NUCLEAR SCIENCE AND ENGINEERING: 18, 280-292 (1964)

Letters to the Editors

The Temperature Dependence of the Density of Liquid Metals

One of the most quantitative approaches used to formulate a density/temperature relation is that described recently by Ree.[1] (Other approaches to this problem have been discussed in the literature.[2-16]) By using the van der Waals, the Berthelot, and the Dieterici equations of state and by imposing the condition that the work done in an isothermal reversible cyclic process is zero, Ree derived the equation

$$\rho_r = 1 + ax^{\frac{1}{2}} + bx + cx^2 + dx^3, \qquad (1)$$

where ρ_r, the reduced liquid density, is the ratio of the density ρ divided by the critical density ρ_c, x is the expression $\left(1 - \frac{T}{T_c}\right)$ in which T_c is the Kelvin critical temperature, and the coefficients a, b, c and d are constants that have different numerical values in each of the above three equations of state. (Ree also derived a ρ-T relation for the vapor in equilibrium with the liquid, but we shall not be concerned with this relation in the present communication.) Ree found that, of the above equations of state, the Berthelot equation described best the existing data for some non-metallic materials. This equation does not, however, yield a reasonable value of ρ_c for Hg when compared with the experimentally determined value,[17] but successively better values of ρ_c are obtained when the Dieterici and the van der Waals equations are applied to Hg. The present investigation was carried out to determine whether an equation of the same functional form as Equation (1) could be fitted to liquid metals.

A comprehensive survey of the literature was made to obtain the available experimental density data on liquid metals; the sources of the data are referred to in Table I. In initial attempts to fit Equation (1) to liquid metals, calculated densities were compared with experimentally determined values using data for Li, Mg, In, Ag, Pb, Hg, U, and Au. These elements were selected because

TABLE I

Sources of Density Data

Element	Method of Measurement	References	Element	Method of Measurement	References	Element	Method of Measurement	References
Ag	*a,b,d*	18-26	In	*a-d*	53,90-92	Sb	*a,b,d*	22,25,31,38,
Al	*a-c*	22,27-34	K	*a-c*	63,84,86,			41,52-54,
Au	*a*	24,35,36			90-96			56,113,120,
Bi	*a-d*	25,37-50	Na	*a-d*	63,93-102			121
Ca	*b*	51	La	*b,c*	59,103	Si	*c,d*	41,122
Cd	*a-d*	6,8,27,40,	Li	*b-d*	42,65,92,96	Sn	*a,b,d*	25,31,33,34,
		45,52-57	Mg	*a,b*	22,104-109			37,40,43,45,
Ce	*b,c*	58,59	Ni	*c,d*	60,61,75			56,92,114,
Co	*c,d*	60,61	Pb	*a-d*	25,27,31,33,			115,123-127
Cs	*b,c*	62-65			40,42,43,45,	Te	*d*	41
Cu	*a-d*	18,20,28,			52-54,56,	Ti	*d*	128
		31,34,66-72			69,109-115	Tl	*d*	52,53
Fe	*a-d*	18,60,61,	Pd	*c*	20	U	*a*	129
		70,71,73-76	Pr	*d*	116	Zn	*a-d*	22,25,27,28,
Ga	*b,c*	77-79	Pt	*c*	20			31,33,40,45,
Ge	*b*	80,81	Pu	*a-c*	39,117			108,115,
Hg	*b-d*	17,45,54,	Rb	*a-c*	62,65,118,			120,124,
		82-89			119			126,130,
								131

a - Archimedean, *b* - pycnometer, *c* - volumetric, *d* - pressure

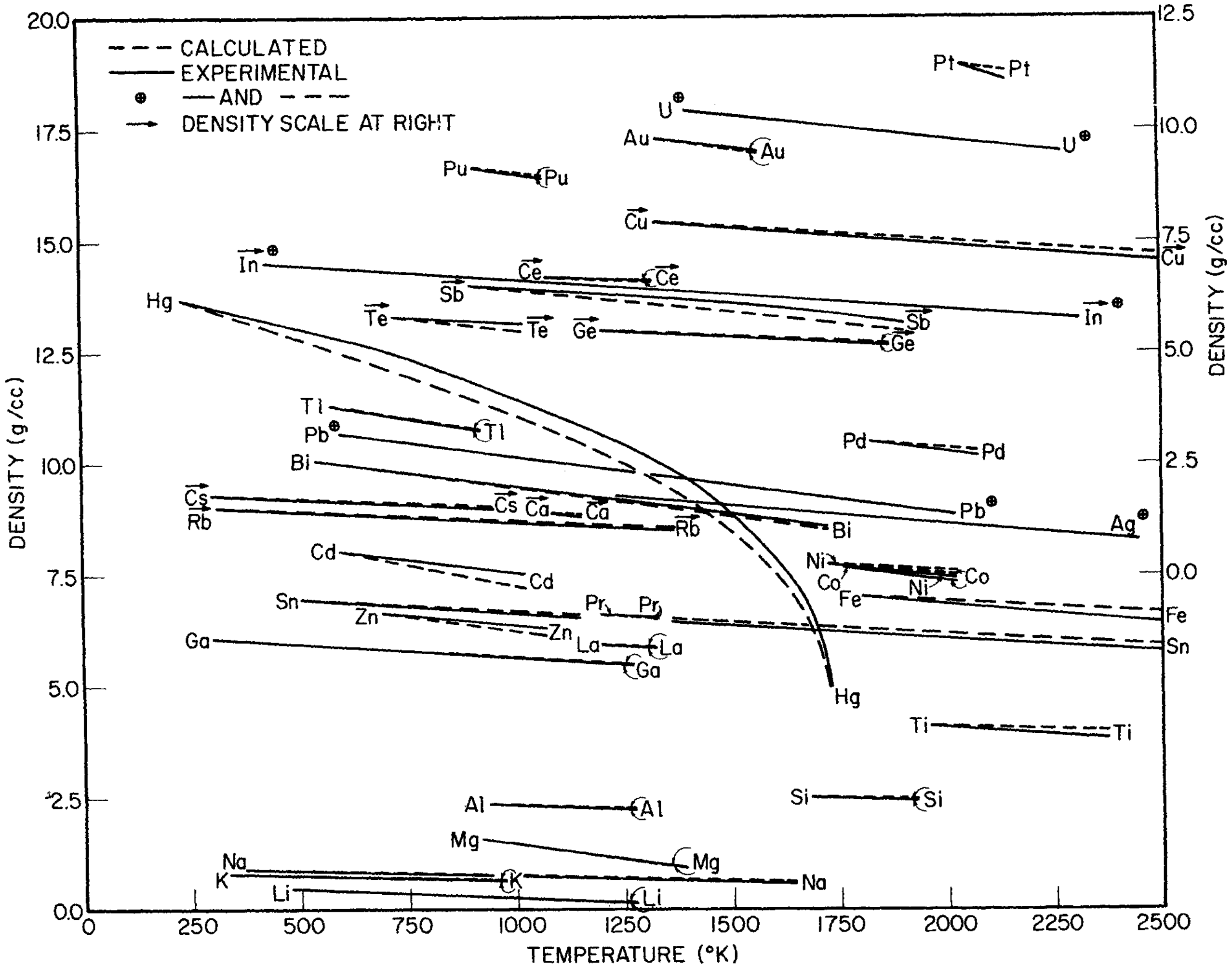

Fig. 1. Density-temperature plots for liquid metals.

their densities are known fairly accurately, and these densities represent low, intermediate, and high values in the density spectrum of liquid metals. It was found empirically in the present study that a very good fit of Equation (1) to these liquid metals was obtained by using the simple numerical values of 2, $\frac{1}{4}$, -1, and $\frac{3}{4}$ for the coefficients a, b, c, and d, respectively; these values of the coefficients were used to fit Equation (1) to the other metals listed in Table I.

It can be shown readily that, by using the values of the coefficients found in the present study, Equation (1) may be written in the form

where ρ_m is the density of the liquid metal at the Kelvin melting temperature, T_m. This equation shows that we need only one experimentally determined value, ρ_m, to estimate the density of the liquid metal as a function of temperature. The density ρ in Equation (2) is expressed as a function of ρ_m (as well as of temperature) rather than in terms of the density at some higher temperature because it is possible in many instances to estimate ρ_m from a knowledge of the densities of solid metals when density data for liquid metals are not available.

Equation (2) was tested by comparing experi-

$$\rho = \rho_m \left[\frac{1+2\left(1-\frac{T}{T_c}\right)^{\frac{1}{2}} + \frac{1}{4}\left(1-\frac{T}{T_c}\right) - \left(1-\frac{T}{T_c}\right)^2 + \frac{3}{4}\left(1-\frac{T}{T_c}\right)^3}{1+2\left(1-\frac{T_m}{T_c}\right)^{\frac{1}{2}} + \frac{1}{4}\left(1-\frac{T_m}{T_c}\right) - \left(1-\frac{T_m}{T_c}\right)^2 + \frac{3}{4}\left(1-\frac{T_m}{T_c}\right)^3} \right], \qquad (2)$$

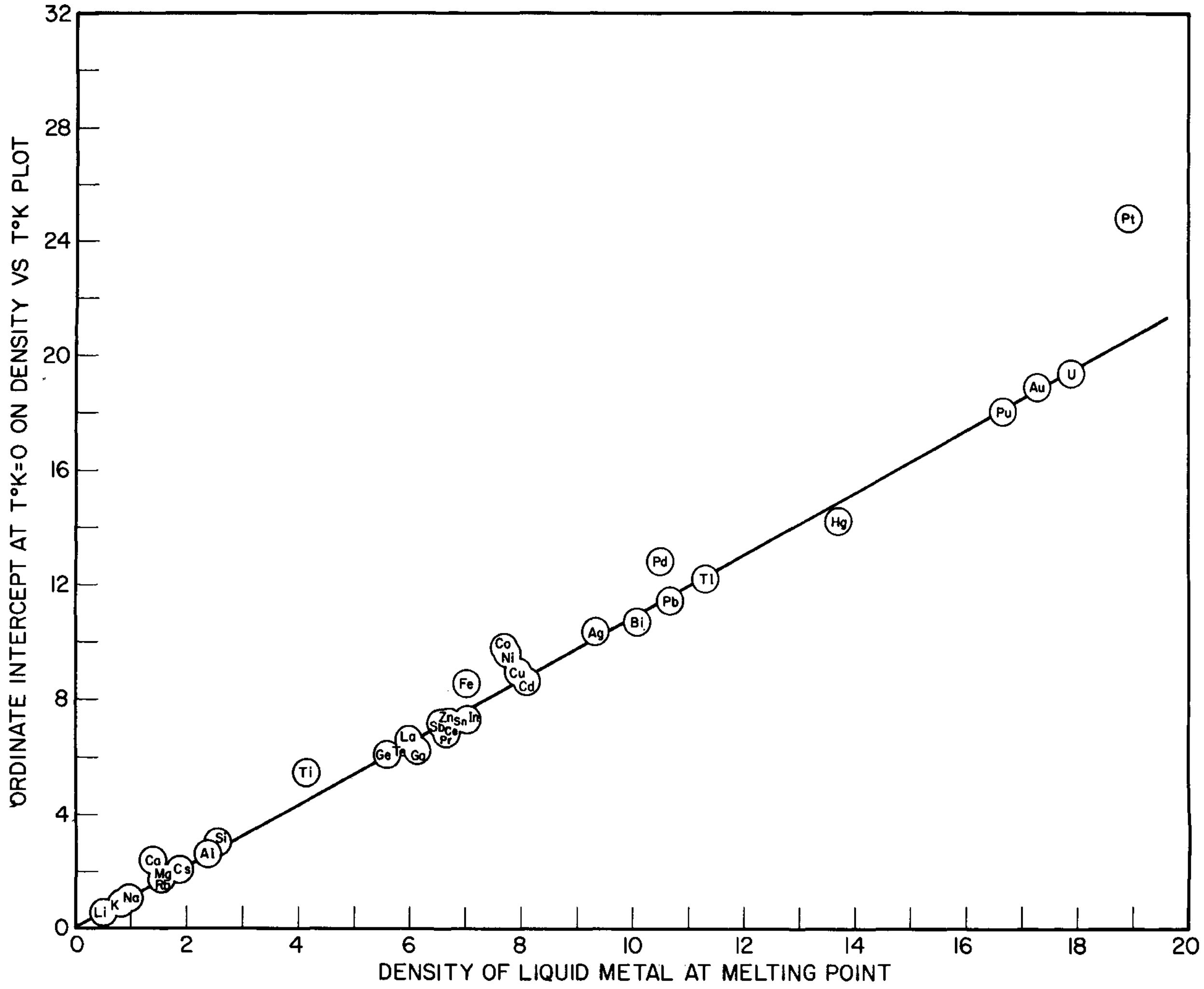

Fig. 2. Ordinate intercept-density plots for liquid metals.

mental and calculated densities of 35 metals (see Fig. 1). Mercury is the only metal whose critical temperature has been determined experimentally,[17] and values of T_c for the other metals were estimated by the method of Gates and Thodos.[132] Over the temperature ranges for which experimental data were available, the differences between experimental and calculated densities were found to be (a) less than seven percent for Ca, Cs, Cd, Fe, Hg, K, Na, Rb, Sb, Sn, and Ti, and (b) less than three percent for the remaining 24 metals.

Equation (2) requires, for application, a knowledge of the critical temperature, which, as indicated above, has been measured only for Hg.[17] A simpler representation of the ρ/T relationship may be made in the form

$$\rho = A + BT \,, \qquad (3)$$

where T is in °K and A and B are constants. Although this equation does not have the fundamental basis that Equation (1) has, and Equation (3) cannot be expected to be applicable at temperatures at which a plot of ρ vs T is no longer linear (whereas Equation (1) does not have this restriction), Equation (3) represents existing data satisfactorily for many practical purposes, and it is obviously somewhat simpler to handle than Equation (1). Equation (3) was therefore adopted to search for an empirical means of estimating unknown temperature coefficients of density for liquid metals. A plot of the coefficient A of Equation (3) vs ρ_m showed that the data fell on a straight line with reasonable scatter (see Fig. 2). A least-squares treatment of the data, omitting the values for Pt, Pd, Co, Ti, and Ca, whose points on the A vs ρ_m plot showed the largest deviations, yielded the expression

$$A = \frac{27}{25}\,\rho_m \;. \qquad (4)$$

From Equation (3) it follows that

$$\rho_m = A + BT_m \;. \qquad (5)$$

A solution of the three simultaneous linear Equations (3), (4), and (5) gives the following empirical density/temperature relation for liquid metals:

$$\rho = \rho_m \left[\frac{27}{25} - \frac{2}{25}\left(\frac{T}{T_m}\right)\right] \quad . \qquad (6)$$

The differences between experimental densities and densities calculated by use of Equation (6) were found to be (a) less than about twenty percent for Ga, In, and Sn, (b) less than eight percent for Ce, Fe, Na, Rb, and Ti, and (c) less than five percent for the remaining twenty-seven metals; in the case of Hg, Equation (6) was applied in the temperature range corresponding to the linear portion of the ρ vs T plot for this metal.

REFERENCES

[1] F. H. REE, *J. Chem. Phys.* 36, 3373-3378 (1962).

[2] E. BAUER, M. MAGAT and M. SURDIN, *Trans. Far. Soc.* 33, 81-87 (1937).

[3] S. W. BENSON, *J. Phys. and Colloid Chem.* 52, 1060-1074 (1948).

[4] J. F. DUGGAR, Jr., *J. Phys. Chem.* 62, 634 (1958).

[5] S. FLUGGE (Editor), *Encyclopedia of Physics*, Vol. 13, pp. 42-57 (1958).

[6] A. V. GROSSE, *J. Inorg. Nucl. Chem.* 22, 23-31 (1961).

[7] E. A. GUGGENHEIM, *J. Chem. Phys.* 13, 253-261 (1945).

[8] R. W. HAKALA, *Chem. and Eng. News* 37, 43-44 (1959).

[9] G. N. LEWIS and M. RANDALL, *Thermodynamics*, Revised by K. S. Pitzer and L. Brewer, pp. 616-623, McGraw-Hill, New York (1961).

[10] P. J. McGONIGAL, *J. Phys. Chem.* 66, 1686-1689 (1962).

[11] J. R. PARTINGTON, *An Advanced Treatise on Physical Chemistry*, Vol. 1, pp. 650-651, Longmans, Green, New York (1949); *ibid.* Vol. 2, pp. 5-57 (1955).

[12] R. C. REID and T. K. SHERWOOD, *The Properties of Gases - and Liquids*, pp. 2-70, McGraw-Hill, New York (1958).

[13] J. S. ROWLINSON, *Liquids and Liquid Mixtures*, Chapter 3, Butterworths, London (1959).

[14] W. T. SMITH, Jr., S. GREENBAUM and G. P. RUTLEDGE, *J. Phys. Chem.* 58, 443-447 (1954).

[15] A. F. STOTT and R. DILLON, *J. Chem. Phys.* 17, 1179-1181 (1949).

[16] F. J. WRIGHT, *J. Appl. Chem.* 11, 193-196 (1961).

[17] F. BIRCH, *Phys. Rev.* 41, 641-648 (1932).

[18] G. URBAIN and L. D. LUCAS, National Phys. Lab. Symposium No. 9, "The Physical Chemistry of Metallic Solutions and Intermetallic Compounds," Vol. 2, Section 4E. Her Majesty's Stationery Office, London (1959).

[19] A. D. KIRSHENBAUM, J. A. KAHILL and A. V. GROSSE, *J. Inorg. Nucl. Chem.* 24, 333-336 (1962).

[20] L. D. LUCAS, *Compt. rend.* 253, 2526-2528 (1961).

[21] I. LAUERMANN and G. METZGER, *Z. Physik Chem.* 216, 37-41 (1961).

[22] F. SAUERWALD, *Z. Metallk* 14, 457-461 (1922).

[23] R. HOFFMAN and W. STAHL, *Metal und Erz* 19, 48-49 (1922).

[24] E. GEBHARDT and S. DORNER, *Z. Metallk* 42, 353-358 (1951).

[25] Y. MATUYAMA, *Sci. Rep. Tohoku Imp. Univ.* 18, 733-744 (1929).

[26] A. JOUNIAUX, *Bull. Soc. Chim.* 47, 528-531 (1930).

[27] A. KNAPPWOST and H. RESTLE, *Z. Electrochem* 58, 112-118 (1954).

[28] E. PELZEL and H. SCHNEIDER, *Z. Metallk* 35, 121-123 (1943).

[29] E. GEBHARDT, M. BECKER and S. DORNER, *Z. Metallk* 44, 510-514; 573-575 (1953).

[30] YU·V. NAIDICH and V. N. YEREMENKO, *The Phys. of Met. and Metallog* 11, 62-66 (1961).

[31] P. PASCAL and A. JOUNIAUX, *Compt. rend.* 158, 414-416 (1914).

[32] J. D. EDWARDS and T. A. MOORMANN, *Chem. and Met. Eng.* 24, 61-64 (1921).

[33] C. M. SAEGER, Jr. and E. J. ASH, *J. Research NBS* 8, 37-60 (1932).

[34] K. BORNEMANN and F. SAUERWALD, *Z. Metallk* 14, 145-159 (1922).

[35] E. GEBHARDT and G. WORWAG, *Z. Metallk* 43, 106-108 (1952).

[36] W. KRAUSE and F. SAUERWALD, *Z. Anorg. Chem.* 181, 347-352 (1929).

[37] E. HERCZYNSKA, *Naturw* 47, 200-201 (1960).

[38] E. PELZEL, *Z. Metallk* 50, 392-395 (1959).

[39] C. Z. SERPAN, Jr. and L. J. WITTENBERG, *Trans. AIMME* 221, 1017-1020 (1961).

[40] V. K. SEMENCHENKO and N. L. POKROVSKI, *Uspekhi Khimii* Vol. 6, Book 6, pp. 777-820 (1937); U. S. Naval Research Laboratory NRL Translation No. 190 (1948).

[41] L. D. LUCAS and G. URBAIN, *Compt. rend.* 255, 2414-2416 (1962).

[42] S. A. BEEN, H. S. EDWARDS, E. E. TEETER, Jr., and V. P. CALKINS, NEPA-1585 (1950).

[43] M. PLUSS, *Z. Anorg. Chem.* 93, 1-44 (1915).

[44] G. METZGER, *Z. Physik-Chem.* 211, 1-25 (1959).

[45] T. R. HOGNESS, *J. Am. Chem. Soc.* 43, 1621-1628 (1921).

[46] K. BORNEMANN and P. SIEBE, *Z. Metallk* 14, 329-334 (1922).

[47] E. GEBHARDT and K. KOSTLIN, *Z. Metallk* 48, 601-602 (1957).

[48] A. JOUNIAUX, *Bull. Soc. Chim.* 31, 677-681 (1932).

[49] A. D. KIRSHENBAUM, "Final Report on High Temperature Inorganic Chemistry to the National Science Foundation," Research Grant NSF-G15540 (October 1961).

[50] S. W. STRAUSS, L. E. RICHARDS and B. F. BROWN, *J. Nucl. Mat.* 5, 12-16 (1962).

[51] M. F. CULPIN, *Proc. Phys. Soc.* 70, 1079-1086 (1957).

[52] A. SCHNEIDER, A. STAUFFER and G. HEYMER, *Naturw* 41, 326-327 (1954).

[53] A. SCHNEIDER and G. HEYMER, *Z. Anorg. Chem.* 286, 118-135 (1956).

[54] H. T. GREENAWAY, *J. Inst. Met.* 74, 33-148 (1948).

[55] R. ARPI, *International Ztsch Metallog* 5, 142-168 (1914).

[56] H. J. FISHER and A. PHILLIPS, *J. Met.* 6, 1058-1070 (1954).

[57] R. SAUERWALD, *Z. Anorg. Chem.* 153, 319-322 (1926).

[58] J. F. EICHELBERGER, Mound Laboratory Report MLM-1113 (1961).

[59] G. R. PULLIAM and E. S. FITZSIMMONS, Ames Laboratory Report ISC-659 (1955).

[60] P. KOZAKEVITCH and G. URBAIN, *J. Iron Steel Inst.* 186, 167-173 (1957).

[61] L. D. LUCAS, *Compt. rend.* 250, 1850-1852 (1960).

[62] D. L. COCHRAN in "Proceedings of 1962 High-Temperature Liquid-Metal Heat-Transfer Technology Meeting" BNL 756 (C-35), pp. 77-94 (1962).

[63] L. HACKSPILL, *Compt. rend.* 152, 259-262 (1911).

[64] M. ECKARDT and E. GRAEFE, *Z. Anorg. Chem.* 23, 378-382 (1900).

[65] E. N. DAC. ANDRADE and E. R. DOBBS, *Proc. Roy. Soc.* 211A, 12-30 (1952).

[66] J. A. CAHILL and A. D. KIRSHENBAUM, *J. Phys. Chem.* 66, 1080-1082 (1962).

[67] J. I. LANG, "Measurement of the Heat Capacity and Density of Liquid Copper," WADC Technical Report 57-488 (1957).

[68] E. PELZEL, *Metall* 11, 954-959 (1957).

[69] T. MALMBURG, *J. Inst. Met.* 89, 137-139 (1961-61).

[70] P. KOZAKEVITCH, S. CHATEL, G. URBAIN and M. SAGE, *Rev. Metall* 52, 139 (1955).

[71] E. WIDAWSKI and F. SAUERWALD, *Z. Anorg. Chem.* 192, 145-160 (1930).

[72] G. BECKER, F. HARDERS and K. KORNFELD, *Arch. Eisenhuttenw* 20, 363-367 (1949).

[73] V. H. STOTT and J. H. RENDALL, *J. Iron Steel Inst.* 175, 374-378 (1953).

[74] A. D. KIRSHENBAUM and J. A. CAHILL, *Trans. AIMME* 224, 816-819 (1962).

[75] C. BENEDICKS, N. ERICSSON and G. ERICSON, *Arch. Eisenhuttenw* 3, 473-486 (1930).

[76] C. H. DESCH and B. S. SMITH, *J. Iron Steel Inst.* 1, 358-363 (1929).

[77] G. L. MACK, J. K. DAVIS and F. E. BARTELL, *J. Phys. Chem.* 45, 846-851 (1941).

[78] T. W. RICHARDS and S. BOYER, *J. Amer. Chem. Soc.* 43, 274-294 (1921).

[79] W. H. HOATHER, *Proc. Phys. Soc.* 48, 699-707 (1936).

[80] W. KLEMM, H. SPITZER, W. LINGERBERG and H. J. JUNKER, *Monatshefte für Chemie* 82, 629-637 (1952).

[81] S. R. SANGSTER and J. N. CARMEN, Jr., *Phys. Rev.* 23, 206-207 (1955).

[82] A. SMITS and G. J. MULLER, *Z. Physik-Chem.* 36B, 288-290 (1937).

[83] R. SUHRMANN and E. O. WINTER, *Z. Naturforsch* 10A, 985-996 (1955).

[84] A. ROEDER and W. MORAWIETZ, *Z. Metallk* 47, 734-741 (1956).

[85] A. H. COOK and N. W. B. STONE, *Phil. Trans. Roy. Soc.* Series A 250, 279-323 (1957); *ibid.* 254A, 126-154 (1961).

[86] J. DEGENKOLBE and F. SAUERWALD, *Z. Anorg. Chem.* 270, 325-327 (1952).

[87] J. BENDER, *Physik Zeitschr* 16, 246-247 (1915); *ibid.* 19, 410-414 (1918).

[88] LANDOLT-BORNSTEIN, "Physikalisch-Chemische Tabellen" Band IV, Teil 1, p. 128 (1955).

[89] *Gmelins Handbuch der Anorganischen Chemie* 34, 287 (1960).

[90] D. D. WILLIAMS and R. R. MILLER, *J. Am. Chem. Soc.* 72, 3821 (1950).

[91] P. J. McGONIGAL, J. A. CAHILL and A. D. KIRSHENBAUM, *J. Inorg. Nucl. Chem.* 24, 1012-1013 (1962).

[92] C. GAMERTSFELDER, *J. Chem. Phys.* 9, 450-457 (1941).

[93] C. T. EWING, J. A. GRAND and R. R. MILLER, *J. Am. Chem. Soc.* 73, 1168-1171 (1951); *J. Phys. Chem.* 58, 1086-1088 (1954).

[94] E. B. HAGEN, *Ann. Physik und Chemie* 19, 436-474 (1883).

[95] E. RINCK, *Compt. rend.* 189, 39-41 (1929); *Ann. de Chimie* 18, 434-445 (1932).

[96] A. BERNINI and C. CANTONI, *Nuovo Cimento* 8, 241-260 (1914).

[97] J. P. STONE, C. T. EWING, J. R. SPANN, E. W. STEINKULLER, T. A. KOVACINA and R. R. MILLER, U. S. Naval Research Laboratory, NRL Memorandum Report 1312 (1962).

[98] C. T. EWING, H. B. ATKINSON, Jr. and T. K. RICE, U. S. Naval Research Laboratory Report No. C-3287 (1948).

[99] National Distillers Chemical Corporation, "Sodium," National Distillers Chemical Corporation, New York (1948).

[100] M. SITTIG, *Sodium, Its Manufacture Properties and Uses*, pp. 366-370, Reinhold, New York (1956).

[101] J. W. TAYLOR, *J. Inst. Met.* 83, 143-152 (1954-55).

[102] M. NISHEBAYASHI, WADC Technical Report 43-308 Part I (1953).

[103] J. F. EICHELBERGER, Mound Laboratory Report MLM-1116 (1961).

[104] J. P. McGONIGAL, A. D. KIRSHENBAUM and A. V. GROSSE, *J. Phys. Chem.* 66, 737-740 (1962).

[105] J. D. EDWARDS and C. S. TAYLOR, *Trans. AIMME* 69, 1070-1075 (1923).

[106] K. ARNDT and G. PLOETZ, *Z. Physik Chem.* 130, 184-186 (1927).

[107] H. GROTHE and C. MANGELSDORF, *Z. Metallk* 29, 352-353 (1937).

[108] E. PELZEL and F. SAUERWALD, *Z. Metallk* 33, 229-232 (1941).

[109] E. GEBHARDT, M. BECKER and E. TRAGNER, *Z. Metallk* 46, 90-94 (1955).

[110] S. W. STRAUSS, L. E. RICHARDS and B. F. BROWN, *Nucl. Sci. Eng.* 7, 442-447 (1960).

[111] O. KUBASCHEWSKI and R. HORNLE, *Z. Metallk* 42, 129-132 (1951).

[112] A. D. KIRSHENBAUM, J. A. CAHILL and A. V. GROSSE, *J. Inorg. Nucl. Chem.* 22, 33-38 (1961).

[113] E. GEBHARDT and K. KOSTLIN, *Z. Metallk* 48, 636-641 (1957).

[114] A. L. DAY, R. B. SOSMAN and J. C. HOSTETTER, *Amer. J. Sci.* 187, 1-39 (1914).

[115] T. C. TOYE and E. R. JONES, *Proc. Phys. Soc.* 71, 88-99 (1958).

[116] J. F. EICHELBERGER, Mound Laboratory Report MLM-1118 (1961).

[117] C. E. OLSEN, T. A. SANDENAW and C. C. HERRICK, Los Alamos Scientific Laboratory Report LA-2358 (1959).

[118] S. COHEN, *Nucl. Sci. Eng.* 2, 530-531 (1957).

[119] F. TEPPER, A. MURCHISON and J. S. ZELENAK, MSA Research Corporation Report MSA 62-122 (1962); ASD-TDR-63-133 (1963).

[120] K. BORNEMANN and F. SAUERWALD, *Z. Metallk* 14, 254-258 (1922).

[121] A. D. KIRSHENBAUM and J. S. CAHILL, *ASM Trans. Quarterly* 55, 849-852 (1962).

[122] R. A. LOGAN and W. L. BOND, *J. Appl. Phys.* 30, 322 (1959).

[123] N. L. POKROWSKI and M. SAIDOV, *Fizika Metallov i Metaldovedenie*, SSSR, Vol. 2, No. 3, pp. 546-551 (1956); U. S. Naval Research Laboratory Translation 614 (1957).

[124] E. UBELACKER and L. D. LUCAS, *Compt. rend.* 254, 1622-1624 (1962).

[125] A. D. KIRSHENBAUM and J. A. CAHILL, *ASM Trans.-Quarterly* 55, 844-848 (1962).

[126] E. PELZEL, *Berg-und Huttenmannische Monatshefte* 93, 248-254 (1948).

[127] F. A. KANDA and D. V. KELLER, "Investigation of Specific Volumes of Liquid Metals and Alloys," Syracuse University Report No. 1 (May 1962).

[128] M. A. MAURAKH, *Trans. Indian Inst. Met.* 14, 209-225 (1961).

[129] A. V. GROSSE, J. A. CAHILL and A. D. KIRSHENBAUM, *J. Am. Chem. Soc.* 83, 4665-4666 (1961).

[130] H. R. THRESH, D. W. G. WHITE, J. O. EDWARDS and J. W. MEIER, Dept. Mines and Tech. Surveys, Mines Branch, Phys. Met. Div., Ottawa, Canada, Report PM-R-61-19, Progress Report No. 1 (July 1961).

[131] E. GEBHARDT, M. BECKER and S. DORNER, *Z. Metallk* 45, 83-35 (1954).

[132] D. S. GATES and G. THODOS, *A. T. Ch. E. Journal* 6, 50-54 (1960).

*S. W. Strauss**

Physical Metallurgy Branch,
Metallurgy Division
U. S. Naval Research Laboratory

Received June 19, 1963
Revised September 25, 1963

*Present Address
Headquarters, Research & Technology Division
Air Force Systems Command
RTD (RTHM) Physics Office
Balling Air Force Base
Washington, D.C. 20332

Considerations on the Mechanism of Inert-Gas Diffusion in UO$_2$

A large amount of work has been done in a number of laboratories in the U. S. A., in Canada and in Europe on the subject of inert-gas diffusion in solid nuclear fuels, and specially of xenon in UO$_2$,[1] this problem being of considerable importance in reactor design.

Information on inert-gas diffusion in some solids has been gained beforehand from the use of Hahn's emanation technique.[2] Moreover, experiments have been performed on the behavior of inert gases in metals,[3,4] and ionic crystals.[5,6] On the other side, a lot is known about some ion diffusion in UO$_2$.[7,8,9]

It turns out, therefore, that while experimental results of xenon diffusion-coefficient measurements in UO$_2$ are so widely scattered that, as Zimen pointed out recently,[6] one may feel justified in publishing almost any figures, some considerations on the migration mechanism are nevertheless possible. The scope of this short paper is merely to review in a critical way and, to a certain extent, to clarify and integrate current ideas on this subject, while suggesting at least one experiment that could cast a little more light on its intricacy.

The idea that inert gases can migrate in solids as charged ions instead of neutral atoms has been set forth by Tobin[4] for the following reasons:

1. When the gas is introduced into a metal through a glow discharge, the kinetic energy of the impinging atoms is of the order of 10 eV, corresponding to their ionization energy, whereas to displace an atom from the interior of the metal about 25 eV would be required:
2. a neutral inert-gas atom, being very large, would be a very serious misfit within a regular metal lattice;
3. the allegedly high strain energy associated with a neutral inert-gas atom within a metal lattice would result in its combination with the first few vacancies, which diffuse toward it;
4. the activation energies and, to a much lesser extent, the temperature-independent diffusion coefficients D_0 of Xe and Kr in Ag fit very well into the frame of a modified Lazarus theory if they are supposed to exist within the metal lattice as quasi-univalent positive ions.

That a neutral inert-gas atom would be such a misfit in any lattice may perhaps be questioned, since inert gases are highly compressible even in the solid state, as should be expected from the lack of strong Van der Waals interactions between atoms.

Stewart[10] has measured and Zucker[11] has calculated the compressibility of solid neon, argon and krypton. One sees that, even at very low temperature, the behavior is such that the compressibilities approach asymptotic temperature-independent values with increasing pressure. For argon the value is about 18×10^{-12} cm^2/dyn, and for xenon, extrapolating from the other inert gases, about 14×10^{-12} cm^2/dyn.

For UO$_2$, the Young modulus E should be, roughly extrapolating from Lang's data,[12] about (1.90 -

---

[1] For a comprehensive review and additional references, see: D. F. TONER and J. L. SCOTT, *Nucl. Safety*, 3, 15, (1961).

[2] See, for instance, F. FELIX and P. SCHMELING, *Euratom Report* EUR-111.e (1962).

[3] A. D. LeCLAIRE and A. H. ROWE, *Rev. Met.*, 52, 94 (1955).

[4] J. M. TOBIN, *Acta Met.*, 7, 701 (1959); 8, 781 (1960).

[5] T. LAGERWALL, *Nukleonik*, 4, 158 (1962).

[6] Oral communications of the Hahn-Meiter Institut Group at the Euratom Meeting on UO$_2$, Brussels (1962).

[7] A. B. AUSKERN and J. BELLE, *J. Nucl. Mat.*, 3, 267 (1961).

[8] J. BELLE and A. B. AUSKERN, *J. Nucl. Mat.*, 3, 311 (1961).

[9] H. M. FERRARI, Westinghouse Report WCAPD-2098 (1962).

[10] J. W. STEWART, *Phys. Rev.*, 97, 578 (1955).

[11] J. J. ZUCKER, *J. Chem. Phys.*, 25, 915 (1956); *Phil. Mag.*, 33, 937 (1958).

[12] See: J. BELLE and P. LUSTMAN, Westinghouse Report WAPD-184 (1957).