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and Progress Reports

Solubility and Diffusivity of Inert Gases in Liquid Sodium, Potassium, and NaK

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CONTENTS

	Page
Abstract	5
I. Introduction	7
II. Published Data	9
A. Solubility	9
B. Diffusivity	11
III. Discussion and Recommendations	13
A. Solubility	13
1. Extrapolation of Data	13
2. Estimated Values	14
B. Diffusivity	21
IV. References	25
Appendix. Solubility of Inert Gases in Liquid Metals	27

TABLES

1. Systems and Temperature Ranges that have been Investigated Concerning the Solubilities of Inert Gases in Liquid Sodium, Potassium, and NaK	13
2. Arrhenius-Type Equations for Solubility of Inert Gases in Liquid Metals (Moles Gas Dissolved per Mole Liquid Metal per Atmosphere Gas Pressure)	17
3. Atomic Radii of Inert Gas and Liquid Metal Atoms	18

FIGURES

1. Solubility of Inert Gases in Liquid Metals (log solubility plotted against reciprocal of absolute temperature)	15
2. Diffusivity of Inert Gases in Liquid Metals (plotted as a function of degrees centigrade)	23

ABSTRACT

The literature was surveyed for data on the solubility and diffusivity of rare gases and nitrogen in liquid sodium, potassium, and NaK. Experimental data were found for the solubilities of helium, argon, krypton, xenon, and nitrogen in liquid sodium; solubility data also were found for helium in liquid potassium. Estimates of solubility were made for the solubility of helium and of argon in eutectic NaK. Nitrogen appears to dissolve as the dimer, while the rare gases dissolve as the monomer. The solubilities of rare gases increased in a consistent manner with decreasing atomic radius of the rare-gas solute atom; conversely, the solubility of a given gas decreased with decreasing atomic radius of the solvent liquid-metal atom. Diffusivity values were calculated for the systems of interest using the Stokes-Einstein equation, derived from Fick's Law.

I. INTRODUCTION

Liquid sodium and NaK are used as heat-transfer media in a number of reactors and are the coolants of primary interest for the Liquid Metal Fast Breeder Reactor (LMFBR) Program. Because sodium and NaK react chemically with the oxygen, moisture, and carbon dioxide in air, it is customary to provide a protective blanket or cover of one of the nonreactive gases (such as argon, helium, or nitrogen) above any free surface of the liquid metal. While the inert gases will not react with sodium or NaK, some of the gas can either dissolve or become entrained in the liquid metal. The accumulation and sudden release of gas in the liquid metal coolant stream of a fast reactor: (1) might interfere with heat transfer by interposing a gaseous layer of relatively high thermal resistance, or (2) can cause reactivity changes in the reactor core leading to undesirable transients in power level. For this reason, there is a need for information on the part of reactor designers and operators about the magnitude of the solubility of inert gases in sodium and NaK, the temperature variation of solubility, and the diffusion of inert gases in liquid metals under a concentration gradient. In addition, two inert gases, krypton and xenon, are born in fission; information on solubility and diffusivity of these gases could help to determine the concentration of these two fission gases in the liquid metal bond of a fuel element, and in the case of vented or failed fuel elements the movement of these fission gases in the coolant stream.

The Liquid Metals Information Center* has received a number of inquiries on topics involving the solubility and diffusivity of inert gases in sodium and NaK. It is the purpose of this report to summarize existing data on solubility and diffusivity of inert gases in sodium and potassium, to show the variation with temperature, to identify areas where data are lacking, and to estimate values for solubilities in eutectic NaK which might be utilized pending the availability of experimental data.

*The Liquid Metals Information Center (LMIC), included within The Liquid Metal Engineering Center (LMEC), is an information analysis center sponsored by the Division of Technical Information of the U.S. Atomic Energy Commission. It provides a focal point for technically evaluated information and data on liquid metal technology and components; initial emphasis is on sodium and NaK alloys as heat transfer and heat transport fluids.

II. PUBLISHED DATA

A. SOLUBILITY

An early attempt to calculate the solubility of an inert gas in sodium was made by Epstein^{(1)*} in 1952. Epstein estimated the solubility of helium gas in liquid sodium, at 1 atm pressure and temperatures of 450 and 900°F. His calculations indicated a solubility of $1.6 \times 10^{-11} \text{ cm}^3 \text{ He (STP)/cm}^3 \text{ Na}$ at 450°F and $1.3 \times 10^{-7} \text{ cm}^3 \text{ He (STP)/cm}^3 \text{ Na}$ at 900°F. The solubility increased with temperature, in contrast to water and other aqueous solutions where the solubility of gases decreases with temperature. Epstein realized that his calculations would be reasonably accurate only if the temperatures involved were not too far removed from the critical temperature of the gas (5.2°K for helium) and that there was the possibility of rather enormous errors in the extrapolation. He assumed that the logarithm of the mole fraction of helium in sodium could be in error by 50% at 450°F which meant that the solubility of helium could vary between 1×10^{-4} to $2 \times 10^{-18} \text{ cm}^3 \text{ He (STP)/cm}^3 \text{ Na}$ (experimental results quoted later in this report indicate a value of about $1 \times 10^{-5} \text{ cm}^3 \text{ He (STP)/cm}^3 \text{ Na}$ at this temperature).

Mitra published data in 1961, as part of his doctoral dissertation, on the solubility of xenon in liquid sodium, mercury, and bismuth.⁽²⁾ The experimental data for xenon in sodium covered the temperature range of 105 to 204°C (221 to 400°F). An "almost instantaneously" frozen aliquot of sodium metal saturated with radioactive xenon gas was placed in an evacuated vessel and the sodium was remelted to liberate dissolved xenon. The activity of the liberated xenon was then measured to determine the amount present.

Slotnick and his co-workers at Pratt and Whitney CANEL issued a report in 1965⁽³⁾ giving experimental data on the solubility of helium in liquid potassium over the temperature range of 482 to 704°C (900 to 1300°F) and of helium in liquid lithium over the temperature range of 649 to 871°C (1200 to 1600°F). They used a predetermined amount of recirculated argon gas to strip the helium from saturated aliquot samples.

*Superscripts refer to the references listed in Section IV of this report.

Vogel, et al., at Argonne National Laboratory⁽⁴⁾ published experimental data in 1964 covering the solubility of krypton in sodium over the temperature range of 200 to 480°C (392 to 896°F) and of argon in sodium over the temperature range 340 to 480°C (644 to 896°F). Veleckis, et al., at ANL⁽⁵⁾ published additional experimental data covering argon in sodium and extended the temperature range to cover 330 to 530°C (626 to 986°F). In 1968, Veleckis, et al., reported experimental data for the solubility of nitrogen in sodium⁽⁶⁾ over the temperature range of 450 to 600°C (842 to 1112°F). F.A. Cafasso at ANL is currently determining the solubility of helium in liquid sodium; preliminary data are published in a recent progress report.⁽⁷⁾

The procedure used at ANL was similar to that used by Pratt and Whitney, except that in certain instances radioactive gases were used to saturate the liquid metal and then the specific activity was measured after sparging.

In addition to the foregoing references covering direct investigations of the solubility of inert gas in sodium or NaK, there have been some scattered references to gas transport effects in a sodium or NaK system. For example, a number of brief power level transients were observed during the course of operation of the Sodium Reactor Experiment (SRE).⁽⁸⁾ Exploratory tests and analog computer-model simulation led to the belief that gas bubble passage through the core was the immediate cause of these period spikes. Investigators at Oak Ridge National Laboratory (ORNL)⁽⁹⁾ also observed the phenomenon of gas transport and accumulation during operation of a NaK test loop, using an argon cover gas. Subsequent evaluative tests indicated that argon gas actually was being dissolved at the hot NaK-cover-gas interface and afterwards released and trapped in pockets in cooler portions of the loop. Operation of the NaK loop under identical flow conditions but under isothermal conditions showed practically no transport of the argon gas, which ruled out entrainment as a major factor. These exploratory tests showed argon gas accumulation rates of 0.2 in.³ (STP)/hr and 1.5 in.³ (STP)/hr at loop temperature differences of 200°F (110°C) and 1100°F (610°C), respectively. Apparently no further tests were performed at ORNL to obtain additional quantitative values related to the solubility of argon in NaK, and no other experimental observations were found in the literature regarding the solubility of argon gas in either liquid potassium or in NaK.

B. DIFFUSIVITY

While extensive investigations have been made on the diffusion of gases into gases and to a lesser extent on diffusion of metals into liquid metals, no experimental data were found on rates of diffusion of gases into liquid metals. Brooks of Knolls Atomic Power Laboratory⁽¹⁰⁾ calculated the rate of escape of fission gases by diffusion through a sodium-filled crack in the cladding of a fuel element. In a numerical example, Brooks gave a diffusion coefficient of 0.5×10^{-5} cm²/sec for fission gases in sodium, but he did not reveal how this value was derived nor did he state the temperature involved.

Bonilla, et al., of Columbia University⁽¹¹⁾ used the familiar Stokes-Einstein equation⁽¹²⁾ to calculate the diffusivities of metals in liquid mercury, and good agreement was obtained between calculated and experimental values for the alkali and alkaline-earth metals as solute atoms in mercury. S. Epstein and J. Rutherford at Brookhaven National Laboratory⁽¹³⁾ have published experimental data on the diffusivity of silver, cadmium, indium, tin, and antimony in liquid sodium. Diffusion coefficients ranged from 3×10^{-5} to 11×10^{-5} cm²/sec over the temperature range of 150 to 350°C (302 to 662°F). If the Stokes-Einstein equation is used to calculate these diffusion coefficients, it yields values ranging from 2×10^{-5} to 11×10^{-5} cm²/sec. These instances of good agreement between experimental and calculated values for solid solutes in liquid metals suggests that this equation may also yield reasonable estimates for gaseous solutes in liquid metals.

III. DISCUSSION AND RECOMMENDATIONS

A. SOLUBILITY

The literature survey revealed that experimental data have been published on the solubility in liquid sodium of helium, argon, krypton, xenon, and nitrogen; there were no experimental data for neon in sodium. No data were found for any of the above gases in NaK, while in potassium (the other constituent of NaK), experimental data were available only for helium. This situation is summarized in Table 1 where the approximate temperature range for which data are available is indicated in a matrix showing the gases and liquid metals of interest.

TABLE 1
SYSTEMS AND TEMPERATURE RANGES THAT HAVE BEEN INVESTIGATED
CONCERNING THE SOLUBILITIES OF INERT GASES IN LIQUID
SODIUM, POTASSIUM, AND NaK

Inert Gas	Liquid Metal					
	Sodium		Potassium		NaK	
	°C	°F	°C	°F	°C	°F
Helium	400-500	752-932	482-704	900-1300	—	—
Neon	—	—	—	—	—	—
Argon	330-530	626-986	—	—	—	—
Krypton	200-480	392-896	—	—	—	—
Xenon	105-204	221-400	—	—	—	—
Radon	—	—	—	—	—	—
Nitrogen	450-600	842-1112	—	—	—	—

1. Extrapolation of Data

It may be seen from Table 1 that there are a number of gaps in the data and even where data are available, the temperature range of interest in LMFBR systems (up to, say, 1200°F) is not covered. It would be of value then to

(1) assess the validity of extrapolating the available data over a wider temperature range, and (2) estimate values for potassium and NaK for at least the two common cover gases, argon and helium.

Least-square-fit equations are plotted in Figure 1 for the five sodium cases and the one potassium case (also for the one lithium case) for which experimental data on gas solubility were available; they are plotted to show log solubility versus the reciprocal of the absolute temperature over the applicable temperature range. Where data are given in the original reference, these data points are also shown in Figure 1. It may be seen that the experimental data points themselves show good linearity plotted in this manner. Thus it appears that pending the availability of additional experimental data, these straight-line plots should give good estimates of solubilities when extrapolated over the ranges where sodium and potassium are liquid.

2. Estimated Values

With solubility values available for helium in both sodium and potassium, it should be possible to estimate reasonably accurate solubility values for helium in NaK. It has been observed that a number of physical properties (e.g., density, specific heat) of a given NaK alloy can be calculated rather accurately by multiplying the molar concentration of sodium and potassium by the specific property value of each of the pure constituents.^(14,15,16) This indicates that in the NaK alloy, the intrinsic interatomic spacing of the two constituents is not altered significantly. Therefore, the solvent capacity of each constituent should remain unchanged. On this basis, it was assumed that the solubility of helium in NaK could be obtained by multiplying the solubility values of helium in pure sodium and in pure potassium by the molar concentration of that particular constituent in the NaK alloy of interest. This was done for eutectic NaK, which corresponds closely to two moles of potassium and one mole of sodium. Values were calculated at 350, 400, 450, and 500°C, and an equation for NaK was derived from these points by the method of least squares.

It was desired also to estimate the solubility of argon in eutectic NaK, using a procedure similar to that described above, in order to answer an inquiry on this topic received by the Liquid Metals Information Center. However, while

7693-4724

Figure 1. Solubility of Inert Gases in Liquid Metals
(log solubility plotted against reciprocal of absolute temperature)

data were available on the solubility of argon in sodium, there were no data on the solubility of argon in potassium, so it was first necessary to estimate these values over the temperature range of interest. It has been demonstrated empirically that the relative order of solubility of several gases in different liquids is almost always the same and, moreover, that the ratios of these solubilities in different liquids are approximately proportional.⁽¹⁷⁾ Since experimental solubility data are available for the helium-in-sodium, helium-in-potassium, and argon-in-sodium systems, this relationship was used to estimate the solubility of argon in potassium using these ratios. Solubility values of argon in potassium were calculated for temperatures of 350, 400, 450, and 500°C; and an equation was derived by the method of least squares. Next, values for the solubility of argon in eutectic NaK were calculated, based on the molar concentration of each NaK constituent, as described above for helium.

The equations derived for the estimated solubilities of helium in NaK-78, argon in potassium, and argon in NaK-78 have been plotted in Figure 1, along with plots of the equations for the other systems obtained from published experimental data. The solubility of helium in NaK-78 was estimated to be slightly less than that reported for helium in potassium, and the solubilities of argon in either potassium or NaK-78 were estimated to be almost the same as that reported for helium in sodium. It would be desirable to verify these estimated solubility values, since one or more of these systems may some day be of particular interest in the design of reactors for space applications. In addition, it would be advisable to experimentally verify any of the diffusivity values in these ten systems where greater accuracies than the calculated values are required.

Table 2 lists least-square-fit equations for each of the ten inert gas-liquid metal systems described herein. The equations are listed in order of decreasing solubility and are applicable at an inert gas pressure of 1 atm. These equations were either given in the literature or derived by the method of least squares from experimental or estimated values, as described previously. The equation for xenon in sodium shows the highest solubility, while nitrogen in sodium shows the lowest.

Except for the reported solubility of xenon in sodium and for nitrogen in sodium, there appears to be a consistent relationship between solubility and the

TABLE 2

ARRHENIUS-TYPE EQUATIONS FOR SOLUBILITY OF INERT GASES
IN LIQUID METALS (MOLES GAS DISSOLVED PER MOLE
LIQUID METAL PER ATMOSPHERE GAS PRESSURE)

-
1. Xenon in Sodium: $\text{Log } S = -3.248 - 522/T \text{ (}^\circ\text{K)}$
[Taken from Reference 2. Assumed valid from 350 to 500°C (662 to 932°F).]
 2. Helium in Potassium: $\text{Log } S = -3.399 - 1530/T \text{ (}^\circ\text{K)}$
[Derived from data in Reference 3. Valid from 482 to 704°C (900 to 1300°F). Extrapolated to 350°C (662°F).]
 3. Helium in NaK-78: $\text{Log } S = -3.580 - 1517/T \text{ (}^\circ\text{K)}$
[Derived from data in References 3 and 7. Assumed valid from 350 to 500°C (662 to 932°F).]
 4. Argon in Potassium: $\text{Log } S = -1.739 - 3702/T \text{ (}^\circ\text{K)}$
[Derived from data in References 3, 5, and 7. Assumed valid from 350 to 500°C (662 to 932°F).]
 5. Argon in NaK-78: $\text{Log } S = -1.880 - 3718/T \text{ (}^\circ\text{K)}$
[Derived from data in References 3, 5, and 7. Assumed valid from 350 to 500°C (662 to 932°F).]
 6. Helium in Sodium: $\text{Log } S = -3.404 - 2654/T \text{ (}^\circ\text{K)}$
[Derived from data in Reference 7. Valid from 400 to 500°C (752 to 932°F). Extrapolated to 350°C (662°F).]
 7. Argon in Sodium: $\text{Log } S = -2.130 - 4542/T \text{ (}^\circ\text{K)}$
[Taken from Reference 5. Valid from 330 to 530°C (626 to 986°F).]
 8. Helium in Lithium: $\text{Log } S = -7.323 - 846/T \text{ (}^\circ\text{K)}$
[Derived from data in Reference 3. Valid from 649 to 871°C (1200 to 1600°F).]
 9. Krypton in Sodium: $\text{Log } S = -4.188 - 3583/T \text{ (}^\circ\text{K)}$
[Taken from Reference 4. Valid from 200 to 480°C (392 to 896°F). Extrapolated to 500°C (932°F).]
 10. Nitrogen in Sodium: $\text{Log } S = -7.352 - 2716/T \text{ (}^\circ\text{K)}$
[Derived from data in Reference 6. Valid from 450 to 600°C (842 to 1112°F).]
-

respective atomic radius of the solute gas atom and the liquid metal solvent atom in the different gas-liquid systems. The atomic radius of these elements is shown in Table 3, arranged in increasing order of the solute and solvent atoms.

TABLE 3
ATOMIC RADII OF INERT GAS AND LIQUID METAL ATOMS⁽¹⁸⁾

Element	Atomic Radius (Å)	Remarks
<u>Solute Atoms:</u>		
Nascent Nitrogen (N)	0.70	Interatomic Spacing = 1.15 Å
Helium	0.93	
Nitrogen Gas (N ₂)	-	
Argon	1.54	
Krypton	1.69	
Xenon	1.90	
<u>Solvent Atoms:</u>		
Lithium	1.52	
Sodium	1.86	
Potassium	2.31	

For any given liquid metal solvent, such as sodium for example, the smaller the atomic radius of the inert gas solute atom the higher the solubility. Thus, helium was found to have the highest solubility in sodium (neglecting xenon), argon the next highest solubility, and krypton the lowest solubility. Conversely, for a given inert gas solute atom, such as helium, the larger the atomic radius of the solvent atom the higher the solubility. Thus, helium in potassium was found to have a higher solubility at a given temperature than helium in sodium or helium in lithium. The solubilities reported for xenon in sodium do not fit this pattern and on this basis may be in error; xenon has a large atomic radius, even greater than krypton. Because of the importance of xenon fission gas behavior in fast breeder reactor studies, it would be desirable to recheck the anomalous appearing solubility data reported by Mitra⁽²⁾ for xenon in sodium. The consistent relationships exhibited between atomic radii and solubility in the

other rare-gas, liquid-metal systems leads to the thought that the solubility of xenon in sodium should be lower than that reported for krypton in sodium.

The published solubility results for nitrogen also do not fit but this possibly may be explained on the basis that nitrogen dissolves in sodium as the diatomic molecule (N_2) rather than as nascent nitrogen (N), which appears to be the case. The interatomic spacing of atoms in a nitrogen molecule is given as 1.15 Å, but the effective radius for solubility could very well be larger than krypton and thus be consistent with the pattern. This question of the nature of nitrogen solubility in sodium was investigated by ANL⁽⁶⁾ using mixtures of nitrogen-14 and nitrogen-15 gas. Their results indicated that nitrogen apparently does dissolve in sodium as the diatomic or molecular species. The following statement is made on page 4 of Reference 6 regarding the dissolution of nitrogen in sodium: "The results, therefore, appear to be consistent with the mechanism of diatomic solution of molecular nitrogen in sodium. Past studies have shown that diatomic gases dissolve or react monatomically in liquid metals; this is the first to show that molecular dissolution also occurs." Thus, the possibility of an effective atomic radius for nitrogen in sodium, which is larger than the atomic radius of krypton, makes the lower solubility reported for nitrogen consistent with the radii relationships exhibited in the other systems (neglecting xenon in sodium, which is not consistent). In fact, the consistent atomic radii relationships exhibited by the other systems add additional credence to the conclusion that nitrogen dissolves in sodium as diatomic molecules rather than as monatomic atoms. On a monatomic dissolution basis (atomic radius 0.70 Å), one would expect to find the solubility of nitrogen in sodium to be higher than that of either helium, argon, or krypton in sodium.

Examination of Figure 1 shows that the solubility of the inert gases in liquid metals increases with temperature. This is in accordance with the behavior predicted by Epstein⁽¹⁾, who pointed out that observations based upon the behavior of gases in water and aqueous media can be misleading, as water has many peculiarities and one of them is a negative rather than a positive temperature coefficient for gas solubility.

As an example of the use of Figure 1, consider the change in the solubility of helium when sodium is cooled from 1200 down to 900°F, assuming a system

at atmospheric pressure. Using values from Figure 1, after first converting the temperature to degrees Kelvin, a value is obtained of 5.2×10^{-7} moles He/mole Na at 1200°F (649°C , 922°K) and 1.2×10^{-7} moles He/mole Na at 900°F (482°C , 755°K). Therefore, this temperature drop should result in the release of 4.0×10^{-7} moles He/mole Na. In a typical LMFBR at a flow rate of 16×10^6 lb Na/hr (40,000 gpm), this corresponds to the release of $\sim 1.6 \text{ ft}^3$ helium per minute. It should be noted that the solubility of inert gases in liquid metals approximates Henry's law;^(4,5) i. e., the solubility of a gas is directly proportional to the pressure of the particular solute gas above the solvent. Therefore, in making any solubility calculations, the effect of pressure, as well as temperature, must be considered. It should also be noted that Figure 1 assumes equilibrium conditions, which requires adequate time for diffusion and/or mixing by stirring.

Referring again to the ORNL tests in a NaK loop, where it was determined that argon cover gas was being dissolved in a surge tank and liberated in cooler portions of a loop, it was stated that 0.2 in.^3 of argon (STP)/hr were being liberated under the following loop conditions:⁽⁹⁾

- 1) 10 psig argon cover gas above surge tank
- 2) 0.07 gpm bypass NaK flow rate
- 3) 1350°F (732°C) NaK surface temperature in surge tank
- 4) 200°F (110°C) ΔT between surge tank surface and bypass exit.

The accumulation of argon gas in cooler portions of the loop was estimated by measuring the rise in NaK level in the surge tank and then calculating the volume change. Using Equation No. 5 in Table 2 derived for estimating the solubility of argon in NaK-78, approximately 0.8 in.^3 of argon (STP)/hr should have been liberated by a temperature change of 200°F (110°C) from 1350°F (732°C). This is a reasonably good check, considering the wide extrapolation of Equation No. 5 from 500°C up to 730°C that was required and considering the fact that the empirical solubility ratios used in the derivation of Equation No. 5 are approximately proportional and are not exact. Even so, the equations estimating the solubilities of helium in NaK-78, argon in NaK-78, and argon in potassium are believed to be valid within at least an order of magnitude

over the given temperature range, and should serve a useful purpose in making differential solubility calculations for reactors that employ these cover-gas, liquid-metal-coolant systems.

The Appendix to this report lists calculated inert gas solubility values at certain temperature intervals for the ten systems listed in Table 2. These solubilities are shown in units of: (1) moles of gas per mole of liquid metal; (2) moles of gas per liter of liquid metal; (3) cubic centimeters of gas at (STP) per cm³ of liquid metal; (4) cubic centimeters of gas, at temperature, per cm³ of liquid metal; and (5) parts per billion. These data are arranged in tables in the same order of decreasing system solubility as in Table 1.

B. DIFFUSIVITY

Diffusivity of materials is defined as the proportionality constant, or diffusion coefficient, in the general diffusion equation for material transfer:⁽¹⁹⁾

$$\frac{dQ}{dt} = -D (dy \, dz) \frac{dc}{dx},$$

where:

Q = moles of material diffusing through an area ($dy \, dz$) in the direction x in time t

$(dy \, dz)$ = area normal to material flow

t = time

x = length in direction of diffusion

$\frac{dc}{dx}$ = rate of increase of volume concentration in the x direction.

D = diffusion coefficient in units of length squared divided by time.

As pointed out in Section II B, no experimental data were found in the literature on the diffusivity of gases in liquid metals. However, Bonilla and others⁽¹¹⁾ made use of the Stokes-Einstein equation (derived from Ficks Law) to calculate

diffusivities and found good agreement for the alkali and alkaline-earth metals as solute atoms in mercury. The Stokes-Einstein equation is

$$D = \frac{RT}{N6\pi\eta r},$$

where

D = diffusivity (cm²/sec)

R = gas constant (8.3144 x 10⁷ dyne-cm/mole - °K)

T = temperature (°K)

N = Avogadro's number (6.02 x 10²³ atoms/mole)

η = solvent viscosity (poise units)

r = atomic radius of diffusing atom (Å, 10⁻⁸ cm)

References 20 and 21 give experimental values of approximately 2 x 10⁻⁵ and 5 x 10⁻⁵ cm²/sec for the diffusivities of oxygen and hydrogen, respectively, in water at ambient temperature. Use of the Stokes-Einstein equation gives calculated diffusivity values of approximately 3 x 10⁻⁵ cm²/sec for oxygen and 7 x 10⁻⁵ cm²/sec for hydrogen diffusing into water at ambient temperature, agreeing quite well with the experimental values. In view of this good agreement, the Stokes-Einstein equation was used to estimate diffusivities in the inert-gas, liquid metal systems.

Figure 2 shows curves of diffusivity vs temperature that were calculated for each system by means of the Stokes-Einstein equation. While some anomalies do exist, in general, these diffusivity curves have the same relative order as the solubility lines in Figure 1 (helium in lithium is higher than argon in sodium; xenon in sodium is less than krypton in sodium; and nitrogen, calculated using the diatomic interatomic spacing of 1.15 Å, shows intermediate diffusivity). It was pointed out previously that on the basis of atomic radii in the xenon-sodium and in the krypton-sodium systems, it appears that the solubility of xenon in sodium might be in error and should fall below that of krypton in sodium. In the case of nitrogen in sodium, the interatomic spacing doubtless does not give an equivalent radius for solubility comparisons. However, it is apparent that nitrogen

7693-4725

Figure 2. Diffusivity of Inert Gases in Liquid Metals
(plotted as a function of degrees centigrade)

dissolves in sodium as the diatomic molecule rather than as the monatomic species, so nitrogen would also be expected to diffuse in the diatomic state.

Referring to the diffusivity curves in Figure 2 and comparing them with the BNL⁽¹³⁾ experimental diffusivity values given before for silver, cadmium, indium, tin, and antimony in liquid sodium, the diffusivity of argon in sodium varies from $3.6 \times 10^{-5} \text{ cm}^2/\text{sec}$ at 150°C to $10 \times 10^{-5} \text{ cm}^2/\text{sec}$ at 350°C , which is excellent agreement for atoms with comparable atomic radii. The atomic radii of the delineated metal atoms range from a minimum of $1.40 \text{ \AA}$ for tin to a maximum of $1.62 \text{ \AA}$ for indium, compared with $1.54 \text{ \AA}$ for argon. Thus, there is some basis for belief that the curves for diffusivity of inert gases presented in Figure 2 should be valid within at least an order of magnitude, and they should be useful as preliminary data in estimating the rate of diffusion of cover gas atoms into liquid-metal-cooled reactor systems.

Finally, Camp and Johnson⁽²²⁾ present a method for the simultaneous determination of diffusivity and solubility in a single experiment. The method involves following the concentration of the diffusing component, or solute, in response to a step change in concentration. The method is applicable to solid or liquid solvent phases, while the solute may be gas or liquid; hence, it should be applicable to inert gases in liquid metals.

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APPENDIX

SOLUBILITY OF INERT GASES IN LIQUID METALS

In the following tables, solubilities are shown in convenient units and evaluated at certain temperatures. All values are at 1 atm gas pressure. Equations are least-square fits in moles gas dissolved per mole of liquid metal.

The reader is cautioned to refer to the text before applying the tabulated data; the significant figures tabulated result from substitution into the respective equations and are not intended to reflect the accuracy of the underlying data.

TABLE A-1
SOLUBILITY OF XENON IN SODIUM

$$\left(\text{Log } S = -3.248 - \frac{522}{T(^{\circ}\text{K})} \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles Xe/ Mole Na	Moles Xe/ Liter Na	cm^3 of Xe(STP)/ cm^3 of Na	cm^3 of Xe/ cm^3 of Na	ppb
350	662	8.20×10^{-5}	3.09×10^{-3}	6.92×10^{-2}	1.58×10^{-1}	4.68×10^5
400	752	9.46×10^{-5}	3.52×10^{-3}	7.88×10^{-2}	1.94×10^{-1}	5.40×10^5
450	842	1.04×10^{-4}	3.93×10^{-3}	8.80×10^{-2}	2.33×10^{-1}	6.11×10^5
500	932	1.19×10^{-4}	4.31×10^{-3}	9.65×10^{-2}	2.73×10^{-1}	6.80×10^5

TABLE A-2
SOLUBILITY OF HELIUM IN POTASSIUM

$$\left(\text{Log } S = - 3.399 - \frac{1530}{T(^{\circ}\text{K})} ; \text{ values below } 500^{\circ}\text{C are extrapolated} \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles He/ Mole K	Moles He/ Liter K	cm^3 of He(STP)/ cm^3 of K	cm^3 of He/ cm^3 of K	ppb
350	662	1.40×10^{-6}	2.72×10^{-5}	6.09×10^{-4}	1.39×10^{-3}	143
400	752	2.13×10^{-6}	4.07×10^{-5}	9.12×10^{-4}	2.25×10^{-3}	218
450	842	3.06×10^{-6}	5.75×10^{-5}	1.29×10^{-3}	3.42×10^{-3}	314
500	932	4.19×10^{-6}	7.75×10^{-5}	1.74×10^{-3}	4.93×10^{-3}	430
550	1022	5.52×10^{-6}	1.00×10^{-4}	2.24×10^{-3}	6.75×10^{-3}	566
600	1112	7.05×10^{-6}	1.26×10^{-4}	2.82×10^{-3}	9.02×10^{-3}	723
650	1202	8.77×10^{-6}	1.54×10^{-4}	3.45×10^{-3}	1.17×10^{-2}	899
700	1292	1.07×10^{-5}	1.85×10^{-4}	4.09×10^{-3}	1.46×10^{-2}	1097

TABLE A-3
SOLUBILITY OF HELIUM IN NaK-78

$$\left(\text{Log } S = - 3.580 - \frac{1517}{T(^{\circ}\text{K})} \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles He/ Mole NaK	Moles He/ Liter NaK	cm^3 of He(STP)/ cm^3 of NaK	cm^3 of He/ cm^3 of NaK	ppb
350	662	9.67×10^{-7}	2.23×10^{-5}	5.00×10^{-4}	1.14×10^{-3}	114
400	752	1.47×10^{-6}	3.36×10^{-5}	7.53×10^{-4}	1.86×10^{-3}	173
450	842	2.10×10^{-6}	4.73×10^{-5}	1.06×10^{-3}	2.81×10^{-3}	248
500	932	2.87×10^{-6}	6.36×10^{-5}	1.42×10^{-3}	4.02×10^{-3}	339

TABLE A-4
SOLUBILITY OF ARGON IN POTASSIUM

$$\left(\text{Log } S = -1.739 - \frac{3702}{T(^{\circ}\text{K})} \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles Argon/ Mole K	Moles Argon/ Liter K	cm^3 of Argon(STP)/ cm^3 of K	cm^3 of Argon/ cm^3 of K	ppb
350	662	2.09×10^{-8}	4.06×10^{-7}	9.09×10^{-6}	2.07×10^{-5}	21
400	752	5.76×10^{-8}	1.10×10^{-6}	2.46×10^{-5}	6.06×10^{-5}	59
450	842	1.38×10^{-7}	2.59×10^{-6}	5.80×10^{-5}	1.54×10^{-4}	140
500	932	2.97×10^{-7}	5.49×10^{-6}	1.23×10^{-4}	3.48×10^{-4}	303

TABLE A-5
SOLUBILITY OF ARGON IN NaK-78

$$\left(\text{Log } S = -1.880 - \frac{3718}{T(^{\circ}\text{K})} \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles Argon/ Mole NaK	Moles Argon/ Liter NaK	cm^3 of Argon(STP)/ cm^3 of NaK	cm^3 of Argon/ cm^3 of NaK	ppb
350	662	1.42×10^{-8}	3.30×10^{-7}	7.39×10^{-6}	1.69×10^{-5}	17
400	752	3.95×10^{-8}	9.03×10^{-7}	2.02×10^{-5}	4.98×10^{-5}	47
450	842	9.51×10^{-8}	2.14×10^{-6}	4.79×10^{-5}	1.27×10^{-4}	112
500	932	2.05×10^{-7}	4.54×10^{-6}	1.02×10^{-4}	2.89×10^{-4}	242

TABLE A-6
SOLUBILITY OF HELIUM IN SODIUM

$$\left(\text{Log } S = -3.404 - \frac{2654}{T(^{\circ}\text{K})}; (350^{\circ}\text{C data point extrapolated}) \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles He/ Mole Na	Moles He/ Liter Na	cm^3 of He(STP)/ cm^3 of Na	cm^3 of He/ cm^3 of Na	ppb
350	662	2.17×10^{-8}	8.19×10^{-7}	1.83×10^{-5}	4.18×10^{-5}	4
400	752	4.49×10^{-8}	1.67×10^{-6}	3.74×10^{-5}	9.22×10^{-5}	8
450	842	8.41×10^{-8}	3.09×10^{-6}	6.92×10^{-5}	1.83×10^{-4}	15
500	932	1.45×10^{-7}	5.25×10^{-6}	1.18×10^{-4}	3.34×10^{-4}	25

TABLE A-7
SOLUBILITY OF ARGON IN SODIUM

$$\left(\text{Log } S = -2.130 - \frac{4542}{T(^{\circ}\text{K})} \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles Argon/ Mole Na	Moles Argon/ Liter Na	cm^3 of Argon(STP)/ cm^3 of Na	cm^3 of Argon/ cm^3 of Na	ppb
330	626	2.18×10^{-10}	8.28×10^{-9}	1.85×10^{-7}	4.09×10^{-7}	0.4
350	662	3.80×10^{-10}	1.43×10^{-8}	3.20×10^{-7}	7.30×10^{-7}	0.7
400	752	1.32×10^{-9}	4.92×10^{-8}	1.10×10^{-6}	2.71×10^{-6}	2.3
450	842	3.87×10^{-9}	1.42×10^{-7}	3.18×10^{-6}	8.42×10^{-6}	6.7
500	932	9.87×10^{-9}	3.57×10^{-7}	8.00×10^{-6}	2.27×10^{-5}	17.1
530	986	1.64×10^{-8}	5.89×10^{-7}	1.32×10^{-5}	3.88×10^{-5}	28.5

TABLE A-8
SOLUBILITY OF HELIUM IN LITHIUM

$$\left(\text{Log } S = -7.323 - \frac{846}{T(^{\circ}\text{K})} \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles He/ Mole Li	Moles He/ Liter Li	cm^3 of He(STP)/ cm^3 of Li	cm^3 of He/ cm^3 of Li	ppb
650	1202	5.76×10^{-9}	3.89×10^{-7}	8.71×10^{-6}	2.94×10^{-5}	3.3
700	1292	6.43×10^{-9}	4.30×10^{-7}	9.63×10^{-6}	3.43×10^{-5}	3.7
750	1382	7.08×10^{-9}	4.70×10^{-7}	1.05×10^{-5}	3.93×10^{-5}	4.1
800	1472	7.75×10^{-9}	5.09×10^{-7}	1.14×10^{-5}	4.48×10^{-5}	4.5
850	1562	8.40×10^{-9}	5.47×10^{-7}	1.23×10^{-5}	5.06×10^{-5}	4.8

TABLE A-9
SOLUBILITY OF KRYPTON IN SODIUM

$$\left(\text{Log } S = -4.188 - \frac{3583}{T(^{\circ}\text{K})} \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles Kr/ Mole Na	Moles Kr/ Liter Na	cm^3 of Kr(STP)/ cm^3 of Na	cm^3 of Kr/ cm^3 of Na	ppb
200	392	1.73×10^{-12}	6.80×10^{-11}	1.52×10^{-9}	2.63×10^{-9}	0.006
250	482	9.14×10^{-12}	3.55×10^{-10}	7.95×10^{-9}	1.52×10^{-8}	0.03
300	572	3.62×10^{-11}	1.39×10^{-9}	3.11×10^{-8}	6.52×10^{-8}	0.1
350	662	1.15×10^{-10}	4.34×10^{-9}	9.72×10^{-8}	2.22×10^{-7}	0.4
400	752	3.08×10^{-10}	1.15×10^{-8}	2.58×10^{-7}	6.36×10^{-7}	1.1
450	842	7.17×10^{-10}	2.63×10^{-8}	5.89×10^{-7}	1.56×10^{-6}	2.6
500	932	1.50×10^{-9}	5.43×10^{-8}	1.26×10^{-6}	3.57×10^{-6}	5.5

TABLE A-10
SOLUBILITY OF NITROGEN IN SODIUM

$$\left(\text{Log } S = -7.352 - \frac{2716}{T(^{\circ}\text{K})} \right)$$

Temperature		Solubility (per atm)				
$^{\circ}\text{C}$	$^{\circ}\text{F}$	Moles N_2 / Mole Na	Moles N_2 / Liter Na	cm^3 of N_2 (STP)/ cm^3 of Na	cm^3 of N_2 / cm^3 of Na	ppb
450	842	7.79×10^{-12}	2.86×10^{-10}	6.41×10^{-9}	1.70×10^{-8}	0.009
500	932	1.36×10^{-11}	4.92×10^{-10}	1.10×10^{-8}	3.11×10^{-8}	0.017
550	1022	2.23×10^{-11}	7.96×10^{-10}	1.78×10^{-8}	5.37×10^{-8}	0.027
600	1112	3.45×10^{-11}	1.21×10^{-9}	2.71×10^{-8}	8.67×10^{-8}	0.042