CONF-770843-3

Lawrence Livermore Laboratory

THERMAL EXPANSION OF METALS OVER THE ENTIRE LIQUID RANGE

J. W. Shaner

August 25, 1977

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

This paper was prepared for 1977 International Symposium on Thermal Expansion of Solids, Manitoba, Canada, August 29-31, 1977

Winnipeg

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

THERMAL EXPANSION OF METALS OVER THE ENTIRE LIQUID RANGE*

J. W. Shaner

University of California Lawrence Livermore Laboratory
Livermore, California 94550

ABSTRACT

This paper reviews the current state of the art for measuring liquid metal densities. Conventional high precision techniques for use below 2000K as well as new techniques for more extreme temperatures are addressed. Pertinent data, which have appeared since the last critical reviews, for elemental metals are discussed.

I. INTRODUCTION

Thermal expansion data requires the measurement of two quantities: a length or density, and a temperature. For solids, high precision measurements of both of these quantities can be made, at least up to modest temperatures ($T \sim 1000K$). The precision available from optical interferometry or capacitance dilatometers, and thermocouples or resistance thermometers can result in $\pm 0.1\%$ accuracies in thermal expansion coefficient around 300K.^{1,2} In fact, the precision may be so high that the effects of even 0.1% Fe impurity on the thermal expansion of ZnS have been detected.³

As the temperature is increased, both length or density, and the temperature become more difficult to measure with precision. Very careful measurements of the linear expansion coefficient of Mo and W wires up to 2800K and 3500K respectively have been made with reported accuracies of $\pm 1\%$.⁴ More recently, an optical interferometer has been used with subsecond pulse heating of metals up to their melting points.⁵ This technique has promise of also giving $\pm 1\%$ measurements of linear expansion coefficients up to

2000-3000K. Modulation techniques⁶ can be exploited to give direct measurement of expansion coefficients, thereby eliminating the errors inherent in differentiating density or length vs temperature data. One might hope that these techniques will become accurate enough to measure non-equilibrium effects associated with vacancy concentration near the melting point.

The theoretical situation for thermal expansion of solids is also relatively well developed. This is particularly true for ionic crystals like alkali halides or molecular crystals where interparticle potentials are known well enough to make realistic calculations of anharmonic effects.⁷ For simple metals, at near normal density, the pseudopotential approach can be used to define effective interparticle potentials, which can then be used to evaluate the thermodynamic functions.⁸

Thermal expansion measurements on liquid metals present several problems not normally encountered in measurements on solids. Firstly, the temperatures are almost always high. Secondly, the samples are not self-supporting. Therefore either a container is necessary, or the heating must be done rapidly while inertially confining the liquid. Thirdly, if a container other than an a.c. magnetic field is used, the extreme corrosiveness of most liquid metals must be considered.

On the other hand, the problem of non-equilibrium defect concentrations may not apply to liquid metals. Therefore, dynamic measurements, which may avoid the container problems, are not so suspect as they are in the solid phase near melting.

Theoretical calculations of the thermal expansion of liquids can be accomplished through differentiation of the equation of state. A standard result of statistical mechanics is that, assuming pairwise additive spherically symmetric potentials, the equation of state can be written as⁹

$$P = \frac{N}{V} kT - \frac{2\pi N^2}{3V} \int_0^\infty \frac{d\phi(r)}{dr} g(r) r^3 dr \quad (1)$$

In this equation, $\phi(r)$ is the interparticle potential and $g(r)$ is the radial distribution function. For simple liquids, like argon, the interparticle potential is known well enough to calculate a reliable equation of state with even a hard sphere distribution function. However, for metals the pseudopotential, $\phi(r)$, is density dependent, so calculations become much less reliable. Pseudopotential calculations of the thermal expansion coefficient of even simple liquid metals at the melting point have come only within 30-40% of experimental values.¹⁰

In view of the uncertainties inherent in a direct calculation of liquid metal equations of state, accurate measurement is desirable for many applications. For example, the volume change upon melting is an important parameter for evaluating residual stresses in weld joints. Also, the equation of state is necessary for designing for accidents in nuclear reactors. For this latter application, equation of state for some materials must be known up to ~ 1 GPa in pressure and $\sim 10,000$ K in order to properly model some advanced reactors.¹¹

The thermal expansion of liquid metals has been the subject for several very extensive reviews covering the period up to 1973.¹²⁻¹⁷ Since the conventional techniques for measuring liquid metal densities up to 2000K are described in detail in these reviews, these techniques will be only briefly discussed in this paper. New data appearing after 1973 will be described along with new techniques capable of significantly increasing accuracy or temperature range.

II. TECHNIQUES REQUIRING A CONTAINER

Perhaps the most familiar apparatus for measuring thermal expansion of a liquid metal is the mercury thermometer. This device is the prototype of all dilatometers, where the liquid metal density is measured directly as a change in surface level with temperature. At temperatures below 1700K, the containing vessel may be quartz for many metals, and the liquid surface may be viewed directly. Under these circumstances the surface level may be corrected for the meniscus shape. For work at higher temperatures a refractory container must be used—tantalum or graphite, for example. Then typically the liquid level is monitored by a contacting electrode. The difficulties in this technique center around calibrating the relation between surface level and sample volume and accurately measuring surface level in spite of optical aberrations, surface curvature, and dilatometer expansion. Since the whole apparatus can be set in a furnace, the temperatures can be accurate to better than 1 K. However, to make density measurements better than $\pm 0.5\%$ at temperatures greater than 1300K is practically impossible.¹⁶

A potentially very accurate capacitance technique for monitoring the liquid level in a zirconia tube has been used by Evan and Magen to obtain density of liquid mercury up to 1800K.¹⁸ The tube has been metallized on the outside to form one electrode of the capacitor, while mercury metal inside forms the other electrode. For mercury, this tube was maintained at room temperature, so no corrections for temperature dependent surface tension (i.e. meniscus shape) need to be made. The dilatometer was connected to the heated mercury sample volume by a capillary of negligible volume. For

other metals, the dilatometer would have to be maintained above the sample melting temperature. The capability for remote sensing with no movable parts permit this apparatus to be used in a pressure vessel (up to 0.2 GPa in the work of Evan and Magen). Evan and Jortner, using this apparatus quote an absolute density uncertainty of 1%.¹⁹

Another of the classical techniques for liquid metal density measurement is the Archimedeian method. In this experiment a sinker of known mass made of some refractory material which will not be attacked by the liquid sample is weighed as it is immersed in the liquid metal. An indirect technique, where the sample is in the sinker, which is immersed in an inert liquid of known density has also been applied. In both of these techniques one major source of error is the force acting on the supporting wire as it passes through the melt surface. Both the liquid surface tension and the contact angle must be calibrated. In addition, the thermal expansion of the sinker must be known accurately. Grosse et al. have used the direct Archimedeian method on liquid uranium up to 1900K with a quoted accuracy of $\pm 0.08\%$.²⁰ However, subsequent measurements have given a liquid density smaller by 3%.²¹ More recent measurements on several metals by Lucas^{13,14} have been more carefully calibrated for the surface tension effect. These measurements quote uncertainties up to $\pm 0.5\%$ at 2000K, which are probably realistic estimates of the accuracy of this technique.

Pycnometry, another classic method, involves the filling of a known volume with liquid metal at a known temperature, and subsequently measuring the mass of the metal after it has cooled. This technique, thoroughly reviewed by Crawley¹⁶, has a potential accuracy of better than $\pm 0.05\%$ in density below 750K. This precision cannot, however, be obtained without extreme care to avoid bubbles or voids when liquid metal fills the cavity.

The last of the classic methods for measuring liquid metal density is the maximum bubble pressure technique. In this method, the pressure necessary to force inert gas through a capillary to form bubbles beneath the surface of the melt is measured. The change in pressure as the depth of the bubbling capillary tip is varied measures the hydrostatic head. Corrections which must be made include the thermal expansion of the capillary and the change in melt level as the capillary is immersed. A detailed analysis of the uncertainties in this technique has been given by Lucas¹², with the result that, at least at modest temperatures, the density can be measured to $\pm 0.2\%$. For higher melting temperature metals an accuracy of $\pm 0.5 - 1\%$ is more appropriate.²²

For all of the methods described in this section attention must be given to the possibility of corrosion of the container and

contamination of the sample. If a correction involving surface tension must be made, then contamination and oxidation of the surface become particularly important. Temperatures can be measured accurately by thermocouples up to 2000K, but care must be taken to prevent contamination of the junction. Optical pyrometry can be used if ± 5 -10 K uncertainties can be tolerated, but again care must be exercised if the metal sample has a high vapor pressure. With all of these cautions in mind, it seems possible to obtain densities accurate to $\pm 0.5\%$ at temperatures up to 2000K by these classic techniques.

III. DROP TECHNIQUES

A drop of a liquid metal can be made to bead on a hot substrate (typically of graphite), it can be allowed to hang or drop from a rod of the same material, or from a tube, and a drop can be levitated by an a.c. magnetic field. Conventionally the volumes of the drops are measured photographically in either visible light or with x-rays. For static drops the shapes are determined by both surface tension and density, so both parameters can be measured. On the other hand, the accuracy of the density is less because the drop is not spherical. If the drop is photographed during free fall, the shape should be spherical unless a large amplitude hydrodynamic oscillation has been excited. However, the photography is more complicated. The uncertainties inherent in drop techniques have been summarized by Saito et al.²² They found the total uncertainty to be $\pm 2\%$ in density up to 2000K, with the measurement of the drop image the largest error. The temperatures in these experiments must be obtained pyrometrically, and this contributes extra uncertainty to the measurements.

Recently double pulse holography has been applied to obtain directly the thermal expansion coefficient of a levitated aluminum drop.²³ The holography allows dimensional changes of less than $1 \mu\text{m}$ to be determined, but the drop size must still be measured photographically. The volume expansion coefficient obtained in this work, $1.95 \times 10^{-4} \text{ K}^{-1}$ at 1100K agrees reasonably with a value of 1.65×10^{-4} chosen by Crawley as the most reliable measurement.¹⁶ The temperature of the levitated sample is difficult to control, and it must be changed rapidly in order to not lose material by evaporation. Also the position of the drop must be kept stable to an optical wavelength. As a result, the double pulse holography may remain just a curiosity in the field of liquid metal density measurements.

Although the $\pm 2\%$ density measurements made with drop techniques are not the most accurate, drop methods have allowed measurements at higher temperatures. In particular, Russian researchers have

exploited drop techniques to obtain liquid densities at the melting point of refractory 4d and 5d transition metals.²⁴⁻²⁸ The reason for the high temperature capability is that a container is unnecessary.

IV. RADIATION ATTENUATION METHOD

The attenuation of gamma rays upon passing through a liquid metal specimen of known thickness has been applied with success to the measurement of density. The highest accuracies are obtained by counting many transmitted gamma rays, but the incident beam cannot be so intense that it disturbs the temperature distribution. As a result, except for very crude measurements this technique will continue to be used with static samples in a container. This restriction will limit the upper temperature range to 2000-3000K. The technique is described in more detail by W. Drotning at this conference.

V. DIRECT TECHNIQUE

A recently reported experimental technique, the isobaric expansion experiment (IEX)²⁹, has been used by us to obtain liquid metal densities over a very extensive temperature range.³⁰⁻³²

In essence, our experimental procedure consists of resistively heating a 1 mm diameter 25 mm long metal wire sample in an inert gas filled pressure vessel. The inert gas provides an isobaric environment, since the gas volume is much larger than the sample volume, and the sample expansion is slow enough to allow acoustic reverberations in the cell. The inert gas also prevents chemical contamination of the hot wire surface, an important feature for our optical pyrometry. In addition, the high pressure raises the boiling temperature, allowing us to investigate most of the liquid metal density range.

Heating of the sample is accomplished by discharging a critically damped capacitor bank through the sample quickly enough to overcome the radiative heat losses and hydrodynamic instabilities ($t < 100 \mu s$), but slowly enough so that the skin effect doesn't cause serious energy inhomogeneities ($t > 10 \mu s$). Typically, a roughly square current pulse of 30 kA with 5 μs rise and fall times is used. With these current pulse parameters the energy density is homogeneous to within 1% by the time the sample is heated to 2000K, where the pyrometry starts responding.

The pressure vessel is designed with four sapphire windows viewing the sample in a radial direction. Back lit shadowgraphs

are taken both with a Q-switched ruby laser and with a cw laser and streaking camera. Since the metal is constrained at the ends by clamping jaws, its expansion in the liquid phase is only radial. Therefore the square of the diameter, measured as a function of time by the streaking camera, is proportional to the sample specific volume. The volume measurement can be made to $\pm 2\%$. The ruby laser snapshot is taken typically 5-10 μ s after the end of the current pulse. If the final state of the sample is not uniform along the axis, the shot is discounted.

The current is measured with a wide-band current transformer, and the voltage drop across the middle 7-10 mm of the wire is measured in a separate high impedance circuit. The oscilloscope traces of both of these signals are digitized, and, after a slight inductive correction is made, the product of current and voltage is integrated to give an enthalpy vs time curve. The enthalpy so obtained is accurate to $\pm 3\%$. This is correlated with volume vs time to give an equation of state.

In order to make pyrometric records, a patch of the sample, approximately 1 x 3 mm, is viewed through a randomly trifurcated fiber optic bundle. The three output bundles deliver the thermal radiation through 50 mm band pass filters (typically at 450, 700, and 900 nm) to silicon photodiodes. The edges of the sample are carefully avoided in the image viewed by the pyrometer, since deviations from Lambert's law have been observed for thermal radiation coming off at grazing angles. The photodiode signals are amplified logarithmically and displayed on oscilloscopes. The log amplifiers reduce the precision of our light measurements, but they allow a whole shot covering the 2000 to 8000K temperature range to be displayed on a single trace. The generally unknown wavelength and temperature dependences of liquid metal emissivities make the calculation of temperatures from the pyrometer readings something of an art. The details of the calculation of temperatures from the pyrometric records is given in reference 31. The temperatures are estimated to be accurate to ± 200 K at 5000K.

The IEX technique, like all the others, has both good and bad features. The density measurements are only accurate to $\pm 2\%$ because of limitations of the measurement technique and difficulties in maintaining a smooth surface at extreme temperatures in a dynamic experiment. Also, the temperature measurement is inaccurate because radiation pyrometry must be used in a range where emissivities are generally unknown. On the other hand, densities of metals can be measured at much higher temperatures than was previously possible. For some simulation applications, the enthalpy is a more convenient variable than temperature, and enthalpy is measured reliably in IEX. Finally, even the most corrosive liquid metals may be studied.

VI. NEW DATA

In the four years since the last critical reviews^{16,17}, there have appeared several new determinations of liquid metal densities in temperature ranges already explored, as well as new measurements at higher temperatures.

Bismuth and Zinc have been remeasured in the temperature ranges 544-900 and 693-900K respectively by the Archimedean method.³³ In both cases, the data can be represented as $\rho = a + b(T-T_m)$, and the liquid densities at the melting point, a , differ by less than 0.3% from the values preferred by Crawley.¹⁶ The values of b differ from Crawley's values by 3% for Zn and 1% for Bi. Dilatometric measurements of the densities of liquid Cd, In, Sn, and Sb, and their binary alloys have been made by Nakajima³⁴ with agreement with Crawley's preferred values of a to $\pm 0.2\%$ in all cases. Larger discrepancies are found in Nakajima's b parameters--as large as 10%. This may be due to the very limited temperature range over which his data has been taken (200-300K).

Gold³⁵, tin³⁵, and silver³⁶ have been measured by sessile drop techniques with results agreeing with Crawley to better than 1% for a . The values of b differ from Crawley's by as much as 70%.

The differences between these new measurements and previous determinations probably represent the accuracy one can expect in the density measurements--i.e. ± 0.1 - 0.3% below 1000K and 1% below 2000K. If density change is measured over a 1000K temperature range, $\pm 0.1\%$ uncertainties in a result in $\pm 10\%$ in b . For smaller temperature intervals, the uncertainties in b are even greater.

Copper and iron have been subject to many density measurements in the last few years. These results are summarized in Tables I and II. Discrepancies of 1% in density and 10-20% in b still appear for these common metals at temperatures above 1500K.

Recent isobaric expansion data for liquid lead are presented in Figure 1. To within experimental error, a straight line fit to the data of the form $\rho = 10.6 - 11.6 \times 10^{-4} (T-600)$ gm/cm³ fits the data over a twofold density change. Agreement with other measurements below 2000K is satisfactory. The thermal expansion coefficient obtained here is slightly smaller than those recommended by Crawley¹⁶ and Steinberg.¹⁷ This may be due to the 0.2 GPa ambient pressure used in the IEX experiments.

Similar data is shown for uranium at 0.2 GPa in Figure 2. In this case the fit to the IEX data is $\rho = 17.6 - 17 \times 10^{-4} (T-1406)$ gm/cc. Since the temperature range of these experiments is very large, the uncertainty in the b coefficient is the same magnitude

TABLE I
LIQUID COPPER DENSITY

$\alpha(\text{gm/cc})$	$-\beta \times 10^4$	Temp. Range(K)	Technique	Ref.
7.936	7.862	1356-1512	Archimedean	37
8.019	7.3	1356-1873	γ -Attenuation	38
7.940	7.367	1376-1556	Pycnometry	39

as the uncertainty in the α coefficient-- $\pm 2\%$.

A number of determinations of high melting point 4d and 5d liquid metal densities have recently appeared. These have been obtained by both drop techniques at or near the melting point²⁴⁻²⁸, and IEX over an extensive temperature range.³⁰⁻³¹ These data are summarized in Table III.

VII. SUMMARY

Density data for liquid metals has been obtained at precisions of better than $\pm 0.2\%$ below 1000K, $\pm 1\%$ below 2000K and $\pm 2\%$ over the entire liquid range. The IEX technique has provided a useful tool for extending the data base to extreme temperatures. Although the density measurements by this latter technique are less accurate than those obtained by conventional methods, the very large temperature range permits more accurate thermal expansion coefficients to be measured.

TABLE II
LIQUID IRON DENSITY

$\alpha(\text{gm/cc})$	$-\beta \times 10^4$	Temp. Range(K)	Technique	Ref.
7.078	6.5	1812-1993	γ -Attenuation	38
7.2	10.4	1833-2023	Dilatometry	40
7.05	6.1	1810-1923	γ -Attenuation	41

TABLE III

Densities of Refractory 4d and 5d Metals

Metal	Melting Pt. (K)	Density at M.P.	$\times 10^4$	Temperature Range (K)
Zr	2123	6.09 (24)		
Nb	2741	7.68 (30) 7.57 (27)	5.4 (31)	(2741-4200)
Mo	2880	9.04 (30)	8.1 (31)	(2380-5000)
Rh	2239	12.20 (26)	50 (26)	(2239-2473)
Hf	2216	11.97 (24)		
Ta	3270	14.43 (30)	13 (31)	(3270-7000)
W	3690	16.26 (30) 16.67 (28)		
Ir	2716	19.39 (25)

Figure 1 Thermal expansion of liquid lead. Archimedeian technique ref. 42; Pycnometry, ref. 13; Direct measurement, ref. 31.

Figure 2 Thermal expansion of liquid uranium. Archimedeian technique, ref. 20; pycnometry, ref. 21.

References

1. F. R. Krueger and C. A. Swenson, J. App. Phys. 48, 853 (1977).
2. K. G. Lyon, G. L. Salinger, C. A. Swenson, and G. K. White, J. App. Phys. 48, 865 (1977).
3. F. W. Sheard, T. F. Smith, G. K. White, and J. A. Birch, J. Phys. C: Solid State 10, 645 (1977).
4. V. A. Petukhov and V. Ya Chekhovskoi, High Temp. High Press. 4, 671 (1972).
5. A. P. Muller and A. Cezairliyan, proceedings of the Seventh Symposium in Thermophysical Properties, National Bureau of Standards, Gaithersburg, Md., May 10-12, 1977.
6. Ya. A. Kraftmakher, High Temp. High Press. 5, 645 (1973).

7. See, for example, S. Bijunko and R. J. Hardy, *J. Phys. Chem. Sol.* 38, 147 (1977).
8. D. C. Wallace, *Thermodynamics of Crystals*, J. Wiley & Sons, N.Y., 1972, p. 404-427.
9. T. M. Reed and K. E. Gubbins, *Applied Statistical Mechanics*, McGraw-Hill, N.Y. 1973, p. 179.
10. H. D. Jones, *Phys. Rev.* A8, 3215 (1973).
11. L. A. Booth, *Nucl. Eng. Design* 24, 263 (1973).
12. L. D. Lucas in *Techniques of Metals Research*, R. F. Bunshah, ed. Vol. IV, part 2, 1970, p. 219.
13. L. D. Lucas, *Mem. Sci. Rev. Met.* 69, 395 (1972).
14. L. D. Lucas, *Mem. Sci. Rev. Met.* 69, 479 (1972).
15. S. D. Veazey and W. C. Roe, *J. Mater. Sci.* 7, 445 (1972).
16. A. F. Crawley, *Int. Metall. Rev.* 19, 32 (1974).
17. D. J. Steinberg, *Metall. Trans.* 5, 1341 (1974).
18. U. Evan and J. Magan, *J. Phys. E. Sci. Inst.* 7, 902 (1974).
19. U. Evan and J. Jortner, *Phil. Mag.* 30, 325 (1974).
20. A. V. Grosse, J. A. Cahill, and A. D. Kirshenbaum, *J. Amer. Chem. Soc.* 83, 4665 (1961).
21. W. G. Rohr and L. J. Wittenberg, *J. Phys. Chem.* 74, 1151 (1970).
22. T. Saito, Y. Shiraishi, and Y. Sakuma, *Trans. Iron Steel Inst. Japan* 9, 118 (1969).
23. H. Spetzler, M. Meyer, and G. Burcher, *High Temp. High Press.* 6, 529 (1974).
24. Yu. N. Ivashchenko and P. S. Martsenyuk, *Indust. Lab.* 39, 64 (1973). [transl. of *Zavod. Lab.* 39, 42 (1973)].
25. P. S. Martsenyuk and Yu. N. Ivashchenko, *Ukrain. Khim. Zhur.* 40, 431 (1974).

26. E. L. Dubinin, V. M. Vlasov, A. I. Timofeev, S. O. Safonov, and A. I. Chegodaev, *Izv. V. U. Z. Tsvetn. Metall.* No. 4, 160 (1975).
27. Yu. N. Ivashchenko and P. S. Martsenyuk, *High Temp.* 11, 1146 (1974). [transl. of *Teplofiz. Vys. Temp.* 11, 1285 (1974)].
28. P. S. Martsenyuk, Yu. N. Ivashchenko and V. N. Eremenko, *High Temp.* 12, 1161 (1975) [transl. of *Teplofiz. Vys. Temp.* 12, 1310 (1975)].
29. G. R. Gathers, J. W. Shaner, and R. L. Brier, *Rev. Sci. Inst.* 47, 471 (1976).
30. J. W. Shaner, G. R. Gathers, and C. Minichino, *High Temp. High Press.* 8, 425 (1976).
31. J. W. Shaner, G. R. Gathers and W. M. Hodgson, *Proc. of 7th Symposium on Thermophysical Properties*, May 10, 1977, Gaithersburg, Md.
32. G. R. Gathers, J. W. Shaner, and D. A. Young, *Phys. Rev. Lett.* 33, 70 (1974).
33. L. Martin-Garin, P. Bedon, and P. Desre, *J. Chim. Phys.* 70, 112 (1973).
34. H. Nakajima, *Trans. J. I. M.* 15, 301 (1974).
35. G. P. Khilya, Yu. N. Ivashchenko, and V. N. Eremenko, *Russian Metall. (Metally)* #6, 72 (1975) [transl. of *Izv. Ak. Nauk Metall.* #6, 87 (1975)].
36. A. P. Andreyev, V. A. Izmaylov, I. S. Ivakhnenko, and V. I. Kashin, *Russian Metallurgy (metally)* #2, 46 (1974) [transl. of *Izv. Ak. Nauk Metall.* #2, (1974)].
37. M. Gomez, L. Martin-Garin, H. Ebert, P. Bedon, and P. Desre, *Z. Metallkde.* 67, 131 (1976).
38. V. I. Yavovskiy, A. A. Ezhov, V. F. Kravchenko, V. S. Uskov, Yu. I. Nebusov, Yu. A. Chernov, and G. A. Dorofeyev, *Russian Metallurgy (Metally)* #4, 44 (1974).
39. C. O. Ruud, M. T. Hepworth, and J. M. Fernandez, *Metall. Trans.* 6B, 351 (1975).
40. S. Watanabe and T. Saito, *Trans. J. I. M.* 14, 120 (1973).

41. Yu. N. Karmalin, G. F. Stasyuk, M. I. Gladkov, V. M. Bagin, L. S. Etelis, and A. B. Borovskii, *Indust. Lab.* 39, 739 (1972) [transl. of *Zavod. Lab.* 39, 553 (1972)].
42. A. D. Kirshenbaum, J. A. Cahill, and A. V. Grosse, *J. Inorg. Nucl. Chem.* 22, 33 (1961).

*Work was performed under the auspices of the U.S. Energy Research and Development Administration under contract no. W-7405-Eng-48.

NOTICE

"This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research & Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately-owned rights."