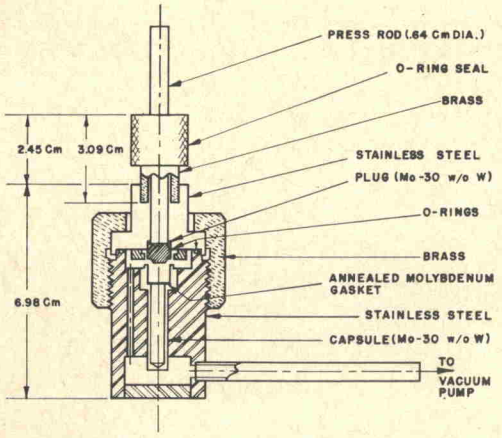

FIG. 2. Capsule charging assembly.

The capsule was charged with a weighed amount of material from the ampoule, evacuated, and then sealed under vacuum by deforming the molybdenum gasket by driving the tapered plug down with the pressing rod (Fig. 2). The sealed capsule was then removed from the capsule charging assembly and welded as shown in Fig. 1.

The capsule was contained in a cylindrical block of molybdenum-30 wt.% tungsten. The original design of the block called for a 3.08 cm diam×15.2 cm long cylinder in three pieces. The three pieces were held in fixed position inside a tantalum can by guide pins. The center piece was drilled radially to hold the capsule. The capsule was held in place by threaded end plugs. Since these plugs tended to seize under the extreme pressures and temperatures of the experiments, an improved design involving a two part split work piece was constructed (Fig. 3). This included a tapered Mo-30 wt.% W block of slightly over 3.08 cm diameter held in place by a 7.62 cm diam sleeve. This afforded more strength and easier removal of the capsule after heating. The work piece was contained in a 0.076 cm thick tantalum can to prevent reaction of the insulation with the molybdenum-tungsten block. The assembly was insulated by 6 μ diam tungsten carbide powder inside a silicon carbide crucible. Silicon carbide crucibles were chosen after testing alumina, magnesia, and zirconia crucibles which tended to crack and disintegrate with little use. Ceramically bonded alpha silicon carbide has a high thermal conductivity plus a low coefficient of thermal expansion and high strength all of which contribute to excellent thermal shock resistance.[9] These silicon carbide crucibles have shown no apparent change in strength or appearance after many (20–50) thermal cycles of the workpiece from room temperature to 2000°K (crucible temperature reached 1500°K maximum). A crucible lid made of Fiberfrax (an alumina-silica insulation made by the Carborundum Co.) held the powder in place. The crucible was supported by a fired lava (hydrous magnesium silicate made by American Lava Corporation) stand which was adjustable in the vertical dimension. As shown in Fig. 3, beam holes and thermocouple holes extending through the work pieces were connected to 0.64 cm tantalum tubing which extended out through the top and bottom of the crucible. The two 0.64 cm diam tantalum tubes extending from the beam holes lined up with two Veeco vacuum fittings in the wall of the vacuum tank. The Veeco fittings contained 1.27 cm stainless steel tubing sealed with a 0.025 cm stainless steel cap which permitted the radiation to pass with little attenuation.

The counting equipment outside the vacuum tank consisted of a single channel analyzer and associated equipment. This equipment included two high voltage sources, two preamplifiers, a scaler, two 2.54×2.54 cm NaI thallium activated crystals packaged with photomultiplier tubes and a recorder.

Selection of Gasket Material for Sealing Capsules

The three criteria for selection of a gasket material for forming the original capsule seal quickly limited the available materials. These three criteria were as follows:

(1) Vickers hardness number less than 350;
(2) resistant to alkali metal corrosion at 2000°C; and
(3) no high energy (>0.5 MeV) gamma produced by neutron irradiation.

Application of these three criteria limited the choice of materials to two; namely, annealed molybdenum and annealed rhenium. Tests of annealed rhenium showed that it was too hard and did not deform properly. Consequently, annealed molybdenum was chosen as the gasket material

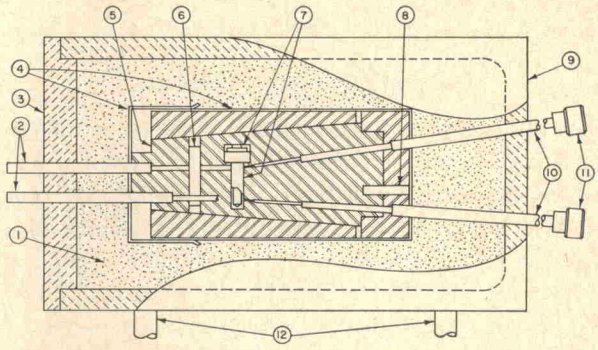

FIG. 3. Induction furnace assembly for vapor-liquid density measurements; 1—insulation; 2—tantalum tubes (0.64 cm o.d.) connecting to temperature measurement holes; 3—Fiberfrax lid; 4—tantalum can and lid; 5—tapered Mo-30 wt.% W cylinder in 7.62 cm o.d. Mo-30 wt.% W sleeve; 6—locating pin; 7—capsule containing test materials (see Fig. 1); 8—location pin; 9—silicon carbide crucible (16.5 cm o.d.×27.9 cm high×1.27 cm wall); 10—tantalum tubes (0.64 cm o.d.) connecting to collimation holes; 11—fitting mounted in vacuum tank wall and sealed with 0.025 cm diaphragm; and 12—refractory supports.

[9] F. C. Roe and E. D. Kwasniewski, Chem. Eng. Progr. **60**, No. 12, 97 (1964).

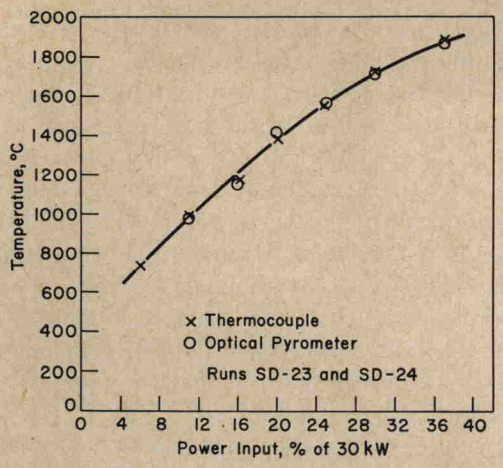

FIG. 4. Comparison of tungsten-5% rhenium/tungsten-26% rhenium thermocouple readings with optical pyrometer readings.

and it performed quite well for alkali metals. For measurement of densities of other metals, different criteria might apply so that other materials could be used for a gasket.

Temperature Measurement

Temperatures were measured by tungsten-5% rhenium/tungsten-26% rhenium thermocouples. These were calibrated using a model 8622-C Leeds & Northrup optical pyrometer. This pyrometer was calibrated with a standard tungsten lamp filament and against a standardized pyrometer. Maximum error in comparison with the other pyrometer and the tungsten lamp was less than 10°C. The pyrometer was then used to check the calibration of thermocouples over the temperature range 700 to 1900°C. A typical calibration curve is shown in Fig. 4. A standard procedure in obtaining the data was to calibrate thermocouples before and after a run to be certain there was no "drift" in thermocouple voltage with repeated temperature cycling.

Radiation Counting Technique

The radioactive tracer technique is based on the fact that small amounts of material may be measured accurately by radioactive counting techniques. Where the tracer isotope is homogeneously distributed through the phase being measured (as in this case) the radioactive count rate is directly proportional to the amount of material present if self-absorption is negligible. Measurements of radioactive count rate of the liquid were compared with density determinations by known standard techniques at lower temperatures (25 to 1300°C) and the relationship between radioactive count rate and density was shown to be constant. This was checked also with the capsule in an inverted position and the same relationship prevailed. This showed that the geometries of the vapor and liquid beam paths were the same. It may readily be shown[10] that the effect of self-absorption is as follows,

$$A_c = R(a\mu r\rho^3 + b\mu^2 r^2\rho^2 + \rho) = R\rho f, \quad (1)$$

where A_c is the activity measured at counter, R the geometry factor determined experimentally by counting liquids of known density, μ the gamma ray absorption coefficient (cm^2/g), r the radius of cylinder (cm), a, b the proportionality constants, ρ the density of material (g/cc), and f the self-shielding factor or fraction of activity escaping. The fraction of activity escaping can be seen to be

$$f = a\mu r\rho^2 + b\mu^2 r^2\rho + 1. \quad (2)$$

For the alkali metals, values of μ are much less than 1 and at high temperatures the density ρ is less than 1. Therefore, for the alkali metals at high temperatures, f is equal to 1 and self-shielding is negligible.

The counting equipment described above was calibrated using irradiated standards. Standards chosen for calibration of the equipment for counting some alkali metals were the following:

Element	Form of standard
Cesium	Cesium nitrate salt
Rubidium	Rubidium metal
Potassium	Potassium nitrate salt
Sodium	Sodium nitrate salt

These standards were irradiated in the CP-5 research reactor at Argonne National Laboratory so as to give a counting of about 50 000 counts/min. The counting rates and spectra determined on a single channel analyzer agreed well with those reported by Crouthamel.[11]

Errors in the counting technique included the usual factors of geometry and statistics and the additional factor of alignment of the beam holes. The source was made sufficiently active so that the counting rate was always greater than 5000 counts/min. Conversion to density from counts per minute was made on a comparative basis; thus, many factors such as geometry and resolving time were eliminated. Variations in efficiency were guarded against by counting standards at frequent intervals during the runs to see that the absolute counting rates did not vary.

OPERATION

Operating Steps

The twelve basic steps in making a run are as follows:

(1) Charging material to the capsule.
(2) Testing the capsule for integrity by heating to 1275°C.

[10] I. G. Dillon, Ph.D. Thesis, Illinois Institute of Technology (1965).
[11] C. E. Crouthamel, *Applied Gamma-Ray Spectrometry* (Pergamon Press, Inc., New York, 1960).