Reprinted from THE REVIEW OF SCIENTIFIC INSTRUMENTS, Vol. 37, No. 5, 614–619, May 1966
Printed in U. S. A.

High Temperature-High Pressure Cell for Measuring Densities of Metals by Radiation Counting Technique*

I. G. DILLON,† P. A. NELSON, AND B. S. SWANSON‡
Chemical Engineering Division, Argonne National Laboratory, Argonne, Illinois 60440

(Received 25 August 1965; and in final form, 7 January 1966)

Equipment and techniques have been developed for measurement of densities of metals at high temperatures (up to 2200°K). The equipment and techniques were demonstrated using alkali metals but are generally applicable not only to most metals but also to many other substances. Determination of vapor and liquid densities of alkali metals at high temperatures and pressures was accomplished by measuring the radiation emanating from the vapor and liquid phases of a radioactive alkali metal contained in a high pressure cell fabricated from a molybdenum-30 wt.% tungsten alloy. The procedure was used to obtain vapor and liquid phase densities of the alkali metals at temperatures up to 2200°K and pressures up to 500 atm, which approach critical conditions. The radiation counting method involves sealing the metal whose density is to be measured in a containment capsule, irradiating the capsule and metal to produce a gamma emitting isotope of the metal, charging the capsule into the high temperature cell, and counting the activity of the vapor phase and the liquid phase at temperatures ranging from room temperature up to the critical point. A calibration of density vs irradiation level is performed at low temperatures where accurate liquid density data are available.

INTRODUCTION

THE advent of space and nuclear power programs has resulted in a great interest in high temperature properties of materials, particularly those of metals. Up to the present time, no convenient technique has been available for accurate measurement of densities at temperatures above 1500°K. Alkali metals are of particular interest to space programs because of their desirable properties for many uses such as ion engine fuels and turbine fluids. The physical and chemical properties of alkali metals make them attractive for use as nuclear reactor coolants. Equipment and techniques for making high temperature density measurements on metals or other materials has been demonstrated in this study by making such measurements on the alkali metals, cesium, rubidium, sodium, and potassium.

Densities below 1500°K are usually measured by pycnometer or dilatometer techniques.[1] Both methods involve weighing an accurately known volume. However, these techniques are accurate only at temperatures well below the boiling point of the material whose density is being measured. As the boiling point is approached, boiling and evaporation begin to occur and meaningful density measurements cannot be obtained. A technique previously used for determination of the densities of mercury liquid and vapor at temperatures up to the critical point employs

* Work performed at Argonne National Laboratory, operated by the University of Chicago under the auspices of the U. S. Atomic Energy Commission, Contract No. W-31-109-eng-38. Based on Ph.D. thesis of I. G. Dillon, Illinois Institute of Technology (1965).

† Present address: School of Engineering, Tuskegee Institute, Tuskegee Institute, Alabama 36088.

‡ Present address: Illinois Institute of Technology, Chicago, Illinois 60600.

[1] F. Tepper, A. Murchison, J. Zelenak, and F. Roelich, "Thermophysical Properties of Rubidium and Cesium," MSA Research Corporation Rept. No. MSAR-63-116 (Callery, Pennsylvania, February 1963).

measurements of electrical conductivity.[2] This method requires elaborate apparatus and is difficult experimentally. However, it has been used recently in work of Hochman and Bonilla[3] for determinations of density of some alkali metals as discussed below.

Basically, the method of density measurement used in this study involves activating the metal by irradiation in a thermal neutron flux to produce a radioactive isotope emitting a gamma ray suitable for counting. The metal must be contained by a material which has sufficient strength and corrosion resistance to the metal at the high temperatures involved in the measurements. The critical temperature and critical density of the metal may be then determined by measuring the densities of the vapor and liquid phases by counting the gamma activity emanating from known volumes of the irradiated metal. The critical temperature and critical density are then obtained from appropriate plots of the density data which cover the range from room temperature to the critical point. These techniques were applied to obtain the critical temperature and critical density of the alkali metals, cesium, rubidium, sodium, and potassium. If a suitable radioactive isotope of the metal whose density is to be measured does not exist, an attenuation method might be used. This would involve directing a collimated beam of radiation from a radioactive source through the metal and measuring the attenuation.

Measurement of properties at these high temperatures (up to 2200°K) and high pressures (up to 500 atm) requires special materials and techniques. Since at elevated temperatures refractory metal containers oxidize readily, it is necessary to provide a very pure inert atmosphere or to operate under high vacuum. The latter technique was used in these studies, since it was desirable to remove continually gas released by the insulation. A 76 cm diam×152 cm high vacuum tank was used as the containment vessel. A combination of a roughing pump and a diffusion pump maintained a vacuum of about 1 μ absolute pressure. In order to obtain the high temperatures required, the work piece was insulated with powdered tungsten carbide. The insulation was contained in a ceramic crucible. The alkali metal itself was contained in a thin wall capsule which in turn was contained in a massive refractory metal cylinder to provide both shielding from radiation and support against the high pressure forces within the capsule. The massive refractory metal cylinder has stepped diameter holes (simulating tapered holes which reduce scatter of radiation) leading from the vapor and liquid phase regions of the capsule wall toward the outside of the vacuum tank to the single channel analyzer which counted the collimated gamma radiation beams from the alkali metal.

DESCRIPTION OF EQUIPMENT

The alkali metal was usually contained in a molybdenum-30 wt.% tungsten capsule 3.81 cm long×0.64 cm o.d. (Fig. 1). Other materials such as molybdenum, tungsten, or molybdenum alloys were also used. These materials of construction were chosen for three reasons:

(1) Data from several sources[4–8] indicated molybdenum and tungsten had excellent corrosion resistance to alkali metals;

(2) this alloy has good high temperature strength; and

(3) neutron irradiation produced short lived radioisotopes of the container materials which would decay rapidly and not interfere with counting of the alkali metal whose density was being measured.

These same considerations would apply when selecting materials of construction for density measurements on other materials. The capsules were charged using the capsule charging assembly shown in Fig. 2. Prior to charging the capsule the alkali metals were contained in glass ampoules sealed under inert gas or vacuum. Charging was carried out in a high purity helium atmosphere glovebox.

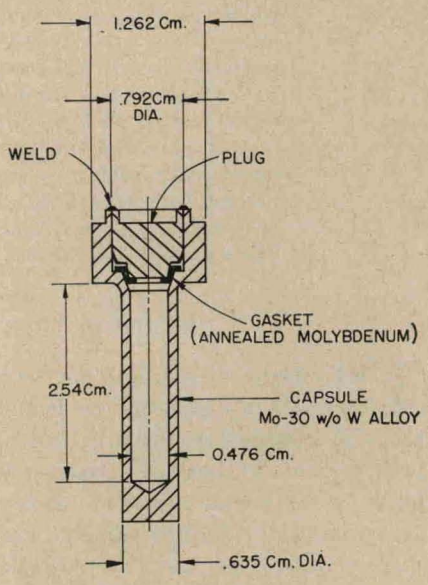

FIG. 1. Capsule for containment of alkali metals.

[2] F. Birch, Phys. Rev. 41, 641 (1932).

[3] J. M. Hochman and C. F. Bonilla, "The Saturated Vapor and Liquid Density of Cesium to 3000°F and the Critical Point of Cesium," USAEC Rept. No. CU-2660 (1965).

[4] A. V. Grosse, Research Institute of Temple University (private communication, 1963).

[5] M. G. Manzone and J. C. Briggs, "Less Common Alloys of Molybdenum," Climax Molybdenum Company (1962).

[6] R. Carlander, "The High Temperature Corrosion Resistance of Hastelloy "B" and Molybdenum to Rubidium," USAEC Rept. No. ORNL-CF-56-8-85 (14 August 1956).

[7] W. D. Weatherford, Jr., J. C. Tyler, and P. M. Ku, "Properties of Inorganic Energy-Conversion and Heat-Transfer Fluids for Space Applications," WADC-TR-61-96 (November 1961).

[8] M. J. Slivka, "A Study of Cesium Vapor Attack on Thermionic Convertor Construction Materials," Advanced Energy Conversion 3, 157 (1963).