.5250	189.9270	13.3512
262-1	0.5250	189.9288	13.3514
263-1	0.5250	189.9104	13.3501
264-1	0.5250	189.9253	13.3511
265-1	0.5250	189.9343	13.3518
261-2	2.625	195.0214	13.2839
262-2	2.625	195.0170	13.2836
263-2	2.625	195.0017	13.2826
264-2	2.625	195.0196	13.2838
265-2	2.625	194.9972	13.2823
261-4	5.169	193.1828	13.1978
262-4	5.169	193.1869	13.1981
263-4	5.169	193.1772	13.1974
264-4	5.169	193.2271	13.2008
265-4	5.169	193.1748	13.1972
282-2	9.998	191.2116	13.0244
283-2	9.998	191.2166	13.0248
284-2	9.998	191.2251	13.0253
282-4	9.998	190.6635	13.0257
283-4	9.998	190.6587	13.0254
284-4	9.998	190.6700	13.0261

TABLE B-V--Continued

Data Point Number	Atomic Percent Cadmium	Weight of Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		119.1°C	
266-0	0.1047	191.4853	13.3003
267-0	0.1047	191.4793	13.2998
268-0	0.1047	191.4793	13.2998
269-0	0.1047	191.4808	13.3000
270-0	0.1047	191.4936	13.3008
266-1	0.5250	189.0832	13.2882
267-1	0.5250	189.0929	13.2889
268-1	0.5250	189.1103	13.2901
269-1	0.5250	189.0746	13.2876
270-1	0.5250	189.0750	13.2876
266-2	2.625	194.1672	13.2220
267-2	2.625	194.1437	13.2204
268-2	2.625	194.1255	13.2191
269-2	2.625	194.1264	13.2192
270-2	2.625	194.1692	13.2221
266-4	5.169	192.3705	13.1377
267-4	5.169	192.3732	13.1379
268-4	5.169	192.3584	13.1369
269-4	5.169	192.3705	13.1377
270-4	5.169	192.3649	13.1373
285-2	9.998	190.4495	12.9688
286-2	9.998	190.4591	12.9694
287-2	9.998	190.4640	12.9698
285-4	9.998	189.9041	12.9693
286-4	9.998	189.9114	12.9698
287-4	9.998	189.9135	12.9699

TABLE B-V--Continued

Data Point Number	Atomic Percent Cadmium	Weight of Amalgam in Pycnometer grams	Amalgam Density gm/cm^3
		150.0°C	
271-0	0.1047	190.4632	13.2265
272-0	0.1047	190.4707	13.2270
273-0	0.1047	190.4735	13.2272
274-0	0.1047	190.4642	13.2266
275-0	0.1047	190.4745	13.2273
271-1	0.5250	188.0818	13.2155
272-1	0.5250	188.0991	13.2167
273-1	0.5250	188.0899	13.2161
274-1	0.5250	188.0841	13.2157
275-1	0.5250	188.1027	13.2170
271-2	2.625	193.1234	13.1491
272-2	2.625	193.1398	13.1502
273-2	2.625	193.1415	13.1503
274-2	2.625	193.1430	13.1504
275-2	2.625	193.1356	13.1499
271-4	5.169	191.3892	13.0684
272-4	5.169	191.3927	13.0686
273-4	5.169	191.3915	13.0685
274-4	5.169	191.3946	13.0688
275-4	5.169	191.4065	13.0696
288-2	9.998	189.5265	12.9042
289-2	9.998	189.5154	12.9034
290-2	9.998	189.5158	12.9035
288-4	9.998	188.9626	12.9027
289-4	9.998	188.9711	12.9033
290-4	9.998	188.9770	12.9037

APPENDIX C

Calculation of the Error in Composition Measurements

CALCULATION OF THE ERROR IN COMPOSITION MEASUREMENTS

The error in the composition determinations from emf measurements depends on the magnitude of the solute concentration in the amalgam. The slope of the logarithm of solute weight percent versus emf curve is itself a function of the composition; furthermore, depending on the composition, different potentiometer ranges were used in the measurements. A Rubicon model 2745 precision potentiometer with the specifications given below was used for all the emf measurements except those for the most dilute cadmium amalgam at temperatures above 50.0°C where a model 2732 Rubicon potentiometer with a larger millivolt range was required.

Specifications for Rubicon Model 2745 Potentiometer

Millivolt Range	Readable to	Error Limit
0 to 16.1	± 0.001 mv	0.01 mv
0 to 80.5	± 0.005 mv	0.05 mv

Galvanometer sensitivity is ± 0.002 mv in circuits with external resistance up to 15 ohms.

As an example, the following calculation gives the error in the composition determination of a 0.577 weight percent zinc amalgam. At 0.577 weight percent zinc, the slope of the emf curve at 50.0°C is -0.034 weight percent zinc per millivolt. From the specifications given above, the error in the emf measurement is found to be 0.05 millivolts. The uncertainty in the amalgam concentration is the product of these two factors which is 0.002 weight percent zinc for the case cited.

APPENDIX D

Radioactive Materials Specifications

Radioactive Materials Specifications

The radioactive isotopes used in this work were obtained from Oak Ridge National Laboratory. The following specifications were given for these materials.

TABLE D-I

SPECIFICATIONS OF RADIOISOTOPES

Item	Isotopes	
	Zinc-65	Cadmium-115m
Half Life	250 days	43 days
Chemical Form	$ZnCl_2$ in HCl	$Cd(NO_3)_2$ in HNO_3
Acidity	0.42N	0.80N
Concentration	52.50±5 mc/ml 6.50 mg Zn/ml	0.181±0.02 mc/ml 1.82 mg Cd/ml
Specific Activity	8076 mc/g	99.4 mc/g
Purity	> 99%	> 90%

The radiation energy spectrums for the isotopes listed above were determined with the counting system used in this research. These curves are given in Fig. D-1. This figure also gives the spectrums for iron-59 and cesium-137. The instrument settings for these measurements were: window width, 020 (0.2 volt); coarse gain, 16; fine gain, 1.6; and voltage, 800. Fig. D-1 shows the good linear characteristic of the amplifier, and it should be noted that this line should not intercept the origin because of a 3 volt bias.

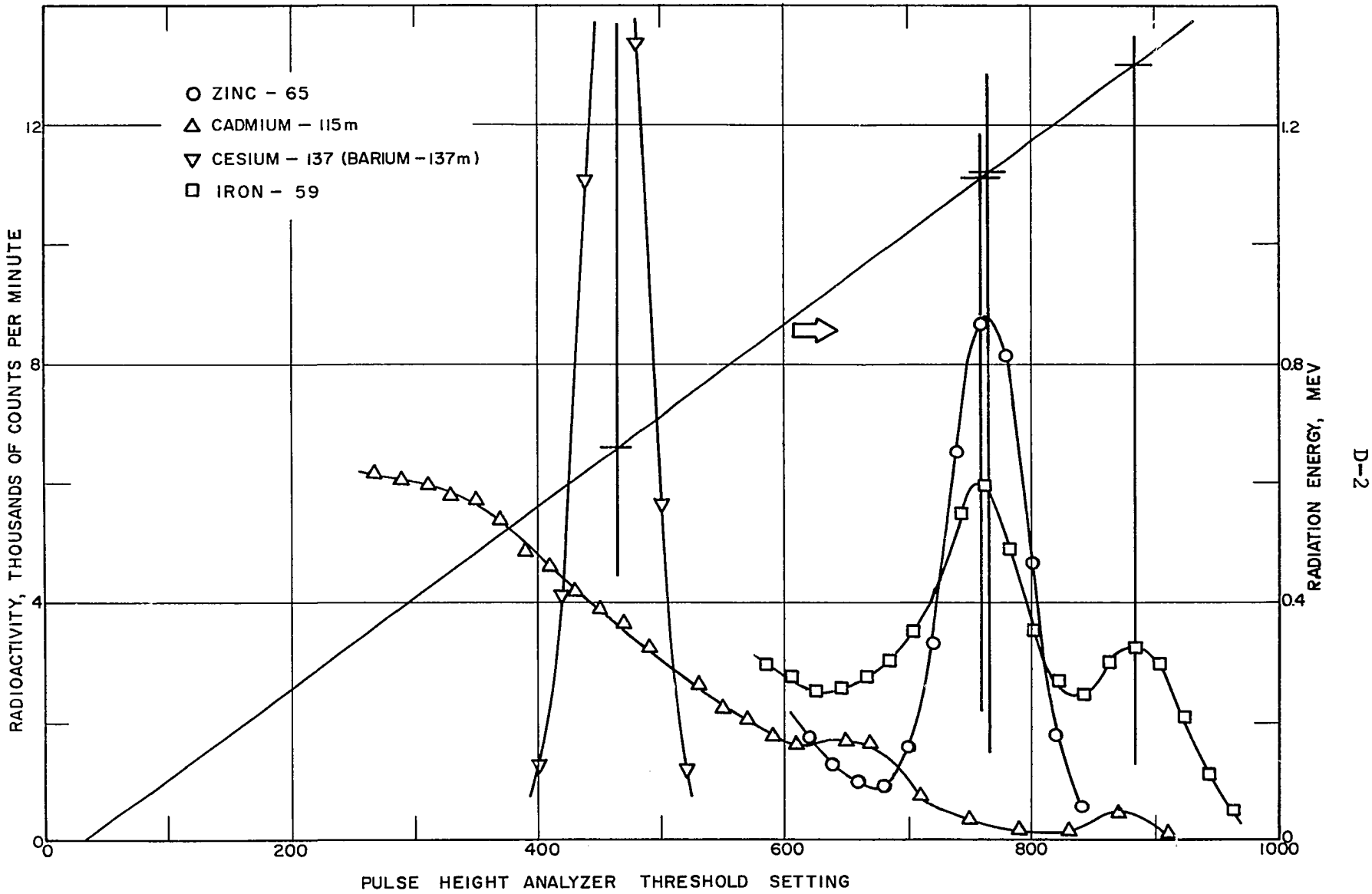


Figure D-1. Radiation Energy Spectrums for Radioactive Isotopes

APPENDIX E

Mathematical Analysis of Capillary-Reservoir Diffusion With Tabulated Values for the Solution of Fick's Equation

Mathematical Analysis of Capillary-Reservoir Diffusion

The mathematical analysis for diffusion in the geometry of the capillary-reservoir system proceeds rather easily by separation of variables assuming diffusion occurs only in the direction of the longitudinal axis of the capillary and that the system is at a constant temperature. The diffusion coefficient is assumed to be independent of the concentration of the diffusing species. The solution of Fick's equation is summarized as follows:

The quantities appearing in Fick's equation,

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial X^2} ,$$

may be transformed by letting

$\xi = \frac{c}{c_o}$, where c is the concentration of the diffusing species at any point and time and c_o is initial uniform concentration in the capillary;

$\beta = \frac{x}{L}$, where x is the distance from the open end of the capillary and L is the inside length of the capillary;

$\theta = \frac{Dt}{L^2}$, where D is the diffusion coefficient and t is the time duration of diffusion.

After introduction of these dimensionless variables, Fick's equation becomes

$$\frac{\partial \xi}{\partial \theta} = \frac{\partial^2 \xi}{\partial \beta^2}$$

and this is then solved by separation of variables. The initial and boundary conditions are:

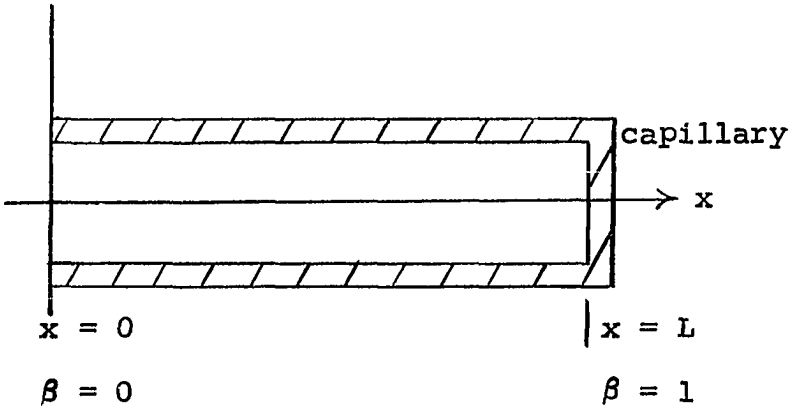
$\xi = 1$ at $0 \leq \beta \leq 1$ for $\theta = 0$

ξ is finite at all β for all θ

$\frac{\partial \xi}{\partial \beta} = 0$ at $\beta = 1$ for all θ

$\xi = k$ at $\beta \leq 0$ for all θ,

where $k = \frac{c_r}{c_o}$ and c_r is the reservoir concentration, which is assumed constant.



The solution so obtained is

$$\xi = k + \frac{(1 - k)4}{\pi} \sum_{n=0}^{\infty} \frac{1}{(2n + 1)} e^{-\frac{(2n + 1)^2 \pi^2 \theta}{4}} \sin \frac{(2n + 1)\pi}{2} \beta.$$

Since

$$c_f \equiv \frac{1}{L} \int_0^L c \, dx,$$

the above series (after integration with respect to β) becomes

$$\psi = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n + 1)^2} e^{- \frac{(2n + 1)^2 \pi^2}{4} \theta},$$

where ψ is defined as $\frac{c_f - c_r}{c_o - c_r}$ and c_f is the final average concentration in the capillary after diffusion.

Tabulated Values of the Solution of Fick's Equation for the Capillary-Reservoir System.

The solution of Fick's equation as indicated above leads to an infinite series expression with the diffusion coefficient contained in an exponential term of the series. The difficulty encountered in solving the equation for the diffusion coefficient has usually resulted in the use of no more than the first several terms of the series or in the use of other approximate solutions.

To facilitate the accurate use of this solution from Fick's equation, the series solution was programmed for the IBM-650 computer; and the diffusion coefficient was evaluated as a function of an experimentally measurable concentration ratio. The programmed equation determined ψ values for a range of θ values from 0.0001 to 0.3600 in increments of 0.0001.

The tabulated solution lists values of ψ versus θ, the number of terms (n) used to compute ψ, and the value of the first term (T) neglected; that is, the (n + 1)th term, so

that an estimate of the error in neglecting the remaining terms of the series may be made.

The values as listed in the table are in the floating decimal point system. This system uses the last two digits on the right-hand end of the usual ten-digit IBM word to designate the magnitude of the value recorded as the first eight digits. This magnitude designation is similar to "scientific notation" in that the last two digits indicate powers of ten. The following examples are given to demonstrate the floating point system.

Floating Point	Scientific Notation
8718408850	8.7184088×10^{-1}
1290000049	1.2900000×10^{-2}
2833075343	2.8330753×10^{-8}
9000000051	9.0×10^{0}
8200000052	8.2×10^{1}

Thus, it may be seen that as the magnitude designations decrease from 50, each step represents another power of 10 less for the magnitude. As the numbers increase from 50, each step increases the magnitude by a factor of 10.

The procedure for computing ψ involved selection of a θ value followed by calculation and addition of each term. As each successive term was added to the series summation, the program checked to determine if the addition of this term would change the value of the sum in the first eight digits. If the term did not change the value computed to that point for ψ, the term was recorded as the first term neglected in

the series. Therefore, the ψ values are significant to eight figures and the θ values are exact.

Hence, the use of the tables involves finding the experimentally determined value for ψ in the tables and reading the corresponding θ value. Knowing θ, L, and t, one may calculate the diffusion coefficient as

$$D = \frac{\theta L^2}{t}$$

from the defining equation for the dimensionless variable θ.

TABLE E-I

SOLUTION OF FICK'S EQUATION FOR CAPILLARY-RESERVOIR DIFFUSION, TABULATED VALUES

Table E-I comprises the remainder of this appendix.

ψ	θ	n	T
9867164250	1000000047	8200000052	4442824243
9840421950	2000000047	6000000052	4973161043
9804561550	3000000047	5100000052	3662790743
9774327350	4000000047	4500000052	3407374743
9747688750	5000000047	4000000052	4652817443
9723606750	6000000047	3700000052	4298327143
9701460350	7000000047	3500000052	3262491443
9680846350	8000000047	3300000052	3159343743
9661467150	9000000047	3100000052	3745697443
9643174750	10000000048	3000000052	2766916443
9625760450	11000000048	2800000052	4555491043
9609116250	12000000048	2700000052	4260337043
9593156150	13000000048	2600000052	4348749443
9577799850	14000000048	2500000052	4817403343
9562983450	15000000048	2500000052	2535685443
9548650150	16000000048	2400000052	3184426343
9534757850	17000000048	2300000052	4282574143
9521270750	18000000048	2300000052	2483093943
9508151650	19000000048	2200000052	3721140043
9495376250	20000000048	2200000052	2257776243
9482912950	21000000048	2100000052	3734480943
9470743850	22000000048	2100000052	2366439543
9458851250	23000000048	2000000052	4279447143
9447209850	24000000048	2000000052	2826541043
9435813250	25000000048	2000000052	1866908043
9424637050	26000000048	1900000052	3803694943
9413667350	27000000048	1900000052	2613476943
9402919450	28000000048	1900000052	1795690243
9392346850	29000000048	1800000052	4067448243
9381962250	30000000048	1800000052	2901503143
9371746550	31000000048	1800000052	2069777143
9361685950	32000000048	1800000052	1476469643
9351796850	33000000048	1700000052	3801956643
9342048950	34000000048	1700000052	2810211243
9332442050	35000000048	1700000052	2077162243
9322973850	36000000048	1700000052	1535330443
9313636050	37000000048	1600000052	4418350043
9304421450	38000000048	1600000052	3377266443
9295328550	39000000048	1600000052	2581489643
9286351450	40000000048	1600000052	1973219643
9277486550	41000000048	1600000052	1508275043
9268727250	42000000048	1500000052	4922329243
9260073650	43000000048	1500000052	3883218543
9251518050	44000000048	1500000052	3063467143
9243061350	45000000048	1500000052	2416766143
9234697950	46000000048	1500000052	1906582443
9226423650	47000000048	1500000052	1504100943
9218238450	48000000048	1500000052	1186583743
9210135250	49000000048	1400000052	4563693543
9202116250	50000000048	1400000052	3708646843
9194175950	51000000048	1400000052	3013671543
9186311250	52000000048	1400000052	2448929743
9178529450	53000000048	1400000052	1990014943
9170815350	54000000048	1400000052	1617099443
9163172450	55000000048	1400000052	1314064643
9155600950	56000000048	1400000052	1067817843
9148093450	57000000048	1300000052	4836614543
9140654050	58000000048	1300000052	4040393143
9133277850	59000000048	1300000052	3375245343
9125963250	60000000048	1300000052	2819600043
9118708650	61000000048	1300000052	2355424643
9111515650	62000000048	1300000052	1967665843
9104376950	63000000048	1300000052	1643739843
9097287450	64000000048	1300000052	1373139943
9090273850	65000000048	1300000052	1147088543
9083302150	66000000048	1300000052	9582496042
9076384250	67000000048	1300000052	8004990042
9069516850	68000000048	1200000052	4465914943
9062698250	69000000048	1200000052	3827692343
9055930850	70000000048	1200000052	3280674743
9049212850	71000000048	1200000052	2811834443
9042540250	72000000048	1200000052	2409993643
9035914650	73000000048	1200000052	2065580043
9029333550	74000000048	1200000052	1770388643
9022797150	75000000048	1200000052	1517381443
9016304550	76000000048	1200000052	1300532943
9009852350	77000000048	1200000052	1114673043
9003445650	78000000048	1200000052	9553754242
8997075250	79000000048	1200000052	8188421942
8990748050	80000000048	1200000052	7016209942

ψ	θ	n	T
8984459650	81000000048	1100000052	4641019343
8978211850	82000000048	1100000052	4246641543
8972000450	83000000048	1100000052	3726754643
8965825450	84000000048	1100000052	3272480743
8959686250	85000000048	1100000052	2872042243
8953585050	86000000048	1100000052	2520600943
8947519550	87000000048	1100000052	2212166443
8941488150	88000000048	1100000052	1941471643
8935491550	89000000048	1100000052	1703902743
8929526650	90000000048	1100000052	1495402443
8923596450	91000000048	1100000052	1312416843
8917696750	92000000048	1100000052	1151621243
8911832650	93000000048	1100000052	1010878143
8906005750	94000000048	1100000052	887180624?
8900194450	95000000048	1100000052	77866?3042
8894419150	96000000048	1100000052	683343404?
8888677450	97000000048	1100000052	599?2?6242
8882963450	98000000048	1100000052	526?395842
8877277?50	99000000048	1000000052	4?5?57843
8871621450	100000000049	1000000052	426486?843
8865993350	101000000049	1000000052	3?2?1?0943
8860394750	102000000049	1000000052	342?72?943
8854822450	103000000049	1000000052	3077014?43
8849276950	104000000049	1000000052	275?7?6743
8843756950	105000000049	1000000052	247?233643
8838264550	106000000049	1000000052	222?034843
8832796450	107000000049	1000000052	199114?043
8827355850	108000000049	1000000052	178?8?6043
8821939650	109000000049	1000000052	1601730943
8816549350	110000000049	1000000052	1436590943
8811180150	111000000049	1000000052	128847?043
8805836850	112000000049	1000000052	1155632643
8800517950	113000000049	1000000052	1036485?43
8795222450	114000000049	1000000052	9296230?42
8789948950	115000000049	1000000052	8337771442
8784699650	116000000049	1000000052	747?139042
8779472350	117000000049	1000000052	670?126842
8774268450	118000000049	1000000052	6015617242
8769084050	119000000049	1000000052	5395401142
8763925550	120000000049	1000000052	483912?242
8758784950	121000000049	1000000052	434?206642
8753666150	122000000049	1000000052	389?723142
8748567?50	123000000049	9000000051	4834616343
8743491950	124000000049	9000000051	4422604443
8738434?50	125000000049	9000000051	4045?00843
8733399550	126000000049	9000000051	3700921343
8728382950	127000000049	9000000051	3385524443
8723384950	128000000049	9000000051	3097005843
8718406850	129000000049	9000000051	2833075343
8713451450	130000000049	9000000051	2591637243
8708512650	131000000049	9000000051	2370772443
8703592450	132000000049	9000000051	2168732343
8698691750	133000000049	9000000051	1983910343
8693808850	134000000049	9000000051	1814839143
8688943050	135000000049	9000000051	1660176543
8684095850	136000000049	9000000051	1518694043
8679266450	137000000049	9000000051	1389267743
8674456550	138000000049	9000000051	1270872643
8669662850	139000000049	9000000051	1162567343
8664885450	140000000049	9000000051	1063491943
8660124850	141000000049	9000000051	972659894?
8655382250	142000000049	9000000051	8899506942
8650655750	143000000049	9000000051	8141080342
8645946350	144000000049	9000000051	7447287542
8641253150	145000000049	9000000051	6812620842
8636574550	146000000049	9000000051	623204104?
8631914550	147000000049	9000000051	5700938642
8627269250	148000000049	9000000051	5215052642
8622640050	149000000049	9000000051	4770656242
8618026250	150000000049	9000000051	436409?342
8613427150	151000000049	9000000051	399218174?
8608843350	152000000049	9000000051	3651963242
8604272650	153000000049	9000000051	334073?642
8599720350	154000000049	9000000051	3056033?42
8595161150	155000000049	9000000051	2795594742
8590656550	156000000049	9000000051	255?350642
8586149750	157000000049	8000000051	4753660943
8581650850	158000000049	8000000051	4426490743
8577169550	159000000049	8000000051	4121837443
8572701650	160000000049	8000000051	3838152243

ψ	θ	n	T
8568248450	1610000049	8000000051	3573991743
8563808950	1620000049	8000000051	3328011843
8559382350	1630000049	8000000051	3098964643
8554971250	1640000049	8000000051	2885678543
8550572350	1650000049	8000000051	2687071843
8546185550	1660000049	8000000051	2502134243
8541814150	1670000049	8000000051	2329924943
8537454850	1680000049	8000000051	2169568043
8533107750	1690000049	8000000051	2020247543
8528774450	1700000049	8000000051	1881204143
8524452550	1710000049	8000000051	1751730343
8520144350	1720000049	8000000051	1631169243
8515851550	1730000049	8000000051	1518904043
8511567750	1740000049	8000000051	1414365543
8507296050	1750000049	8000000051	1317021943
8503037250	1760000049	8000000051	1226377943
8498789850	1770000049	8000000051	1141972543
8494555450	1780000049	8000000051	1063376343
8490332450	1790000049	8000000051	9901894842
8486121450	1800000049	8000000051	9220397642
8481922750	1810000049	8000000051	8585812842
8477734550	1820000049	8000000051	7994894542
8473559250	1830000049	8000000051	7444646442
8469393750	1840000049	8000000051	6932268942
8465239550	1850000049	8000000051	6455155742
8461096750	1860000049	8000000051	6010879942
8456965350	1870000049	8000000051	5597181342
8452845950	1880000049	8000000051	5211955742
8448735550	1890000049	8000000051	4853242942
8444637350	1900000049	8000000051	4519223242
8440550450	1910000049	8000000051	4208187942
8436474150	1920000049	8000000051	3918559542
8432407450	1930000049	8000000051	3648865142
8428350550	1940000049	8000000051	3397732042
8424305850	1950000049	8000000051	3163683342
8420271650	1960000049	8000000051	2946129142
8416247150	1970000049	8000000051	2743361942
8412231550	1980000049	8000000051	2554550242
8408226550	1990000049	8000000051	2378733442
8404232850	2000000049	8000000051	2215019442
8400248950	2010000049	8000000051	2062570842
8396273950	2020000049	8000000051	1920614442
8392308550	2030000049	8000000051	1788428242
8388353050	2040000049	8000000051	1665339742
8384407950	2050000049	8000000051	1550722842
8380471850	2060000049	7000000051	4797890743
8376546250	2070000049	7000000051	4538784943
8372627950	2080000049	7000000051	4293676443
8368722650	2090000049	7000000051	4061800543
8364823850	2100000049	7000000051	3842450643
8360935550	2110000049	7000000051	3634942743
8357056150	2120000049	7000000051	3438644443
8353186450	2130000049	7000000051	3252943743
8349325750	2140000049	7000000051	3077274643
8345473050	2150000049	7000000051	2911089343
8341630150	2160000049	7000000051	2753881443
8337795350	2170000049	7000000051	2605160743
8333969450	2180000049	7000000051	2464473943
8330153350	2190000049	7000000051	2331382343
8326344450	2200000049	7000000051	2205478243
8322545350	2210000049	7000000051	2086375543
8318754250	2220000049	7000000051	1973702843
8314972150	2230000049	7000000051	1867116743
8311198150	2240000049	7000000051	1766284843
8307433050	2250000049	7000000051	1670899943
8303675250	2260000049	7000000051	1580664243
8299927150	2270000049	7000000051	1495303743
8296185550	2280000049	7000000051	1414551243
8292453750	2290000049	7000000051	1338161143
8288729150	2300000049	7000000051	1265894943
8285013450	2310000049	7000000051	1197532743
8281305150	2320000049	7000000051	1132861143
8277606550	2330000049	7000000051	1071683043
8273912750	2340000049	7000000051	1013807843
8270228750	2350000049	7000000051	9590590242
8266551950	2360000049	7000000051	9072659642
8262884950	2370000049	7000000051	8582707642
8259223650	2380000049	7000000051	8119206742
8255569550	2390000049	7000000051	7680744042
8251924450	2400000049	7000000051	7265952942

ψ	θ	n	T
8248285750	2410000049	7000000051	6873561842
8244655250	2420000049	7000000051	6502368042
8241031150	2430000049	7000000051	6151213842
8237416850	2440000049	7000000051	5819029342
8233809050	2450000049	7000000051	5504777842
8230207650	2460000049	7000000051	5207502742
8226614350	2470000049	7000000051	4926276442
8223029250	2480000049	7000000051	4660242242
8219449750	2490000049	7000000051	4408570342
8215876750	2500000049	7000000051	4170493742
8212312750	2510000049	7000000051	3945270242
8208755950	2520000049	7000000051	3732213342
8205204050	2530000049	7000000051	3530658642
8201661050	2540000049	7000000051	3339992142
8198123650	2550000049	7000000051	3159618942
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3470851750	3438000050	2000000051	2465136640
3469994650	3439000050	2000000051	2449978440
3469137550	3440000050	2000000051	2434911340

ψ	θ	n	T
3468280950	3441000050	2000000051	2419938440
3467424250	3442000050	2000000051	2405055840
3466568050	3443000050	2000000051	2390267340
3465711950	3444000050	2000000051	2375567440
3464855950	3445000050	2000000051	2360959940
3463999450	3446000050	2000000051	2346439940
3463144850	3447000050	2000000051	2332011940
3462288750	3448000050	2000000051	2317669640
3461434750	3449000050	2000000051	2303418240
3460579850	3450000050	2000000051	2289252440
3459725350	3451000050	2000000051	2275175540
3458870850	3452000050	2000000051	2261183340
3458016750	3453000050	2000000051	2247279640
3457163050	3454000050	2000000051	2233458440
3456308950	3455000050	2000000051	2219724440
3455455650	3456000050	2000000051	2206073740
3454602150	3457000050	2000000051	2192508140
3453749250	3458000050	2000000051	2179024540
3452896550	3459000050	2000000051	2165625640
3452043550	3460000050	2000000051	2152306740
3451191050	3461000050	2000000051	2139072640
3450338750	3462000050	2000000051	2125917240
3449486750	3463000050	2000000051	2112844640
3448635150	3464000050	2000000051	2099850940
3447783350	3465000050	2000000051	2086938440
3446932150	3466000050	2000000051	2074104240
3446080450	3467000050	2000000051	2061350340
3445229650	3468000050	2000000051	2048673240
3444379150	3469000050	2000000051	2036075940
3443528350	3470000050	2000000051	2023554140
3442678250	3471000050	2000000051	2011110840
3441827850	3472000050	2000000051	1998742240
3440977850	3473000050	2000000051	1986452340
3440128550	3474000050	2000000051	1974235240
3439278650	3475000050	2000000051	1962096040
3438429550	3476000050	2000000051	1950029240
3437580350	3477000050	2000000051	1938038240
3436731550	3478000050	2000000051	1926119240
3435882950	3479000050	2000000051	1914275840
3435034350	3480000050	2000000051	1902502740
3434186250	3481000050	2000000051	1890804640
3433337950	3482000050	2000000051	1879175740
3432490250	3483000050	2000000051	1867620940
3431642950	3484000050	2000000051	1856134940
3430795250	3485000050	2000000051	1844721640
3429948150	3486000050	2000000051	1833376540
3429101050	3487000050	2000000051	1822102840
3428254450	3488000050	2000000051	1810896940
3427407850	3489000050	2000000051	1799761440
3426561450	3490000050	2000000051	1788693240
3425715450	3491000050	2000000051	1777694440
3424869350	3492000050	2000000051	1766761540
3424023750	3493000050	2000000051	1755897840
3423178350	3494000050	2000000051	1745099140
3422332950	3495000050	2000000051	1734368440
3421487950	3496000050	2000000051	1723701940
3420642950	3497000050	2000000051	1713102640
3419798350	3498000050	2000000051	1702567040
3418954050	3499000050	2000000051	1692097740
3418109450	3500000050	2000000051	1681691740
3417265650	3501000050	2000000051	1671350740
3416421750	3502000050	2000000051	1661072240
3415578150	3503000050	2000000051	1650858640
3414734750	3504000050	2000000051	1640709640
3413891450	3505000050	2000000051	1630617040
3413048550	3506000050	2000000051	1620588440
3412205650	3507000050	2000000051	1610623640
3411363250	3508000050	2000000051	1600718240
3410521050	3509000050	2000000051	1590875240
3409678750	3510000050	2000000051	1581091440
3408836950	3511000050	2000000051	1571369240
3407995050	3512000050	2000000051	1561705240
3407153650	3513000050	2000000051	1552102140
3406312550	3514000050	2000000051	1542556840
3405471050	3515000050	2000000051	1533071640
3404630550	3516000050	2000000051	1523643440
3403789550	3517000050	2000000051	1514274240
3402949250	3518000050	2000000051	1504961340
3402109050	3519000050	2000000051	1495707540
3401268850	3520000050	2000000051	1486508340

ψ	θ	n	T	ψ	θ	n	T
3400429050	3521000050	2000000051	1477368140	3367000050	3561000050	2000000051	1154332640
3399589150	3522000050	2000000051	1468282440	3366174450	3562000050	2000000051	1147232240
3398749950	3523000050	2000000051	1459253640	3365343650	3563000050	2000000051	1140179340
3397910850	3524000050	2000000051	1450280040	3364512750	3564000050	2000000051	1133167140
3397071650	3525000050	2000000051	1441361440	3363681850	3565000050	2000000051	1126198840
3396232950	3526000050	2000000051	1432497440	3362851550	3566000050	2000000051	1119272840
3395394250	3527000050	2000000051	1423688740	3362021050	3567000050	2000000051	1112390740
3394555950	3528000050	2000000051	1414932940	3361191050	3568000050	2000000051	1105549440
3393718050	3529000050	2000000051	1406232940	3360361350	3569000050	2000000051	1098750940
3392879650	3530000050	2000000051	1397584640	3359531550	3570000050	2000000051	1091993840
3392042150	3531000050	2000000051	1388990540	3358702250	3571000050	2000000051	1085279940
3391204350	3532000050	2000000051	1380448240	3357872750	3572000050	2000000051	1078604440
3390367150	3533000050	2000000051	1371959440	3357043550	3573000050	2000000051	1071971940
3389530250	3534000050	2000000051	1363522740	3356215050	3574000050	2000000051	1065379440
3388693050	3535000050	2000000051	1355138140	3355386150	3575000050	2000000051	1058827840
3387856550	3536000050	2000000051	1346804240	3354557950	3576000050	2000000051	1052317240
3387019750	3537000050	2000000051	1338522440	3353729450	3577000050	2000000051	1045845440
3386183650	3538000050	2000000051	1330290340	3352901550	3578000050	2000000051	1039414740
3385347650	3539000050	2000000051	1322110140	3352073750	3579000050	2000000051	1033021840
3384511550	3540000050	2000000051	1313979740	3351245950	3580000050	2000000051	1026670040
3383675950	3541000050	2000000051	1305899640	3350418650	3581000050	2000000051	1020356240
3382840350	3542000050	2000000051	1297868640	3349591250	3582000050	2000000051	1014081540
3382005350	3543000050	2000000051	1289887640	3348764550	3583000050	2000000051	1007844840
3381170350	3544000050	2000000051	1281955540	3347937850	3584000050	2000000051	1001648040
3380335150	3545000050	2000000051	1274072240	3347111050	3585000050	2000000051	9954874439
3379500650	3546000050	2000000051	1266237040	3346284750	3586000050	2000000051	9893663639
3378666150	3547000050	2000000051	1258450540	3345458450	3587000050	2000000051	9832814439
3377832050	3548000050	2000000051	1250711240	3344632650	3588000050	2000000051	9772354039
3376998250	3549000050	2000000051	1243020640	3343806950	3589000050	2000000051	9712257639
3376164150	3550000050	2000000051	1235375940	3342981150	3590000050	2000000051	9652529239
3375330650	3551000050	2000000051	1227779540	3342155850	3591000050	2000000051	9593174039
3374496950	3552000050	2000000051	1220229040	3341330650	3592000050	2000000051	9534186439
3373664050	3553000050	2000000051	1212725340	3340505750	3593000050	2000000051	9475548439
3372831250	3554000050	2000000051	1205266940	3339680850	3594000050	2000000051	9417286039
3371998150	3555000050	2000000051	1197856640	3338856450	3595000050	2000000051	9359369239
3371165850	3556000050	2000000051	1190489640	3338032150	3596000050	2000000051	9301819239
3370333150	3557000050	2000000051	1183168840	3337207850	3597000050	2000000051	9244613639
3369501250	3558000050	2000000051	1175892640	3336384150	3598000050	2000000051	9187768439
3368669450	3559000050	2000000051	1168662140	3335560150	3599000050	2000000051	9131255639
3367837450	3560000050	2000000051	1161475040	3334736350	3600000050	2000000051	9075108439

APPENDIX F

Sample Data Sheet For Self-Diffusion Measurements And Sample Calculations

SAMPLE DATA PAGE

Self-Diffusion

System Zn-Hg | Run No. 91 | Date 7/1/62

	Reservoir Data	Diffusion Run
CTB #B	Amalgam #13	Start 7/1-2206:25
Temp 93.2°C	CPM-BG 33.0	End 7/2-1109:00
Ref Cnts 128,000		Elapsed 46,955 sec
Capillary Data	BG Count (1)	BG Count (2)
RPM 1/2	Start 6/30-2312	Start 7/2-2236
Dia 2 mm	Counts 8000	Counts 8000
Counts 128,000	Time 573.50	Time 580.81
Amalgam AZn-10	CPM 13.9	CPM 13.9

Capillaries				
Number	211	212	213	214
Length	3.513	3.538	3.500	3.508
Initial Counting				
Date-Hour	7/1-1928	7/1-1952	7/1-2009	7/1-2038
Ref Cnt Time (1)	4.34	4.33	4.36	4.37
CPM-BG	29,479	29,547	29,344	29,277
Counting Time	9.70	9.97	9.93	9.83
C_o', CPM-BG	13,182.0	12,824.6	12,876.3	13,007.5
Ref Cnt Time (2)	4.33	4.36	4.37	4.37
CPM-BG	29,547	29,344	29,277	29,277
Ave Ref Cnt Rate	29,513	29,446	29,310	29,277
Final Counting				
Date-Hour	7/3-0852	7/3-0940	7/3-1055	7/3-1124
Ref Cnt Time (1)	4.41	4.38	4.36	4.34
CPM-BG	29,011	29,210	29,344	29,479
Counting Time	15.56	15.76	15.55	15.34
C_f', CPM-BG	8,212.4	8,108.0	8,217.7	8,330.4
Ref Cnt Time (2)	4.38	4.36	4.34	4.36
CPM-BG	29,210	29,344	29,479	29,344
Ave Ref Cnt Rate	29,110	29,277	29,412	29,412
Correction Factor, F	1.01384	1.00577	0.99653	0.99541

SAMPLE CALCULATIONS

For Run No. 91, Capillary No. 1

Correction for Instrument Variation and Radioisotope Decay

The correction factor, F, for instrument variation and radioisotope decay is determined from the reference counting data. These reference counts were made before and after each capillary count as is shown on the preceding Sample Data Page. The individual reference count rates, corrected for background, are used to determine an average reference count rate corresponding to each instance of counting the capillary.

The average reference count rate at the initial counting of the capillary was

$$\tfrac{1}{2}\left[\frac{128,000}{4.34} - 13.9 + \frac{128,000}{4.33} - 13.9\right]$$

$$= 29,513 \text{ counts per minute.}$$

At the final counting of the capillary, the average reference count rate was 29,110 counts per minute.

The correction factor, F, is $\frac{29,513}{29,110}$, or 1.01384.

Calculation of the Diffusion Coefficient

$$\psi = \frac{C_f - C_r}{C_o - C_r} = \frac{FC_f' - C_r}{GC_o' - C_r} = \frac{(1.01384)\ (8,212.4) - 33.0}{(0.98774)\ (13,182.0) - 33.0}$$

$$= 0.63854.$$

The factor G in the above equation is the correction for volume expansion of the amalgam between the capillary

filling temperature of 25°C and the diffusion temperature.

From Appendix E, the value of θ corresponding to the ψ calculated above is found to be 0.1026. The diffusion coefficient is calculated from the defining equation for θ.

$$D = \frac{L^2\theta}{t} = \frac{(3.513)^2(0.1026)}{46,955}$$

$$= 2.697 \times 10^{-5} \text{ cm}^2/\text{sec.}$$

APPENDIX G

Effect Of Capillary Turning Rate On The Diffusion Coefficient For Various Capillary Diameters

TABLE G-I

EFFECT OF CAPILLARY TURNING RATE,

DIFFUSION DATA

For 0.569 ± 0.002 Weight Percent Zinc Amalgam at 50.0°C

Run-Capillary Number	Capillary Diameter mm	RPM	C_o' cpm	C_f' cpm	C_r cpm	F
32-1	2.0	0.0	1119.3	745.7	17.4	1.02380
-2	1.5	0.0	639.2	427.3	11.1	1.02840
-3	1.0	0.0	284.8	184.5	5.1	1.03455
-4	1.5	0.0	652.2	436.0	11.1	1.01107
33-1	2.0	0.0	1112.3	866.2	17.8	1.00746
-2	1.5	0.0	661.0	504.8	11.4	1.01254
-3	1.0	0.0	295.2	222.6	5.3	1.02844
-4	1.5	0.0	624.2	479.3	11.4	1.00630
34-1	2.0	0.42	1088.6	734.3	18.3	1.00943
-2	1.5	0.42	676.7	458.9	11.7	1.00468
-3	1.0	0.42	289.2	200.3	5.5	1.00315
-4	1.5	0.42	663.9	457.0	11.7	0.99852
35-1	2.0	0.42	1109.4	823.5	18.8	1.00612
-2	1.5	0.42	655.4	499.7	12.1	0.99390
-3	1.0	0.42	284.0	218.4	5.7	0.99236
-4	1.5	0.42	638.7	483.4	12.1	1.00785
36-1	2.0	0.76	1086.7	785.7	19.2	1.00628
-2	1.5	0.76	637.8	459.4	12.4	1.00151
-3	1.0	0.76	287.0	205.8	5.9	0.99987
-4	1.5	0.76	623.4	448.3	12.4	1.01090
37-1	2.0	0.76	1096.8	799.9	19.6	1.00778
-2	1.5	0.76	626.3	466.1	12.8	1.00316
-3	1.0	0.76	278.5	208.6	6.1	1.00007
-4	1.5	0.76	630.2	468.1	12.8	0.99085
38-1	2.0	1.27	1118.1	705.3	20.1	0.99850
-2	1.5	1.27	626.0	377.2	13.2	0.99697
-3	1.0	1.27	286.3	176.2	6.3	0.99544
-4	1.5	1.27	649.0	403.2	13.2	1.00159

TABLE G-I--Continued

Run-Capillary Number	Capillary Diameter mm	RPM	C_o' cpm	C_f' cpm	C_r cpm	F
39-1	2.0	1.27	1079.2	566.5	20.7	1.00931
-2	1.5	1.27	652.3	345.7	13.6	1.01702
-3	1.0	1.27	285.2	157.8	6.5	1.01247
-4	1.5	1.27	672.7	364.0	13.6	1.02342

TABLE G-II

EFFECT OF CAPILLARY TURNING RATE,

DIFFUSION COEFFICIENTS

For 0.569 Weight Percent Zinc Amalgam At 50.0°C

Run-Capillary Number	Capillary Diameter mm	RPM	$\frac{C_f - C_r}{C_o - C_r}$	L cm	Time sec	$D \times 10^5$ $\frac{cm^2}{sec}$
32-1	2.0	0.0	0.68017	3.490	50,400	1.941
-2	1.5	0.0	0.68510	3.602	50,400	2.005
-3	1.0	0.0	0.66726	3.574	50,400	2.205
-4	1.5	0.0	0.67339	3.569	50,400	2.118
33-1	2.0	0.0	0.78251	3.541	27,000	1.725
-2	1.5	0.0	0.77284	3.579	27,000	1.921
-3	1.0	0.0	0.77497	3.637	27,000	1.950
-4	1.5	0.0	0.77202	3.465	27,000	1.814
34-1	2.0	0.42	0.67856	3.556	52,200	1.966
-2	1.5	0.42	0.67883	3.553	52,200	1.956
-3	1.0	0.42	0.69205	3.592	52,200	1.841
-4	1.5	0.42	0.68488	3.516	52,200	1.847
35-1	2.0	0.42	0.74590	3.531	27,000	2.341
-2	1.5	0.42	0.75671	3.571	27,000	2.196
-3	1.0	0.42	0.76180	3.602	27,000	2.141
-4	1.5	0.42	0.76171	3.594	27,000	2.134

TABLE G-II--Continued

Run-Capillary Number	Capillary Diameter mm	RPM	$\frac{C_f - C_r}{C_o - C_r}$	L cm	Time sec	D x 10^5 $\frac{cm^2}{sec}$
36-1	2.0	0.76	0.72599	3.515	39,600	1.841
-2	1.5	0.76	0.71916	3.584	39,600	2.009
-3	1.0	0.76	0.71433	3.551	39,600	2.041
-4	1.5	0.76	0.72476	3.581	39,600	1.927
37-1	2.0	0.76	0.73353	3.538	34,200	2.040
-2	1.5	0.76	0.74471	3.561	34,200	1.898
-3	1.0	0.76	0.74689	3.556	34,200	1.860
-4	1.5	0.76	0.73389	3.564	34,200	2.065
38-1	2.0	1.27	0.62596	3.567	68,400	2.055
-2	1.5	1.27	0.59487	3.350	68,400	2.115
-3	1.0	1.27	0.60672	3.548	68,400	2.236
-4	1.5	1.27	0.61725	3.553	68,400	2.124
39-1	2.0	1.27	0.52302	3.515	102,600	2.154
-2	1.5	1.27	0.53162	3.546	102,600	2.113
-3	1.0	1.27	0.55249	3.541	102,600	1.923
-4	1.5	1.27	0.54709	3.548	102,600	1.978

TABLE G-III

DIFFUSION COEFFICIENT AS A FUNCTION OF CAPILLARY DIAMETER AND TURNING RATE

For 0.596 Weight Percent Zinc Amalgam At 50.0°C

Capillary Diameter mm	Capillary Turning Rate, RPM			
	0.0	0.42	0.76	1.27
2.0	1.833*	2.154	1.940	2.104
1.5	1.964	2.033	1.975	2.082
1.0	2.078	1.991	1.950	2.080
For All Capillaries	1.960	2.053	1.960	2.087

*Average value of the diffusion coefficient ($cm^2/sec \times 10^5$), from Table G-II.

APPENDIX H

Self-Diffusion Coefficients and Data For The Zinc-Mercury System

TABLE H-I

DIFFUSION DATA VERSUS TEMPERATURE

FOR VARIOUS ZINC AMALGAMS

Run-Capillary Number	Temp. °C	Radioactivity of Zinc			Corr. Factor, F
		C_o' cpm	C_f' cpm	C_r cpm	
0.0212 ± 0.0001 Weight Percent Zinc (0.0651 Atomic Percent)					
64-1	50.0	1654.0	991.9	2.0	1.00742
-2	50.0	1668.3	973.9	2.0	1.00375
-3	50.0	1661.7	960.4	2.0	0.99514
-4	50.0	1659.5	914.2	2.0	0.99270
67-1	50.0	1614.1	781.2	5.0	1.01233
-2	50.0	1667.6	827.2	5.0	1.00378
-3	50.0	1684.3	850.8	5.0	0.99755
-4	50.0	1661.0	880.2	5.0	1.00486
0.0219 ± 0.0001 Weight Percent Zinc (0.0671 Atomic Percent)					
41-1	50.0	1901.9	1122.1	0.8	0.59093
-2	50.0	1889.1	1123.1	0.8	0.59142
-3	50.0	1910.6	1163.7	0.8	0.60677
-4	50.0	1919.3	1191.7	0.8	0.61607
42-2	50.0	1910.7	791.7	3.6	0.41226
-3	50.0	1938.7	783.1	3.6	0.40186
43-1	50.0	1932.5	1150.5	6.3	0.59586
-2	50.0	1898.9	1102.5	6.3	0.58182
-3	50.0	1950.5	1161.8	6.3	0.59947
44-1	50.0	1910.4	844.0	8.2	0.53039
-3	50.0	1894.5	814.2	8.2	0.51760
-4	50.0	1894.5	816.1	8.2	0.51821

TABLE H-I--Continued

Run-Capillary Number	Temp. °C	Radioactivity of Zinc C_o' cpm	C_f' cpm	C_r cpm	Corr. Factor, F
45-2	50.0	1546.5	981.1	10.0	0.62984
-3	50.0	1560.9	933.9	10.0	0.59638
50-1	50.0	371.3	214.4	4.4	1.01619
-2	50.0	380.3	216.1	4.4	1.01724
-3	50.0	368.8	204.8	4.4	1.01717
-4	50.0	364.5	211.6	4.4	1.01607
51-1	50.0	374.5	204.6	4.4	0.83700
-2	50.0	363.8	196.3	4.4	0.83828
-3	50.0	367.2	198.7	4.4	0.82908
-4	50.0	372.1	202.1	4.4	0.83258
0.0226 ± 0.0001 Weight Percent Zinc (0.0694 Atomic Percent)					
68-1	50.0	2715.0	1597.4	1.9	0.99171
-2	50.0	2715.0	1582.3	1.9	0.98104
-3	50.0	2685.4	1674.0	1.9	0.96943
69-1	50.0	2668.3	1705.7	5.4	1.00368
-2	50.0	2615.9	1698.8	5.4	1.00242
-3	50.0	2657.1	1739.9	5.4	1.00486
-4	50.0	2621.4	1662.9	5.4	1.00119
70-1	70.2	2665.9	1530.6	8.6	1.01327
-2	70.2	2558.2	1361.0	8.6	1.01451
-3	70.2	2579.0	1419.7	8.6	1.01206
-4	70.2	2619.0	1475.6	8.6	1.00118
73-1	93.2	2578.8	1456.7	11.8	1.03241
-2	93.2	2632.5	1476.4	11.8	1.02423
-3	93.2	2629.7	1468.6	11.8	1.02790
-4	93.2	2592.0	1429.3	11.8	1.02527
75-2	119.1	2567.9	1602.6	14.4	1.01800
-3	119.1	2581.0	1583.4	14.4	1.00723

TABLE H-I--Continued

Run-Capillary Number	Temp. °C	Radioactivity of Zinc			Corr. Factor, F
		C_o' cpm	C_f' cpm	C_r cpm	
0.1175 ± 0.0004 Weight Percent Zinc (0.3597 Atomic Percent)					
98-1	50.0	2811.6	2042.6	40.0	1.00221
-2	50.0	2816.6	2028.1	40.0	1.00560
-3	50.0	2736.2	1952.8	40.0	1.00337
-4	50.0	2809.1	2061.2	40.0	0.99771
95-1	70.2	2881.7	2131.7	32.1	1.00677
-2	70.2	2866.1	2025.8	32.1	1.01825
-3	70.2	2908.2	2075.1	32.1	1.02363
-4	70.2	2929.7	2112.5	32.1	1.00224
96-1	93.2	2902.8	1786.5	35.4	1.00344
-2	93.2	2935.1	1743.9	35.4	1.00344
-3	93.2	2965.3	1787.5	35.4	1.00458
-4	93.2	2881.7	1725.7	35.4	1.01332
85-1	119.1	6000.4	3440.0	24.8	1.02306
0.577 ± 0.002 Weight Percent Zinc (1.750 Atomic Percent)					
87-1	50.0	12,621.9	9227.6	6.3	1.00928
-2	50.0	12,941.0	9404.4	6.3	1.01037
-3	50.0	12,760.7	9161.3	6.3	1.00924
-4	50.0	12,967.9	9530.8	6.3	1.01608
89-1	70.2	12,902.3	9294.8	18.8	0.98769
-2	70.2	12,648.2	8825.5	18.8	0.95197
-3	70.2	12,915.4	9050.9	18.8	0.98223
-4	70.2	12,697.1	8789.0	18.8	0.98421
91-1	93.2	13,182.0	8212.4	33.0	1.01384
-2	93.2	12,824.6	8108.0	33.0	1.00577
-3	93.2	12,876.3	8217.7	33.0	0.99653
-4	93.2	13,007.5	8330.4	33.0	0.99541
92-1	119.1	12,967.4	9175.0	45.8	1.00577
93-1	119.1	12,571.7	6652.5	60.5	1.00109
-2	119.1	12,915.0	6968.9	60.5	1.00457
-4	119.1	12,760.2	7152.7	60.5	1.00226

TABLE H-I--Continued

Run-Capillary Number	Temp. °C	Radioactivity of Zinc C_o' cpm	Radioactivity of Zinc C_f' cpm	Radioactivity of Zinc C_r cpm	Corr. Factor, F
1.159 ± 0.001 Weight Percent Zinc (3.473 Atomic Percent)					
71-1	50.0	12,167.3	8138.0	7.9	1.01711
-2	50.0	11,862.8	8025.3	7.9	1.01220
-3	50.0	12,260.5	8280.6	7.9	1.01578
-4	50.0	11,999.0	7905.9	7.9	1.01948
72-1	50.0	11,527.0	8794.8	20.0	1.01441
-2	50.0	11,970.1	9082.1	20.0	1.02029
-4	50.0	12,049.2	8960.9	20.0	1.02416
74-1*	50.0	3,099.5	2261.0	6.4	1.01073
-2	50.0	3,047.3	2209.6	6.4	1.00957
-3	50.0	3,118.5	2211.5	6.4	1.00838
-4	50.0	3,122.4	2246.9	6.4	1.00595
76-1	70.2	11,913.9	7975.0	33.5	1.01431
-2	70.2	11,869.6	7980.0	33.5	1.00712
-3	70.2	12,026.1	7965.0	33.5	1.00833
-4	70.2	11,795.7	7714.5	33.5	1.01312
78-1	93.2	11,750.1	8663.0	43.7	0.99645
-2	93.2	11,970.4	8645.4	43.7	1.00235
-3	93.2	11,870.3	8501.3	43.7	1.00355
-4	93.2	11,600.6	8148.3	43.7	1.00590
80-2	119.1	11,643.0	7926.3	52.2	0.99295
1.900 ± 0.002 Weight Percent Zinc (5.609 Atomic Percent)					
84-1	50.0	11,654.1	8788.7	5.1	0.99886
-2	50.0	11,383.9	8698.8	5.1	1.00238
-3	50.0	11,229.5	8378.8	5.1	1.01059
-4	50.0	11,353.6	8286.3	5.1	0.99886
86-1	70.2	11,684.9	8936.4	14.2	1.00234
-2	70.2	11,684.9	8849.7	14.2	1.00468
-3	70.2	11,433.7	8809.1	14.2	1.00354
-4	70.2	11,495.5	8675.2	14.2	0.99734

*1 mm diameter capillaries were used in run No. 74.

TABLE H-I--Continued

Run-Capillary Number	Temp. °C	Radioactivity of Zinc C_o' cpm	C_r' cpm	C_r cpm	Corr. Factor, F
88-1	93.2	11,464.5	8026.4	15.3	0.99771
-2	93.2	11,413.3	8036.5	15.3	0.99761
-3	93.2	11,474.8	7897.2	15.3	0.99182
-4	93.2	11,342.8	7991.2	15.3	0.99533
90-1	119.1	11,548.2	7725.0	36.6	1.01399
-2	119.1	11,569.1	8670.1	36.6	1.00860
-4	119.1	11,383.4	8374.1	36.6	1.00117

TABLE H-II

SELF-DIFFUSION COEFFICIENTS OF ZINC VERSUS TEMPERATURE FOR VARIOUS ZINC AMALGAMS

Run-Capillary Number	Temp. °C	$\frac{C_f - C_r}{C_o - C_r}$	Length cm	Time sec	$Dx10^5$ $\frac{cm^2}{sec}$
0.0212 $\pm$ 0.0001 Weight Percent Zinc (0.0651 Atomic Percent)					
64-1	50.0	0.60641	3.546	75,565	2.025
-2	50.0	0.58812	3.576	75,565	2.255
-3	50.0	0.57725	3.505	75,565	2.283
-4	50.0	0.54880	3.358	75,565	2.388
67-1	50.0	0.49059	3.475	102,940	2.397
-2	50.0	0.49866	3.579	102,940	2.461
-3	50.0	0.50470	3.575	102,940	2.396
-4	50.0	0.53350	3.515	102,940	2.053

TABLE H-II--Continued

Run-Capillary Number	Temp. °C	$\frac{C_f - C_r}{C_o - C_r}$	Length cm	Time sec	$Dx10^5 \frac{cm^2}{sec}$
0.0219 ± 0.0001 Weight Percent Zinc (0.0671 Atomic Percent)					
41-1	50.0	0.59093	3.505	80,100	2.015
-2	50.0	0.59142	3.520	80,100	2.028
-3	50.0	0.60677	3.551	80,100	1.912
-4	50.0	0.61607	3.574	80,100	1.845
42-2	50.0	0.41226	3.561	175,550	1.981
-3	50.0	0.40186	3.518	175,550	2.006
43-1	50.0	0.59586	3.531	74,505	2.147
-2	50.0	0.58182	3.467	74,505	2.217
-3	50.0	0.59947	3.561	74,505	2.145
44-1	50.0	0.53039	3.520	97,745	2.198
-3	50.0	0.51760	3.508	97,745	2.304
-4	50.0	0.51821	3.546	97,745	2.348
45-2	50.0	0.62984	3.594	64,800	2.145
-3	50.0	0.59638	3.353	64,800	2.221
50-1	50.0	0.58449	3.500	88,920	1.868
-2	50.0	0.57572	3.561	88,920	2.016
-3	50.0	0.56216	3.538	88,920	2.120
-4	50.0	0.58752	3.579	88,920	1.925
51-1	50.0	0.45298	3.599	148,050	2.303
-2	50.0	0.44766	3.533	148,050	2.032
-3	50.0	0.44397	3.551	148,050	2.081
-4	50.0	0.44769	3.553	148,050	2.055
0.0226 ± 0.0001 Weight Percent Zinc (0.0694 Atomic Percent)					
68-1	50.0	0.58584	3.515	74,865	2.223
-2	50.0	0.57404	3.523	74,865	2.362
-3	50.0	0.60677	3.543	74,865	2.036
69-1	50.0	0.64378	3.548	62,145	2.020
-2	50.0	0.65322	3.543	62,145	1.908
-3	50.0	0.66028	3.543	62,145	1.830
-4	50.0	0.63724	3.520	62,145	2.061

TABLE H-II--Continued

Run-Capillary Number	Temp. °C	$\frac{C_f - C_r}{C_o - C_r}$	Length cm	Time sec	$Dx10^5$ $\frac{cm^2}{sec}$
70-1	70.2	0.58519	3.614	64,830	2.724
-2	70.2	0.54261	3.322	64,830	2.798
-3	70.2	0.56022	3.416	64,830	2.734
-4	70.2	0.56728	3.510	64,830	2.795
73-1	93.2	0.58851	3.548	59,995	2.791
-2	93.2	0.57964	3.515	59,995	2.858
-3	93.2	0.57926	3.500	59,995	2.840
-4	93.2	0.57039	3.467	59,995	2.905
75-2	119.1	0.64418	3.581	36,080	3.533
-3	119.1	0.62639	3.543	36,080	3.818
0.1175 ± 0.0004 Weight Percent Zinc (0.3597 Atomic Percent)					
98-1	50.0	0.72750	3.490	33,590	2.114
-2	50.0	0.72342	3.586	33,590	2.301
-3	50.0	0.71516	3.411	33,590	2.206
-4	50.0	0.73156	3.500	33,590	2.064
95-1	70.2	0.74803	3.513	29,030	2.307
-2	70.2	0.72249	3.419	29,030	2.436
-3	70.2	0.74394	3.536	29,030	2.218
-4	70.2	0.72558	3.492	29,030	2.482
96-1	93.2	0.62053	3.350	49,845	2.546
-2	93.2	0.59869	3.510	49,845	3.127
-3	93.2	0.60834	3.570	49,845	3.081
-4	93.2	0.60949	3.513	49,845	2.966
85-1	119.1	0.59487	3.500	39,660	3.981
0.577 ± 0.002 Weight Percent Zinc (1.750 Atomic Percent)					
87-1	50.0	0.74108	3.505	36,695	1.768
-2	50.0	0.73745	3.531	36,695	1.840
-3	50.0	0.72771	3.518	36,695	1.963
-4	50.0	0.75004	3.566	36,695	1.695
92-1	119.1	0.72283	3.533	27,180	2.769

TABLE H-II--Continued

Run-Capillary Number	Temp. °C	$\frac{C_f - C_r}{C_o - C_r}$	Length cm	Time sec	$Dx10^5$ $\frac{cm^2}{sec}$
89-1	70.2	0.71696	3.546	36,175	2.186
-2	70.2	0.66921	3.353	36,175	2.671
-3	70.2	0.69353	3.459	36,175	2.441
-4	70.2	0.68640	3.429	36,175	2.509
91-1	93.2	0.63854	3.513	46,955	2.697
-2	93.2	0.64283	3.538	46,955	2.671
-3	93.2	0.64295	3.500	46,955	2.614
-4	93.2	0.64449	3.508	46,955	2.602
93-1	119.1	0.53655	3.399	59,010	3.305
-2	119.1	0.54921	3.518	59,010	3.349
-4	119.1	0.56937	3.558	59,010	3.126
1.159 ± 0.001 Weight Percent Zinc (3.473 Atomic Percent)					
71-1	50.0	0.68316	3.551	58,920	1.687
-2	50.0	0.68766	3.551	58,920	1.639
-3	50.0	0.68895	3.538	58,920	1.615
-4	50.0	0.67454	3.434	58,920	1.665
72-1	50.0	0.77709	3.498	31,665	1.507
-2	50.0	0.77726	3.602	31,665	1.598
-4	50.0	0.76472	3.515	31,665	1.697
74-1*	50.0	0.74010	3.574	43,590	1.544
-2	50.0	0.73480	3.546	43,590	1.592
-3	50.0	0.71776	3.571	43,590	1.831
-4	50.0	0.72661	3.576	43,590	1.722
76-1	70.2	0.68303	3.531	47,165	2.086
-2	70.2	0.68175	3.495	47,165	2.060
-3	70.2	0.67239	3.467	47,165	2.148
-4	70.2	0.66708	3.365	47,165	2.090
78-1	93.2	0.74280	3.597	29,205	2.304
-2	93.2	0.73192	3.531	29,205	2.408
-3	93.2	0.72662	3.538	29,205	2.516
-4	93.2	0.71414	3.421	29,205	2.573

*1 mm diameter capillaries were used in run No. 74.

TABLE H-II--Continued

Run-Capillary Number	Temp. °C	$\frac{C_f - C_r}{C_o - C_r}$	Length cm	Time sec	$D \times 10^5 \frac{cm^2}{sec}$
80-2	119.1	0.68613	3.538	34,855	2.780
1.900 ± 0.002 Weight Percent Zinc (5.609 Atomic Percent)					
84-1	50.0	0.75658	3.513	34,260	1.675
-2	50.0	0.76932	3.543	34,260	1.532
-3	50.0	0.75735	3.470	34,260	1.624
-4	50.0	0.73219	3.264	34,260	1.751
86-1	70.2	0.77259	3.576	28,245	1.838
-2	70.2	0.76687	3.579	28,245	1.936
-3	70.2	0.77925	3.558	28,245	1.712
-4	70.2	0.75856	3.543	28,245	2.035
88-1	93.2	0.70677	3.518	43,925	1.902
-2	93.2	0.71077	3.508	43,925	1.841
-3	93.2	0.69064	3.495	43,925	2.091
-4	93.2	0.70952	3.526	43,925	1.877
90-1	119.1	0.68892	3.538	32,585	2.920
-2	119.1	0.76807	3.528	32,585	1.612
-4	119.1	0.74830	3.536	32,585	1.911

APPENDIX I

Self-Diffusion Coefficients and Data For The Cadmium-Mercury System

TABLE I-I

DIFFUSION DATA VERSUS TEMPERATURE
FOR VARIOUS CADMIUM AMALGAMS

Run-Capillary Number	Temp. °C	Radioactivity of Cadmium C_o' cpm	Radioactivity of Cadmium C_f' cpm	Radioactivity of Cadmium C_r cpm	Corr. Factor, F*
0.0498±0.002 Weight Percent Cadmium (0.08892 Atomic Percent)					
1-1	50.0	371.8	223.9	0.0	0.99966
-2	50.0	347.6	216.2	0.0	0.99966
-3	50.0	366.3	218.2	0.0	0.99966
-4	50.0	360.3	214.5	0.0	0.99966
2-1	70.2	380.6	269.0	0.0	1.01578
-2	70.2	393.4	270.6	0.0	1.01578
-3	70.2	389.3	264.6	0.0	1.01578
-4	70.2	387.2	282.9	0.0	1.01578
3-1	93.2	385.1	254.6	0.0	1.01190
-2	93.2	390.9	262.7	0.0	1.01190
-3	93.2	391.5	263.0	0.0	1.01190
-4	93.2	389.3	248.6	0.0	1.01190
4A-1	119.1	341.1	218.7	0.0	1.03410
-2	119.1	344.6	210.2	0.0	1.03410
-3	119.1	338.1	218.4	0.0	1.03410
-4	119.1	347.9	210.4	0.0	1.03410
0.266±0.001 Weight Percent Cadmium (0.4741 Atomic Percent)					
5-1	50.0	1531.0	1136.7	0.9	1.02149
-2	50.0	1597.8	1230.0	0.9	1.02149
-3	50.0	1562.2	1158.3	0.9	1.02149
-4	50.0	1575.6	1212.1	0.9	1.02149

*Correction factor calculated using over-all average of reference count rates for each set (initial and final) of capillary count-rate measurements. This gives one average correction factor for each diffusion run.

TABLE I-I--Continued

Run-Capillary Number	Temp. °C	Radioactivity of Cadmium C_o' cpm	C_f' cpm	C_r cpm	Corr. Factor, F
6-1	70.2	1611.8	1062.4	3.1	1.09106
-2	70.2	1640.6	1066.9	3.1	1.09106
-3	70.2	1626.1	1069.8	3.1	1.09106
-4	70.2	1610.7	1008.7	3.1	1.09106
7A-1	93.2	1566.5	1027.3	7.8	1.00883
-2	93.2	1589.5	1023.1	7.8	1.00883
-4	93.2	1522.3	964.4	7.8	1.00883
8-1	119.1	1608.3	1016.3	10.0	1.01931
-2	119.1	1578.4	1019.0	10.0	1.01931
-4	119.1	1592.1	982.7	10.0	1.01931
1.307 ± 0.005 Weight Percent Cadmium (2.309 Atomic Percent)					
10-1	50.0	8156.2	6054.6	8.7	1.02465
-2	50.0	8093.4	5951.3	8.7	1.02465
-3	50.0	7910.6	5728.5	8.7	1.02465
-4	50.0	7861.3	5873.3	8.7	1.02465
11-1	50.0	7716.9	5625.4	15.2	1.02454
-2	50.0	7861.3	5590.3	15.2	1.02454
-3	50.0	7920.5	5697.0	15.2	1.02454
-4	50.0	7890.8	5610.3	15.2	1.02454
12-1	70.2	7804.3	5691.8	21.7	1.00198
-2	70.2	7804.3	5449.3	21.7	1.00198
-3	70.2	7931.9	5565.5	21.7	1.00198
-4	70.2	7992.2	5645.7	21.7	1.00198
13-1	93.2	7746.8	5098.2	28.3	1.04834
-2	93.2	7746.8	5024.5	28.3	1.04834
-3	93.2	7775.4	5052.9	28.3	1.04834
-4	93.2	7775.4	5000.4	28.3	1.04834
14-1	119.1	7971.7	5217.3	35.0	1.03747
-2	119.1	7737.0	5024.5	35.0	1.03747
-3	119.1	7921.6	5535.5	35.0	1.03747
-4	119.1	7765.6	5044.8	35.0	1.03747

TABLE I-I--Continued

Run-Capillary Number	Temp. °C	Radioactivity of Cadmium C_o' cpm	Radioactivity of Cadmium C_f' cpm	Radioactivity of Cadmium C_r cpm	Corr. Factor, F
2.646 ± 0.010 Weight Percent Cadmium (4.626 Atomic Percent)					
15-1	50.0	7569.7	5430.3	3.5	1.04019
-2	50.0	7418.2	5265.9	3.5	1.04019
-3	50.0	7569.7	5274.7	3.5	1.04019
-4	50.0	7374.8	5174.4	3.5	1.04019
16-1	70.2	7076.0	5119.4	9.3	0.98113
-2	70.2	7383.0	5288.1	9.3	0.98113
-3	70.2	7265.9	5226.3	9.3	0.98113
-4	70.2	7230.4	5200.2	9.3	0.98113
17-1	93.2	7314.4	4897.9	15.4	1.05039
-2	93.2	7297.4	4960.1	15.4	1.05039
-3	93.2	7314.4	4863.6	15.4	1.05039
-4	93.2	7400.3	4894.1	15.4	1.05039
18-1	119.1	7230.7	4706.0	21.5	1.02786
-2	119.1	7340.2	4695.3	21.5	1.02786
-3	119.1	7280.8	4642.9	21.5	1.02786
-4	119.1	7124.3	4381.9	21.5	1.02786
4.275 ± 0.004 Weight Percent Cadmium (7.382 Atomic Percent)					
20-1	70.0	8648.9	6172.2	8.1	1.03360
-2	70.0	8590.0	6112.1	8.1	1.03360
-3	70.0	8744.7	6359.8	8.1	1.03360
-4	70.0	8555.1	6196.6	8.1	1.03360
21-1	93.2	8590.2	6247.6	12.3	1.03364
-2	93.2	8578.6	6180.0	12.3	1.03364
-3	93.2	8429.7	6078.3	12.3	1.03364
-4	93.2	8543.8	6060.7	12.3	1.03364
22-1	119.1	8648.0	6143.8	28.5	1.01564
-2	119.1	8496.6	6008.6	28.5	1.01564
-3	119.1	8439.8	5946.0	28.5	1.01564
-4	119.1	8496.6	6014.3	28.5	1.01564

TABLE I-II

SELF-DIFFUSION COEFFICIENTS OF CADMIUM VERSUS TEMPERATURE FOR VARIOUS CADMIUM AMALGAMS

Run-Capillary Number	Temp. °C	$\frac{C_f - C_r}{C_o - C_r}$	Length cm	Time sec	$D \times 10^5 \frac{cm^2}{sec}$
0.0498±0.0002 Weight Percent Cadmium (0.08892 Atomic Percent)					
1-1	50.0	0.60472	3.548	56,970	2.711
-2	50.0	0.62458	3.500	56,970	2.380
-3	50.0	0.59818	3.612	56,970	2.904
-4	50.0	0.59783	3.528	56,970	2.775
2-1	70.2	0.72382	3.515	27,055	2.735
-2	70.2	0.70444	3.518	27,055	3.138
-3	70.2	0.69607	3.261	27,055	2.852
-4	70.2	0.74825	3.553	27,055	2.324
3-1	93.2	0.67729	3.495	42,615	2.345
-2	93.2	0.68847	3.575	42,615	2.285
-3	93.2	0.68820	3.531	42,615	2.234
-4	93.2	0.65419	3.467	42,615	2.649
4A-1	119.1	0.67439	3.520	50,195	2.056
-2	119.1	0.64159	3.480	50,195	2.434
-3	119.1	0.67944	3.505	50,195	1.975
-4	119.1	0.63611	3.510	50,195	2.553
0.266±0.001 Weight Percent Cadmium (0.4741 Atomic Percent)					
5-1	50.0	0.76171	3.309	24,350	2.006
-2	50.0	0.78980	3.490	24,350	1.736
-3	50.0	0.76068	3.411	24,350	2.150
-4	50.0	0.78927	3.510	24,350	1.766
6-1	70.2	0.72453	3.513	42,065	1.749
-2	70.2	0.71481	3.607	42,065	1.976
-3	70.2	0.72316	3.518	42,065	1.771
-4	70.2	0.68828	3.508	42,065	2.232
7A-1	93.2	0.66812	3.465	32,075	3.238
-2	93.2	0.65569	3.553	32,075	3.664
-4	93.2	0.64520	3.261	32,075	3.279

TABLE I-II--Continued

Run-Capillary Number	Temp. °C	$\frac{C_f - C_r}{C_o - C_r}$	Length cm	Time sec	$Dx10^5$ $\frac{cm^2}{sec}$
8-1	119.1	0.65296	3.526	36,345	3.236
-2	119.1	0.66719	3.586	36,345	3.078
-4	119.1	0.63762	3.490	36,345	3.455
1.307 ± 0.005 Weight Percent Cadmium (2.309 Atomic Percent)					
10-1	50.0	0.76383	3.574	31,035	1.803
-2	50.0	0.75661	3.536	31,035	1.873
-3	50.0	0.74509	3.399	31,035	1.900
-4	50.0	0.76875	3.515	31,035	1.672
11-1	50.0	0.74975	3.528	42,135	1.453
-2	50.0	0.73135	3.523	42,135	1.670
-3	50.0	0.73976	3.543	42,135	1.585
-4	50.0	0.73122	3.536	42,135	1.684
12-1	70.2	0.73602	3.594	30,585	2.310
-2	70.2	0.70454	3.419	30,585	2.620
-3	70.2	0.70802	3.513	30,585	2.823
-4	70.2	0.71282	3.510	30,585	2.610
13-1	93.2	0.69736	3.581	43,580	2.116
-2	93.2	0.68722	3.536	43,580	2.203
-3	93.2	0.68857	3.505	43,580	2.148
-4	93.2	0.68138	3.518	43,580	2.263
14-1	119.1	0.68925	3.513	31,510	2.971
-2	119.1	0.68384	3.510	31,510	3.069
-3	119.1	0.73621	3.500	31,510	2.125
-4	119.1	0.68408	3.495	31,510	3.039
2.646 ± 0.010 Weight Percent Cadmium (4.626 Atomic Percent)					
15-1	50.0	0.74947	3.569	40,755	1.541
-2	50.0	0.74162	3.533	40,755	1.605
-3	50.0	0.72798	3.548	40,755	1.795
-4	50.0	0.73301	3.503	40,755	1.686
16-1	70.2	0.71529	3.414	28,340	2.620
-2	70.2	0.70814	3.480	28,340	2.859
-3	70.2	0.71114	3.487	28,340	2.810
-4	70.2	0.71106	3.510	28,340	2.852

TABLE I-II--Continued

Run-Capillary Number	Temp. °C	$\frac{C_f - C_r}{C_o - C_r}$	Length cm	Time sec	$Dx10^5 \frac{cm^2}{sec}$
17-1	93.2	0.71148	3.490	42,985	1.853
-2	93.2	0.72222	3.510	42,985	1.737
-3	93.2	0.70648	3.512	42,985	1.943
-4	93.2	0.70265	3.505	42,985	1.985
18-1	119.1	0.67947	3.526	29,440	3.408
-2	119.1	0.66777	3.490	29,440	3.587
-3	119.1	0.66569	3.498	29,440	3.649
-4	119.1	0.64194	3.309	29,440	3.745
4.275 ± 0.004 Weight Percent Cadmium (7.382 Atomic Percent)					
20-1	70.0	0.74343	3.574	32,780	2.015
-2	70.0	0.74124	3.515	32,780	1.983
-3	70.0	0.75766	3.515	32,780	1.738
-4	70.0	0.75457	3.487	32,780	1.755
21-1	93.2	0.76094	3.561	35,685	1.599
-2	93.2	0.75351	3.536	35,685	1.671
-3	93.2	0.75420	3.543	35,685	1.669
-4	93.2	0.74195	3.505	35,685	1.800
22-1	119.1	0.73302	3.500	33,090	2.073
-2	119.1	0.72963	3.487	33,090	2.109
-3	119.1	0.72687	3.500	33,090	2.169
-4	119.1	0.73032	3.508	33,090	2.124

APPENDIX J

Analysis of Errors in Diffusion Measurements

Estimation of the Expected Error in Measurements of the Diffusion Coefficient

An estimate of the random errors associated with the measurement and calculation of the diffusion coefficient can be made from examination of the individual parameters involved. As indicated in Appendix E, the important parameters in such an estimate of error are the capillary length, time duration of the diffusion, and the quantity θ which is dependent upon the measured concentration ratio.

The estimated error in the capillary length is ± 0.1 percent and that in the time measurement is from ± 0.01 to ± 0.04 percent, depending upon the elapsed time of the diffusion. The error associated with the concentration ratio is estimated to be ± 1 percent as an average since it is dependent upon the number of counts used to determine the radioactive count rate. The number of counts taken in these measurements was varied with the specific activity of the amalgam to give a reasonable counting time for the capillary activity evaluation.

Examination of the solution to the mathematical analysis of capillary-reservoir diffusion presented in Appendix E indicates the functional dependence of θ on ψ to be such that any given error in the ψ value will be amplified in the variation of θ. This amplified error effect in the θ value, which is directly proportional to the diffusion coefficient, is also a function of ψ itself. Using the tabulated solution from Appendix E, the following table has been prepared to show the percent variation in θ as a function of ψ for a 1 percent error in ψ.

TABLE J-I

PERCENT ERROR IN THE θ TERM OF THE DIFFUSION EQUATION FOR A ONE PERCENT ERROR IN ψ, AS A FUNCTION OF ψ

ψ	Percent Error in θ
0.90	± 17.72
0.85	± 11.30
0.80	± 7.96
0.75	± 6.02
0.70	± 4.67
0.65	± 3.69
0.60	± 3.02
0.55	± 2.45
0.50	± 2.01
0.45	± 1.70
0.40	± 1.41
0.35	± 1.19

The errors involved in the capillary length and diffusion time measurements are negligible compared to the variation in θ. Thus, the random error expected in the diffusion measurements will be essentially the error in determining θ and will vary with the magnitude of the concentration ratio as is evident in Table J-I above.

Calculation of the Standard Deviation of the Diffusion Coefficients

An indication of the random error in the diffusion measurements can be obtained by calculating the standard deviation of the diffusion coefficients which were presented in Tables H-II and I-II. The standard deviation is defined as

$$\sigma = \sqrt{\frac{\Sigma(D^2) - (\Sigma D)^2/n}{n - 1}} ,$$

where σ is the standard deviation of the individual diffusion

coefficients, D, and n is the number of determinations. The random error associated with the determination of the average value of the diffusion coefficient at the various temperatures and concentrations is calculated as the standard deviation of the average, $\bar{\sigma}$, defined as

$$\bar{\sigma} = \frac{\sigma}{\sqrt{n}} .$$

These two statistical quantities have been calculated for the diffusion data and are presented in Tables J-II and J-III.

A joint standard deviation of the diffusion coefficient can be calculated from the results given in Tables J-II and J-III for the individual data groups.

$$\sigma_j^2 = \frac{(n_1-1)\sigma_1^2+(n_2-1)\sigma_2^2+\ldots+(n_t-1)\sigma_t^2}{n_1+n_2+\ldots+n_t-t}$$

In the above equation, σ_j is the joint standard deviation, n is the number of measurements in each group, the subscripts indicate the data groups, and t is the total number of such groups. Thus, joint estimates of the deviation in the data can be made as a function of temperature or concentration, and these quantities are given in Tables J-IV and J-V. Also, an over-all estimate for the deviation of all the data was calculated to be $\pm$ 0.180 $\times$ 10^{-5} cm^2/sec for the zinc self-diffusion coefficients in zinc amalgams and $\pm$ 0.204 $\times$ 10^{-5} cm^2/sec for the cadmium self-diffusion coefficients in cadmium amalgams.

TABLE J-II

STANDARD DEVIATION OF THE SELF-DIFFUSION COEFFICIENTS OF ZINC IN ZINC AMALGAMS

Temperature °C	Number of Determinations	Standard Deviation $10^5 cm^2/sec$	Standard Deviation of the Average
0.0694 Atomic Percent Zinc			
50.0	37	0.172	0.028
70.2	4	0.039	0.020
93.2	4	0.047	0.024
119.1	2	0.202	0.142
0.3597 Atomic Percent Zinc			
50.0	4	0.105	0.052
70.2	4	0.121	0.060
93.2	4	0.265	0.132
119.1	1	--	--
1.750 Atomic Percent Zinc			
50.0	4	0.114	0.057
70.2	4	0.202	0.100
93.2	4	0.045	0.023
119.1	4	0.264	0.132
3.473 Atomic Percent Zinc			
50.0	11	0.090	0.027
70.2	4	0.037	0.019
93.2	4	0.119	0.060
119.1	1	--	--
5.609 Atomic Percent Zinc			
50.0	4	0.092	0.046
70.2	4	0.138	0.069
93.2	4	0.112	0.056
119.1	3	0.316	0.183

TABLE J-III

STANDARD DEVIATION OF THE SELF-DIFFUSION COEFFICIENTS OF CADMIUM IN CADMIUM AMALGAMS

Temperature °C	Number of Determinations	Standard Deviation $10^5 cm^2/sec$	Standard Deviation of the Average
0.08892 Atomic Percent Cadmium			
50.0	4	0.706	0.353
70.2	4	0.338	0.169
93.2	4	0.186	0.108
119.1	4	0.282	0.141
0.4741 Atomic Percent Cadmium			
50.0	4	0.198	0.099
70.2	4	0.389	0.194
93.2	3	0.235	0.136
119.1	3	0.189	0.109
2.309 Atomic Percent Cadmium			
50.0	8	0.150	0.053
70.2	4	0.211	0.106
93.2	4	0.065	0.032
119.1	4	0.453	0.226
4.626 Atomic Percent Cadmium			
50.0	4	0.110	0.055
70.2	4	0.112	0.056
93.2	4	0.110	0.055
119.1	4	0.142	0.071
7.382 Atomic Percent Cadmium			
70.2	4	0.147	0.073
93.2	4	0.084	0.042
119.1	4	0.040	0.020

TABLE J-IV

JOINT STANDARD DEVIATION FOR THE DIFFUSION COEFFICIENTS VERSUS TEMPERATURE

Temperature °C	Joint Standard Deviation for Zinc Coefficients $\frac{cm^2}{sec} \times 10^5$	Joint Standard Deviation for Cadmium Coefficients $\frac{cm^2}{sec} \times 10^5$
50.0	0.150	0.170
70.2	0.124	0.221
93.2	0.142	0.142
119.1	0.445	0.266

TABLE J-V

JOINT STANDARD DEVIATION FOR THE DIFFUSION COEFFICIENTS VERSUS CONCENTRATION

Concentration, Atomic Percent	Joint Standard Deviation $cm^2/sec \times 10^5$
Zinc Amalgams	
0.0694	0.161
0.3597	0.178
1.750	0.177
3.473	0.089
5.609	0.310
Cadmium Amalgams	
0.08892	0.264
0.4741	0.212
2.309	0.239
4.626	0.119
7.382	0.100

APPENDIX K

Thermodynamic Quantities Calculated From Concentration Cell Potentials For Cadmium and Zinc Amalgams At Various Temperatures

Sample Calculations

For the concentration cell

Zn-Hg (saturated amalgam) | Zn^{++} 0.1 M | Zn-Hg (dilute amalgam),

the thermodynamic activity of zinc in the dilute amalgam, a_{Zn}, may be calculated as

$$\ln \frac{a_{Zn}}{a_{Zn,s.s.}} = -\frac{nFE}{RT} .$$

$a_{Zn,s.s.}$ = thermodynamic activity of zinc in the standard state; in this case, the saturated amalgam at the given temperature ($a_{Zn,s.s.} = 1$).

n = number of equivalent per mole, 2 in this case (also for cadmium).

F = 23,074 calories/ volt-equivalent.

E = cell potential in volts.

R = gas constant, 1.987 cal/mole-°K.

T = temperature in °K.

Substitution of numerical values for constants in the equation gives

$$\log_{10} a_{Zn} = -10.085 \frac{E}{T} , \text{ for E in millivolts.}$$

This equation was used to calculate the thermodynamic activities of zinc or cadmium in their respective amalgams using the measured emf values given in Tables K-I and K-II. Results of these activity calculations are presented in the same tables.

The activity coefficients given in Tables K-I and K-II were calculated from the defining equation,

$$\gamma = \frac{a}{N} .$$

γ = activity coefficient of zinc or cadmium in the amalgam.

a = activity of zinc or cadmium in the amalgam.

N - atomic fraction of zinc or cadmium in the amalgam.

The thermodynamic factor, dlna/dlnN, in the diffusion equation may be calculated from values of the activity coefficient as a function of the atomic fraction.

$$\frac{d\ln a}{d\ln N} = 1 + N \frac{d\ln\gamma}{dN}$$

$$= 1 + 2.303N \frac{d\log_{10}\gamma}{dN}$$

Values of $\log_{10}\gamma$ were plotted parametrically in temperature against the atomic fraction of solute for each amalgam system. The slopes of these curves were determined to give $d\log_{10}\gamma/dN$ for various values of N. Using the above equation, the thermodynamic factor was evaluated and these results are included as Table K-III of this appendix.

TABLE K-I

THERMODYNAMIC ACTIVITIES AND ACTIVITY COEFFICIENTS OF ZINC IN ZINC AMALGAMS, CALCULATED FROM CONCENTRATION CELL POTENTIALS AT VARIOUS TEMPERATURES

Weight Percent Zinc	Atomic Percent Zinc	emf mv	Activity of Zinc	Activity Coefficient of Zinc
		50.0°C		
3.348*	9.608	0.000	1.0000	10.408
3.300	9.479	0.130	0.9907	10.452
3.198	9.204	0.412	0.9708	10.548
3.000	8.667	1.000	0.9308	10.740
2.980	8.613	1.050	0.9273	10.766
2.949	8.528	1.132	0.9219	10.810
2.898	8.389	1.296	0.9111	10.861
2.496	7.283	2.652	0.8265	11.348
1.900	5.609	5.702	0.6639	11.836
1.500	4.464	8.020	0.5620	12.590
1.150	3.447	11.060	0.4518	13.107
0.970	2.918	13.111	0.3894	13.345
0.799	2.412	15.245	0.3345	13.868
0.577	1.750	19.420	0.2488	14.217
0.400	1.217	24.120	0.1768	14.528
0.250	0.763	30.290	0.1135	14.875
0.170	0.520	35.520	0.07793	14.987
0.114	0.349	41.400	0.05109	14.639
0.0864	0.265	44.890	0.03976	15.004
0.0510	0.156	52.130	0.02363	15.147
0.0400	0.123	55.530	0.01851	15.049
0.0214	0.0656	64.280	0.009873	15.050
0.0150	0.0460	69.320	0.006874	14.943
		70.2°C		
4.645*	13.003	0.000	1.0000	7.691
4.630	12.965	0.030	0.9980	7.698
4.501	12.634	0.247	0.9834	7.784
4.252	11.992	0.797	0.9475	7.901
3.996	11.325	1.321	0.9145	8.075

*Equilibrium solubility at temperature.

TABLE K-I--Continued

Weight Percent Zinc	Atomic Percent Zinc	emf mv	Activity of Zinc	Activity Coefficient of Zinc
3.720	10.599	2.097	0.8678	8.188
2.992	8.646	3.904	0.7680	8.883
2.898	8.389	4.217	0.7519	8.963
2.496	7.283	5.718	0.6793	9.327
1.900	5.609	9.021	0.5436	9.692
1.378	4.111	12.890	0.4182	10.173
0.799	2.412	19.250	0.2720	11.277
0.400	1.217	28.675	0.1438	11.816
0.250	0.763	35.280	0.09201	12.059
0.170	0.520	40.860	0.06309	12.133
0.0864	0.265	50.860	0.03208	12.106
0.0510	0.156	58.470	0.01918	12.295
0.0400	0.123	62.100	0.01500	12.195
0.0214	0.0656	71.570	0.007907	12.053
		93.2°C		
6.540*	17.676	0.000	1.0000	5.657
6.495	17.569	0.065	0.9959	5.669
6.201	16.864	0.496	0.9691	5.747
5.995	16.366	0.802	0.9504	5.807
5.300	14.656	1.899	0.8866	6.049
5.000	13.904	2.366	0.8608	6.191
4.530	12.709	3.315	0.8105	6.377
3.720	10.599	5.192	0.7197	6.790
3.000	8.667	7.170	0.6349	7.325
2.496	7.283	9.203	0.5581	7.663
1.900	5.609	12.734	0.4462	7.955
1.378	4.111	16.610	0.3490	8.489
0.799	2.412	23.810	0.2212	9.171
0.400	1.217	33.910	0.1166	9.581
0.250	0.763	40.960	0.07461	9.779
0.170	0.520	46.940	0.05108	9.823
0.0864	0.265	57.700	0.02583	9.747
0.0510	0.156	65.760	0.01550	9.936
0.0400	0.123	69.550	0.01219	9.911
0.0214	0.0656	79.590	0.006452	9.835

*Equilibrium solubility at temperature.

TABLE K-II

THERMODYNAMIC ACTIVITIES AND ACTIVITY COEFFICIENTS OF CADMIUM IN CADMIUM AMALGAMS, CALCULATED FROM CONCENTRATION CELL POTENTIALS AT VARIOUS TEMPERATURES

Weight Percent Cadmium	Atomic Percent Cadmium	emf mv	Activity of Cadmium	Activity Coefficient of Cadmium
		50.0°C		
9.710*	16.102	0.000	1.0000	6.210
9.435	15.677	0.502	0.9646	6.153
9.009	15.016	1.304	0.9106	6.064
8.005	13.442	3.419	0.7822	5.819
6.975	11.802	5.780	0.6602	5.594
5.860	9.998	8.745	0.5335	5.336
2.964	5.169	19.340	0.2492	4.821
1.488	2.665	28.935	0.1251	4.766
0.295	0.525	51.635	0.02449	4.760
0.0587	0.105	74.060	0.004890	4.657
		70.2°C		
13.758*	22.161	0.000	1.0000	4.512
13.476	21.750	0.390	0.9740	4.478
12.990	21.038	1.082	0.9294	4.418
12.000	19.573	2.682	0.8341	4.261
9.999	16.546	6.934	0.6257	3.782
9.009	15.016	8.974	0.5450	3.629
8.005	13.442	11.258	0.4670	3.474
6.975	11.802	13.613	0.3983	3.375
5.860	9.998	16.655	0.3242	3.243
2.964	5.169	27.535	0.1553	3.004
1.488	2.625	38.000	0.07655	2.916
0.295	0.525	62.130	0.01497	2.851
0.0587	0.105	86.420	0.002896	2.758

*Equilibrium solubility at temperature.

TABLE K-II--Continued

Weight Percent Cadmium	Atomic Percent Cadmium	emf mv	Activity of Cadmium	Activity Coefficient of Cadmium
		93.2°C		
19.310*	29.927	0.000	1.0000	3.341
19.299	29.912	0.015	0.9990	3.340
18.500	28.831	1.161	0.9291	3.223
18.042	28.205	1.845	0.8897	3.154
18.000	28.148	1.851	0.8893	3.159
17.286	27.165	2.775	0.8387	3.087
15.739	25.001	5.411	0.7097	2.839
14.437	23.143	7.457	0.6234	2.694
12.000	19.573	11.821	0.4728	2.416
9.999	16.546	15.815	0.3671	2.219
9.009	15.016	17.950	0.3206	2.135
8.005	13.442	20.395	0.2746	2.043
6.975	11.802	22.900	0.2343	1.985
5.860	9.998	26.080	0.1915	1.915
2.964	5.169	37.750	0.09144	1.769
1.488	2.625	48.905	0.04510	1.718
0.295	0.525	74.630	0.008834	1.683
0.0587	0.105	100.030	0.001767	1.683

*Equilibrium solubility at temperature.

TABLE K-III

VALUES OF THE THERMODYNAMIC FACTOR IN THE DIFFUSION EQUATION CALCULATED FROM CONCENTRATION CELL POTENTIALS FOR CADMIUM AND ZINC AMALGAMS AT VARIOUS TEMPERATURES

Atomic Percent Solute	Thermodynamic Factor		
	50.0°C	70.2°C	93.2°C
	Zinc Amalgams		
0.00	1.000	1.000	1.000
2.00	0.912	0.919	0.925
4.00	0.824	0.838	0.851
6.00	0.776	0.764	0.776
8.00	0.720	0.733	0.711
9.60	0.664		
10.00		0.692	0.664
12.00		0.630	0.655
13.00		0.599	
14.00			0.645
16.00			0.617
17.67			0.577
	Cadmium Amalgams		
0.00	1.000	1.000	1.000
2.00	1.012	1.020	1.017
4.00	1.036	1.042	1.041
6.00	1.098	1.073	1.083
8.00	1.153	1.114	1.118
10.00	1.226	1.189	1.200
12.00	1.296	1.274	1.262
14.00	1.371	1.358	1.326
16.00	1.424	1.479	1.416
18.00		1.580	1.506
20.00		1.695	1.589
22.00		1.765	1.684
24.00			1.757
26.00			1.820
28.00			1.883
29.90			1.943