DP-1096

AEC RESEARCH AND DEVELOPMENT REPORT

# DEVELOPMENT OF $^{60}Co$ CAPSULES FOR HEAT SOURCES

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Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.65

664208

DP-1096

Isotopes - Industrial Technology
Propulsion Systems and Energy Conversion
(TID-4500)

# DEVELOPMENT OF $^{60}Co$ CAPSULES FOR HEAT SOURCES

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June 1967

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*CONTRACT AT(07-2)-1 WITH THE*
*UNITED STATES ATOMIC ENERGY COMMISSION*

## ABSTRACT

Radioactive cobalt containing 100 curies of $^{60}Co$ per gram of metal has been encapsulated in "Inconel"* 600 and "Hastelloy"** C containers. Such capsules, producing up to 250 watts, have been tested successfully at 850°C for 100 hours in air. Longer tests are under way.

Preliminary tests indicate that TD Nickel† (2% thoria-dispersed) and TD Nickel Chromium† (78 nickel - 20 chrome) are also promising materials for encapsulation.

About 8 megacuries of $^{60}Co$ at activities up to 400 Ci/g will be available for heat source development and demonstrations by May 1967.

Progress is reported in the areas of $^{60}Co$ production, fuel form properties, capsule design, capsule material properties, and capsule fabrication and testing.


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# INTRODUCTION

Radioisotope power sources will be essential for maximum development of remote regions on land, under the sea, and in space. They operate unattended for long times, they are simple and self-contained, and individual units can produce from milliwatts to kilowatts of electric power or process heat.

Cobalt-60 is a promising isotope for many large-scale uses because it can be produced in large quantities at low cost. Production capacity of the Savannah River reactors, if not used for other purposes, is hundreds of megacuries per year, and the cost of production is less than 25¢ per curie. Each megacurie produces 15.6 kw of heat initially and 13.7 kw after one year of decay. $^{60}Co$ has a convenient half-life (5.3 years) both for reasonable mission duration and ultimate disposal, and can be produced at specific activities above 300 curies per gram (41 watts/cm$^3$ of cobalt metal). A comparison of power densities of useful isotopes is given in Table I and Figure 1.

Because high activity $^{60}Co$ is a compact energy source, the shields that must be provided to recover the energy from the gamma radiation (1.17 and 1.33 Mev) and to provide biological shielding are not excessively heavy. Shielding weighs less than that required for $^{90}SrTiO_3$ sources at powers above about 2 kilowatts thermal. The weight of spherical shields of depleted uranium for the two isotopes is shown in Figure 2. Although this shielding arrangement is idealized for minimum weight, it approximates arrangements that would be used for practical heat- and power-producing units.

This report summarizes progress during 1966 in the Savannah River Laboratory (SRL) program to develop data for designing, fabricating, and operating $^{60}Co$ heat sources. It is the second such status report on this program, the first being DP-1012, Radioactive Cobalt for Heat Sources, October 1965,[1] which was published near the beginning of the program and included information on the production and properties of radioactive cobalt. Additional property data are given in DP-1051, Rev. 1, Properties of $^{60}Co$ and Cobalt Metal Fuel Forms, October 1966.[2] The status of this and other SRL isotope programs is reported quarterly.[3,4] The SRL program includes:

- Designing irradiation conditions for producing large amounts of high-activity cobalt.
- Establishing specifications for cobalt fuel forms and determining effects of irradiation and decay.

- Designing suitable capsules based on predicted operating conditions.

- Determining limiting operating conditions for encapsulating materials based on strength, compatibility, and oxidation.

- Developing encapsulation processes and testing experimental capsules to confirm performance under heat source operating conditions.

Goals of the program include developing and establishing allowable operating limits for:

- Capsules of radioactive cobalt metal in an oxidation-resistant alloy by the end of FY 1968.

- Capsules of a higher temperature fuel form of $^{60}Co$ in a refractory metal alloy by the end of FY 1971.

The program does not include:

- Development or testing of large heat sources (several capsules with associated shielding).

- Development of power conversion systems.

TABLE I

Power Densities of Radioisotope Fuel Forms

| Isotope | Half-Life, years[5] | Specific Power of Pure Isotope, w/g | Fuel Form | Fuel Form Density, g/cm³ | Initial Power Density, watts/cm³[6] |
|---|---|---|---|---|---|
| $^{238}Pu$ | 89 | 0.55 | $PuO_2$ | 10.0 | 3.5 |
| $^{137}Cs$ | 30 | 0.48 | Cs glass | 3.2 | 0.21 |
| $^{90}Sr$ | 28 | 0.93 | $SrTiO_3$ | 3.9 | 0.85 |
| $^{244}Cm$ | 18 | 2.7 | $Cm_2O_3$ | 10.6 | 26 |
| $^{60}Co$ | 5.3 | 17 | Metal | 8.8 | 41 (300 Ci/g) |
| $^{147}Pm$ | 2.7 | 0.36 | $Pm_2O_3$ | 6.7 | 2.0 |
| $^{144}Ce$ | 0.78 | 25 | $Ce_2O_3$ | 6.1 | 20 |
| $^{210}Po$ | 0.38 | 140 | Metal matrix | 8.5 | 75 (50% void vol) |
| $^{170}Tm$ | 0.34 | 12 | $Tm_2O_3$ | 8.6 | 15 (900 Ci/g) |

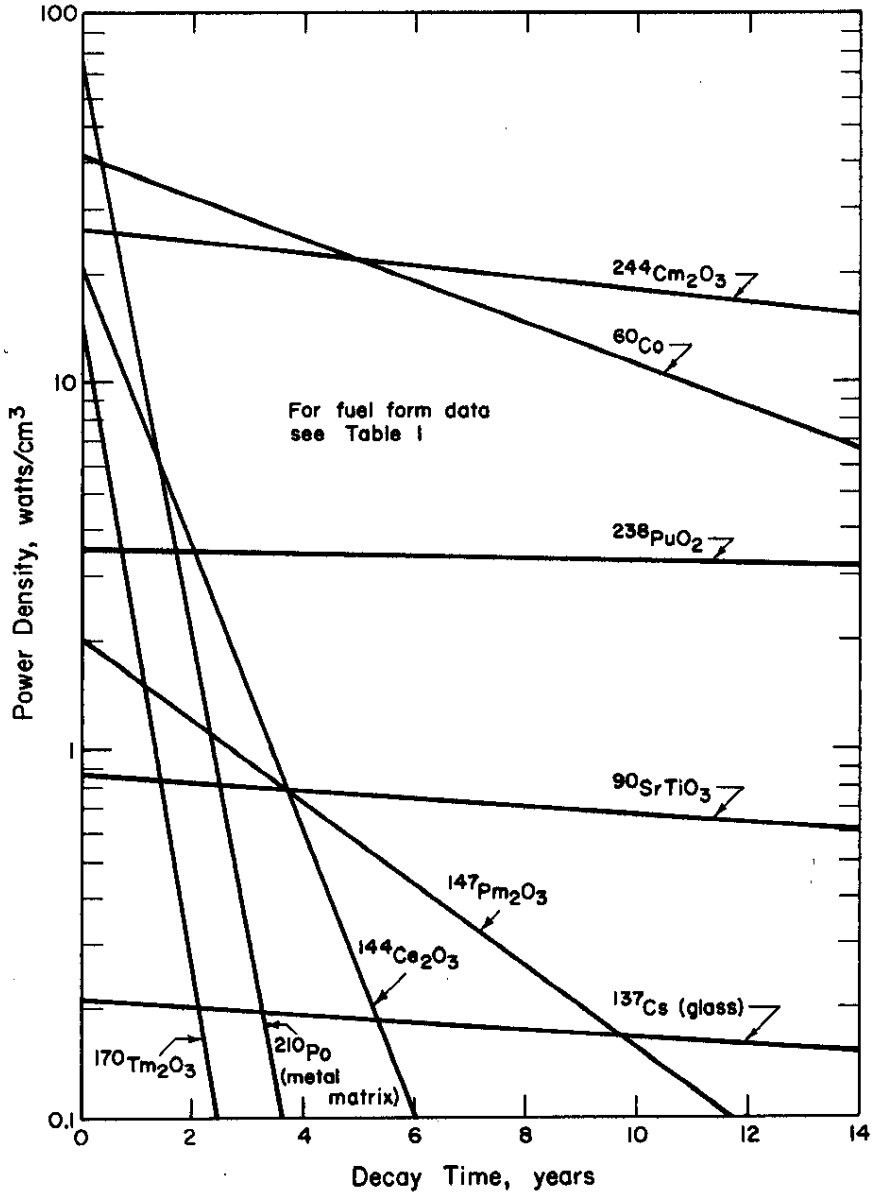


FIG. 1 POWER DENSITIES OF RADIOISOTOPE FUEL FORMS

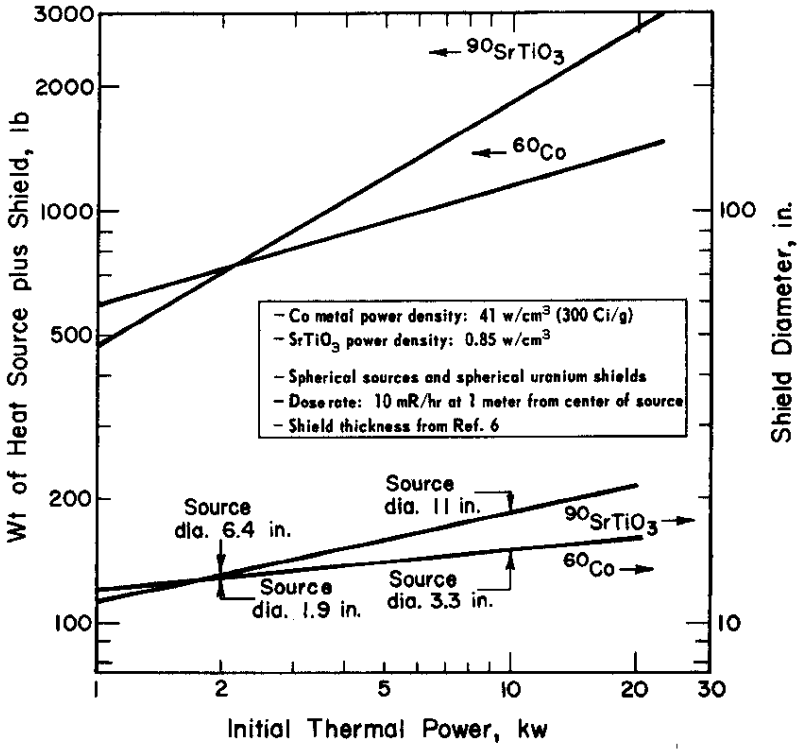


FIG. 2 SHIELD WEIGHT COMPARISON FOR $^{60}Co$ AND $^{90}SrTiO_3$ HEAT SOURCES

## SUMMARY

The Savannah River Laboratory program on $^{60}Co$ includes all aspects of heat source development from the specification of raw materials through reactor irradiation to fabrication of experimental capsules and testing at heat source temperatures.

About 8 megacuries of $^{60}Co$ (125 kw) at up to 400 curies per gram of metal (55 $w/cm^3$) are being produced for use in heat source development and for possible use in experimental heat sources. Most of this material is in the form of 0.745-inch-diameter wafers that are 0.040-inch thick and plated with up to 1 mil of nickel.

Metallographic examinations revealed no significant dimensional or structural changes in cobalt metal irradiated to 700 Ci/g (96 $w/cm^3$) or in cobalt metal irradiated to 300 Ci/g and allowed to decay for one half-life.

Design of capsules for $^{60}Co$ fuel forms is governed by long-term strength, compatibility, and oxidation considerations. Wall thicknesses of 50 to 100 mils should be adequate for oxidation-resistant alloys to contain cobalt metal at 800 to 1000°C for up to 5 years in inert atmospheres and at least 1 year in oxidizing atmospheres.

Preferred encapsulating materials based on compatibility tests for 1 week at 800, 1000, and 1200°C are "Inconel"* 600 and TD Nickel† (2% thoria-dispersed). Of these, "Inconel" 600 has the better oxidation resistance and TD Nickel the better long-term strength. Long-term tests of nonradioactive capsules made from these alloys are underway. Preliminary tests of TD Nickel Chromium† show that oxidation resistance of TD Nickel is improved markedly by the addition of chromium. Compatibility of this alloy is being evaluated.

Capsules of "Inconel" 600 and "Hastelloy"** C containing 16,000 curies (250 watts) and 9000 curies (140 watts), respectively, were tested successfully for 100 hours at 850°C in air. Capsules are being fabricated for tests at higher temperatures and for longer times. Requirements to prevent excessive temperatures during assembly of radioactive capsules in the high level caves have been established by calculations and by experience with 100-Ci/g cobalt. Experimental capsules producing up to 1000 watts and containing up to 350 Ci/g will be fabricated and tested.


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## PRODUCTION OF RADIOACTIVE COBALT

Large quantities of high activity $^{60}Co$ can be produced in Savannah River reactors either as the major product of a reactor charge or as one of several products in mixed-lattice charges. Both methods are being studied. These studies depend only loosely on the choice of cobalt fuel form; a wide variety of cobalt shapes can be irradiated in target positions up to 3-1/2 inches in diameter.[1]

To provide the high activity cobalt metal that will be required for development and demonstration of heat sources, significant quantities of $^{60}Co$ are being produced in control rods of high-flux reactor charges. About 800,000 curies of $^{60}Co$ (12.5 kw) at an average of 220 curies per gram (30 w/cm$^3$) is on hand as 0.75-inch-diameter x 0.040-inch-thick nickel-plated wafers. Another 6 million curies (93 kw) as 0.745-inch-diameter wafers plus 2 million curies (32 kw) as 0.80-inch-diameter wafers and 3-inch-long slabs of various cross sections are being produced. Dimensions of the targets and the amount of $^{60}Co$ produced are given in Tables II and III. Typical impurity content of this material is given in Table IV.

Up to 1 megacurie of this $^{60}Co$ will be used for property studies, encapsulation studies, and small-scale tests at the Savannah River Laboratory. The remainder will be available by May 1967 for use in demonstration heat sources or could be supplied for use in radiation sources.

TABLE II

High Specific Activity $^{60}Co$ Inventory[a]

| Specific Activity, Ci/g | | Average Specific Power | | No. of Wafers[b] | Total Weight, g | Total Power, w | Total Activity, Ci |
|---|---|---|---|---|---|---|---|
| Range | Avg | w/g | w/cm$^3$ | | | | |
| 30-40 | 36 | 0.56 | 5.0 | 190 | 480 | 270 | 17,400 |
| 85-115 | 100 | 1.6 | 14 | 95 | 240 | 380 | 24,200 |
| 100-140 | 120 | 1.9 | 16.5 | 95 | 240 | 450 | 29,000 |
| 130-180 | 155 | 2.4 | 21 | 95 | 240 | 580 | 37,400 |
| 180-280 | 220 | 3.3 | 30 | 1520 | 3840 | 12,700 | 817,000 |
| | | | | | Total | 14,380 | 925,000 |

(a) Inventory as of December 1966

(b) Diameter: 0.750 ±0.003 inch; thickness 40 ±3 mils
Density: 8.80 ±0.05 g/cm$^3$
Nickel-plating thickness: 0.05 to 1.0 mil

TABLE III

High Specific Activity $^{60}Co$ Production[a]

| | No. of Pieces | Total Grams | Total MCi | Total kw |
|---|---|---|---|---|
| Nickel-plated wrought wafers<br>Diameter 0.745 ±0.001 in.<br>Thickness 40 ±3 mils[b] | 6,688 | 16,830 | 6.0 | 93 |
| Nickel-plated sintered wafers<br>Diameter 0.800 ±0.001 in.<br>Thickness 40 ±3 mils[b] | 527 | 1,490 | 0.53 | 8.3 |
| Nickel-plated wrought slabs<br>Length 3.00 ±0.03 in.<br>Width 0.640 ±0.002 in.<br>Thickness 60 ±1 mil[b] | 93 | 1,550 | 0.54 | 8.4 |
| Stainless-steel-clad wrought slabs[c]<br>Length 2.96 ±0.03 in.<br>Width 0.735 - 0.740 in.<br>Thickness 92 ±1 mil | 93 | 1,250 | 0.44 | 6.9 |
| Stainless-steel-bonded wrought slabs[c]<br>Length 3.00 ±0.03 in.<br>Width 0.740 ±0.002 in.<br>Thickness 72 ±2 mils | 124 | 1,470 | 0.52 | 8.1 |
| | | Total | 8.0 | 125 |

(a) Estimated production as of April 1967. (Avg specific activity: 350 Ci/g. Avg specific power: 48 w/cm$^3$. Activities range from 300 to 400 Ci/g.)
(b) Includes 0.5 to 1.0 mil of nickel plate.
(c) Cladding is minimum of 15 mils thick.

TABLE IV

Chemical Composition of Cobalt Raw Material

| | Tentative Specification | | Typical Current Analysis | |
|---|---|---|---|---|
| Element | Minimum wt % | Maximum ppm (wt) | wt % | ppm (wt) |
| Co + Ni | 99.9 | | 99.3 | |
| Ni | | 1500 | | 3800 |
| Fe | | 1000 | | 140 |
| Cu | | 100 | | 15 |
| O | | 100 | | 300 |
| Si | | 100 | | 3000 |
| Th | | 100 | | <1 |
| U | | 100 | | <1 |
| Al | | 50 | | 140 |
| Cd | | 50 | | <1 |
| Mn | | 50 | | 3000 |
| Pb | | 50 | | 1 |
| S | | 50 | | 15 |
| Cr | | 20 | | 30 |
| Mo | | 20 | | <1 |
| V | | 20 | | 1 |
| W | | 20 | | 6 |
| P | | 10 | | 5 |
| B | | 5 | | 1 |
| Gd | | 5 | | <1 |
| Li | | 5 | | <1 |

# FUEL FORM PROPERTIES

## CHEMICAL CONTENT OF COBALT METAL

Current technology indicates that commercially available cobalt has satisfactory properties at expected heat source temperatures. Pieces of the desired dimensions are coated with a layer of nickel about one mil thick to minimize the spread of contamination during handling after irradiation. The nickel layer is electrodeposited on the cobalt.

To confirm that impurities normally present in cobalt metal do not adversely affect irradiation of the cobalt or its use in heat sources, representative samples from several sources were evaluated. Evaluation consisted of measurements of the density, chemical composition, and melting point, and examination of the microstructure. Results of these tests were used to establish the allowable impurity content for cobalt metal (Table IV).

Of the common impurities in commercially available cobalt, sulfur is the most deleterious because it lowers the melting point and causes embrittlement. Since a typical manufacturing process produces cobalt with up to 50 ppm sulfur, and since reducing the sulfur content would add appreciably to the price, samples of various sulfur contents will be annealed at high temperature to determine whether low melting phases are formed from migration of sulfur to grain boundaries and whether this sulfur affects the compatibility between cobalt and encapsulating material. If adverse effects are found the specification on sulfur may be reduced to below 50 ppm.

Small concentrations of some elements are specified to prevent potentially detrimental effects. These elements include:

- Impurities that generate gases during irradiation. Such impurities are boron, lithium, uranium, and thorium. Gases generated from these impurities could cause swelling during high temperature operation of a heat source.

- Impurities that lower the melting point. These include lead, oxygen, phosphorus, and carbon.

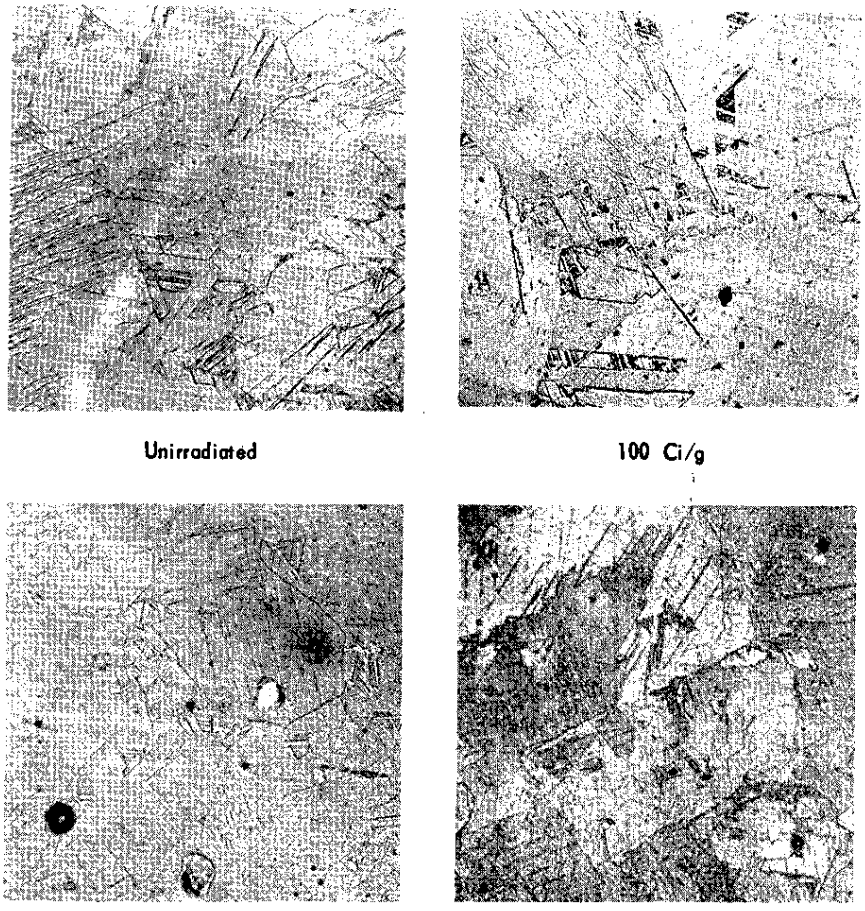
- Elements that affect reactivity during irradiation. These include gadolinium, cadmium, and boron.

Special samples of material not meeting these specifications are being irradiated to determine if higher concentrations of impurities are allowable.

## EFFECTS OF IRRADIATION AND DECAY

Changes in dimensions, density, hardness, and microstructure of irradiated cobalt metal are being measured to define the effects of irradiation and radioactive decay on the properties of the metal. Other properties will be investigated as required for particular applications.

In February 1966, a total of 980 curies of $^{60}Co$ with the highest specific activity yet attained, 700 Ci/g (96 w/cm$^3$), was discharged from the Savannah River reactor that had been operated at high neutron fluxes (up to 6 x $10^{15}$ n/(cm$^2$)(sec)). Metallographic examination of this cobalt, which consists of two 1-cm-diameter by 1-mm-thick wafers, indicated that the microstructure was almost identical to that of unirradiated cobalt (Figure 3).



Unirradiated | 100 Ci/g

700 Ci/g | 300 Ci/g decayed to 140 Ci/g (~6 years)

FIG. 3 EFFECT OF IRRADIATION AND DECAY ON MICROSTRUCTURE OF COBALT METAL (200X)

The same conclusion was reached for pellets irradiated to 300 Ci/g and allowed to decay to 140 Ci/g (Figure 3). Earlier work showed that the microstructure of cobalt with normal impurities was not altered significantly, and the impurities were not redistributed, by irradiation to 100 Ci/g,[1] by thermal cycling,[7] or by annealing in a thermal gradient that spanned the allotropic transformation temperature.[7]

After examination, the high activity cobalt samples will be stored for later examination to determine the structural effects of further radioactive decay and the associated increase in nickel content. During a 5.3-year half-life, the $^{60}Co$ concentration of the 700-Ci/g material will decrease from 62 to 31% and the nickel content will increase from the present 11 to 42%. $^{60}Co$ concentration in the sample that has decayed from 300 to 140 Ci/g is ~13%, and nickel content is ~22%. A portion of the 700-Ci/g material will be available for specialized applications.

## HEAT GENERATION IN RADIOACTIVE COBALT

Heat produced in irradiated slugs and individual cobalt pieces is dissipated by natural convection and radiation to the environment. About 4% of the energy released by $^{60}Co$ decay is beta energy; in heat transfer calculations, all this energy is assumed to be released in the cobalt itself. The remaining 96% is released as 1.17- and 1.33-Mev gamma rays; the absorption of the energy in the cobalt is estimated from Figure 4, which is based on data from Reference 8.

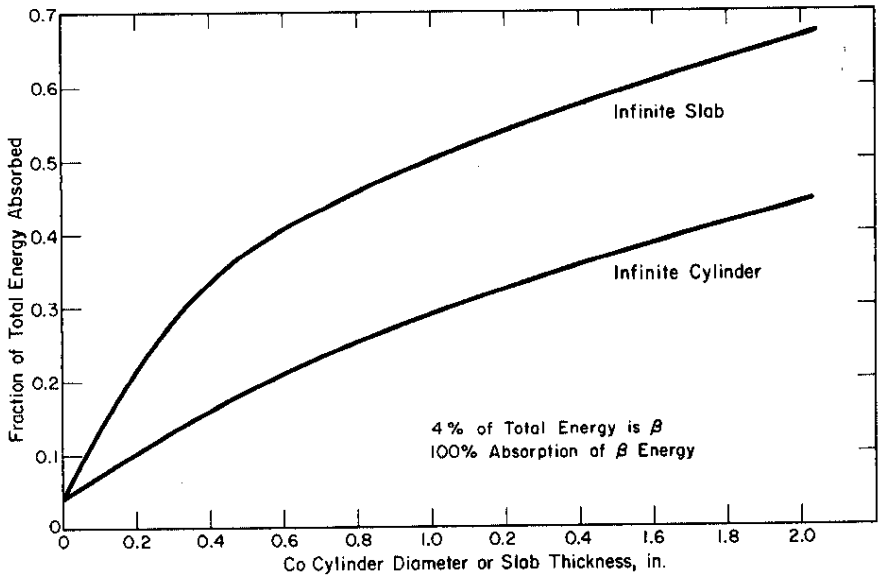



FIG. 4 SELF-ABSORPTION OF COBALT METAL SHAPES

### Irradiated Slugs

As irradiated, cobalt is usually contained in aluminum slugs that provide a relatively large heat transfer surface (Figure 5). Temperatures in irradiated slugs in either air or water are not excessive if a capacity to absorb the total heat generated is provided during shipment or storage. The maximum $\Delta T$ between slug surface and still air is 250°C for 400-Ci/g cobalt.

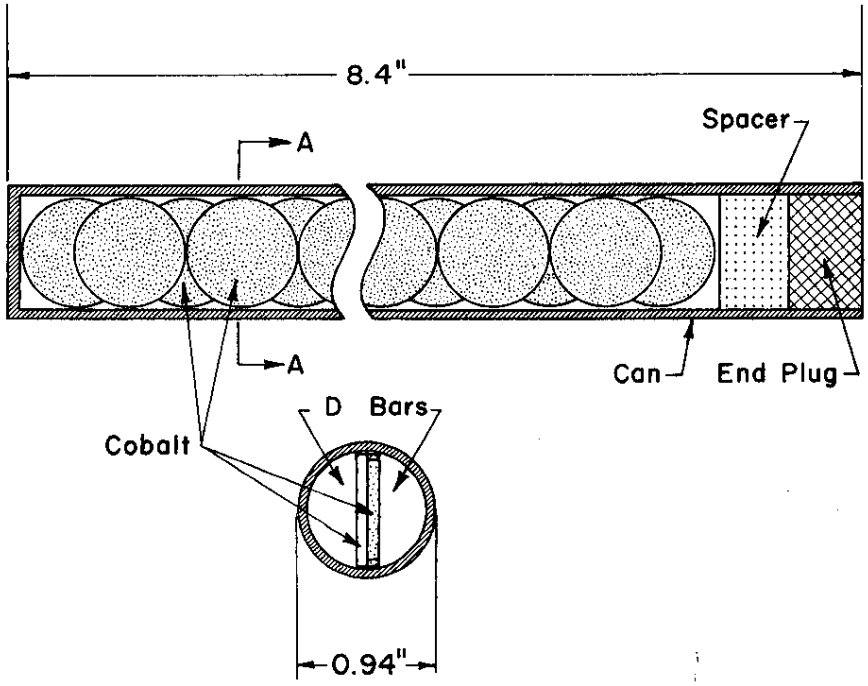



FIG. 5 ALUMINUM SLUG FOR COBALT METAL WAFERS

### Individual Cobalt Pieces

Although radioactive cobalt with high specific activity has an intense volumetric energy rate, only 5 to 10% of the energy appears as heat within the thin pieces normally irradiated. Consequently, the temperature of individual pieces does not exceed about 300°C even when they are placed on nonconducting surfaces in air (Table V). Pieces can be further cooled by immersion in liquid or by contact with conductive surfaces. The values in Table V are not applicable when pieces are stacked or piled.

TABLE V

Heat Transfer from Individual Cobalt Pieces

(on nonconducting surface)

| | Wafers[a] | | Slabs[b] | |
|---|---|---|---|---|
| Specific activity, Ci/g | 200 | 400 | 200 | 400 |
| Total activity, Ci | 510 | 1020 | 3340 | 6680 |
| Total heat generation, watts | 8 | 16 | 51 | 102 |
| Internal heat generation, watts[c] | 0.5 | 1.0 | 4.6 | 9.2 |
| Adiabatic heating rate, °C/min | 24 | 48 | 36 | 72 |
| Maximum heat flux, Btu/(hr)(ft$^2$) (from edges and one flat surface) | 430 | 860 | 960 | 1920 |
| ΔT, °C | | | | |
| Cobalt to 25°C air | 80 | 150 | 180 | 290 |
| Cobalt to 25°C water | 3 | 6 | 7 | 13 |

(a) 0.75-inch diameter, 40 mils thick.
(b) 3 inch by 0.064 inch, 60 mils thick.
(c) 6% internal absorption for wafers; 9% for slabs.

## HIGH TEMPERATURE FUEL FORMS

The use of cobalt metal in heat sources will be limited to some temperature below its melting point of 1495°C because the cobalt either will be incompatible with the cladding material or will become molten during temperature excursions associated with interruption of coolant flow. $^{60}$Co may be suitable for use in thermionic power conversion systems if a fuel form can be found that is stable above 1500°C, the minimum temperature for efficient operation of a thermionic converter. The SRL program includes a small effort towards identification of such fuel forms.

### Cobalt-Rhenium Alloy

The most promising high-temperature forms of cobalt are the solid solution alloys of the cobalt-rhenium system. Alloys containing 30 to 60 vol % Co melt between 2200 and 1700°C; power densities of 12.5 to 25 w/cm$^3$ could be attained in these alloys with 300-Ci/g $^{60}$Co. These alloys would be prepared after irradiation, because irradiation would convert rhenium to osmium, which lowers the melting point significantly.

**Cobalt Oxides**

Cobaltous oxide (CoO) and solid solutions of cobalt and magnesium oxides are being evaluated as possible high-temperature forms of cobalt that could be prepared before irradiation. CoO has a melting point of 1745°C and a power density of 24 $w/cm^3$, based on 300-Ci/g cobalt. Chemical stability of CoO in contact with the refractory metals is at best marginal. The free energy change for the reduction of CoO by refractory metals is negative in all cases.

The CoO-MgO system shows complete solid solubility and a somewhat lower power density for a given melting point than the Co-Re system. Since MgO is one of the most stable oxides, alloying CoO with MgO might increase the chemical stability and perhaps change the sign of the free energy change for the reduction of the solid solution.

## CAPSULE DESIGN

The initial objective of the $^{60}Co$ program is to develop satisfactory capsules of radioactive cobalt metal contained in oxidation-resistant materials such as nickel- or cobalt-based superalloys. The operating temperature limit for such capsules is expected to be above 800°C and perhaps as high as 1000°C for lifetimes up to 5 years in inert gases and at least 1 year in air. Operation for up to 5 years in air may be possible at somewhat lower temperatures.

Capsules must also be able to withstand emergency conditions such as impact, thermal shock, loss at sea, or elevated temperatures for short times due to fire or loss of coolant. However, long-term properties such as compatibility, creep or stress rupture, and oxidation, will be controlling in selection of materials and design of capsules for normal operating temperatures, and these properties are emphasized in the Savannah River program.

### DESIGN CHARACTERISTICS FOR LONG-TERM OPERATION

The thickness of a capsule wall must provide adequate strength to withstand the maximum calculated stress over the capsule's operating lifetime. On the assumption that neither the zones formed by diffusion of cobalt nor the oxidized surfaces contribute to the capsule's strength, the initial wall thickness required is the sum of three layers (Figure 6):

- Wall thickness required to provide mechanical strength
- Depth of interaction with cobalt
- Depth of oxidation penetration

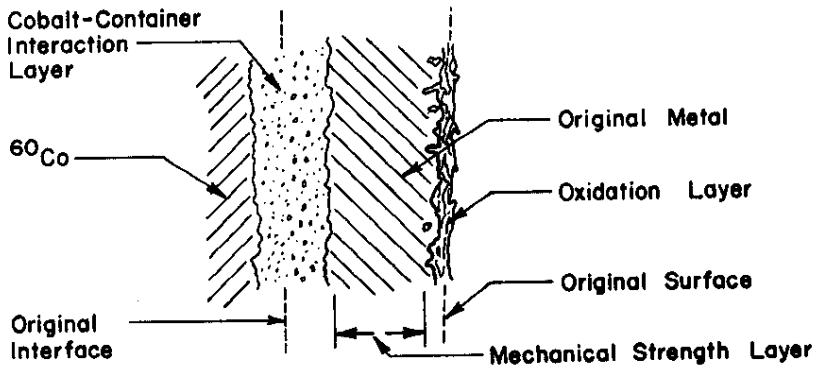



FIG. 6 DIFFUSION AND OXIDATION OF CAPSULE WALL

The wall must also be thick enough to prevent excessive diffusion of $^{60}Co$ to the exterior. This $^{60}Co$ would become part of the oxidation layer and could produce a radiation hazard if carried outside the biological shield in systems where the coolant contacts the capsules.

## STRESSES IN CYLINDRICAL CAPSULES

Experimental capsules are being designed so that end caps and cap-to-wall junctions are stronger than the cylindrical wall. Pressure rupture tests at room temperature have demonstrated preferential failure in 50-mil-thick capsule walls.

Capsule internal pressure, primarily from heating the gases initially present, will be about 60 psig for a capsule welded at ambient temperature and heated to 1000°C. This pressure results in a maximum circumferential stress of 300 psi in a 1-inch-diameter capsule with a 100-mil-thick wall. This relatively low stress allows consideration of container materials for use at temperatures where their strength is low by normal industrial standards. Oxidation-resistant materials being considered for $^{60}Co$ encapsulation will have satisfactory creep rates with this stress at temperatures above 800°C and perhaps as high as 1000°C.

Measurements indicate that gases released from encapsulated cobalt will not pose significant problems to capsule designers. In one test, pressure was measured as a cobalt-filled "Hastelloy" C capsule was heated to 1200°C to determine the pressure contribution from released gases. Helium present when the capsule was welded was assumed to remain as a gas. As shown in Figure 7, the apparent gas release reached a maximum of 0.004 cc(STP)/g of cobalt at about 500°C.

In a second test, four capsules were heated and the contained gases were sampled at temperatures up to 1000°C. Net gas release was calculated from the analysis of the samples, and the four data points are also shown in Figure 7. Gas release in these capsules was a maximum of 0.006 cc(STP)/g of cobalt at 350°C. Apparent gas release in the tests was much less than the 0.05 cc(STP)/g of cobalt released by vacuum extraction at 1200°C. Absorption of the gas (primarily hydrogen) and permeation through the 50-mil-thick capsule wall are believed to account for the decrease in gas "release" above 350-500°C. Figure 8 shows the expected increase of pressure with temperature in capsules of nickel- or cobalt-based alloys containing radioactive cobalt metal. Capsules are assumed to be loaded with a void volume equal to 5% of the cobalt volume, and the closure weld is conservatively assumed to be made with the capsule at room temperature and atmospheric pressure.

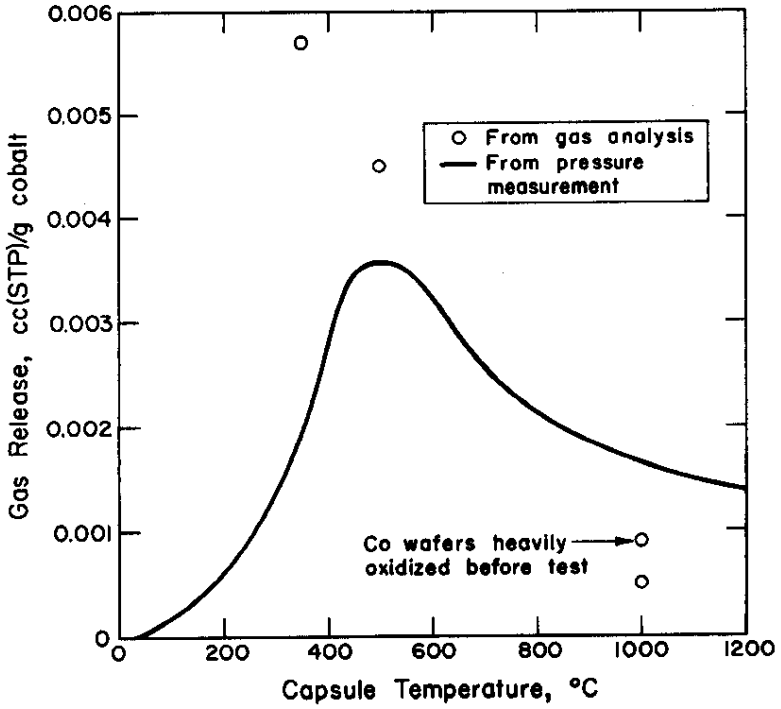



FIG. 7 GAS RELEASE FROM ENCAPSULATED COBALT (UNIRRADIATED)

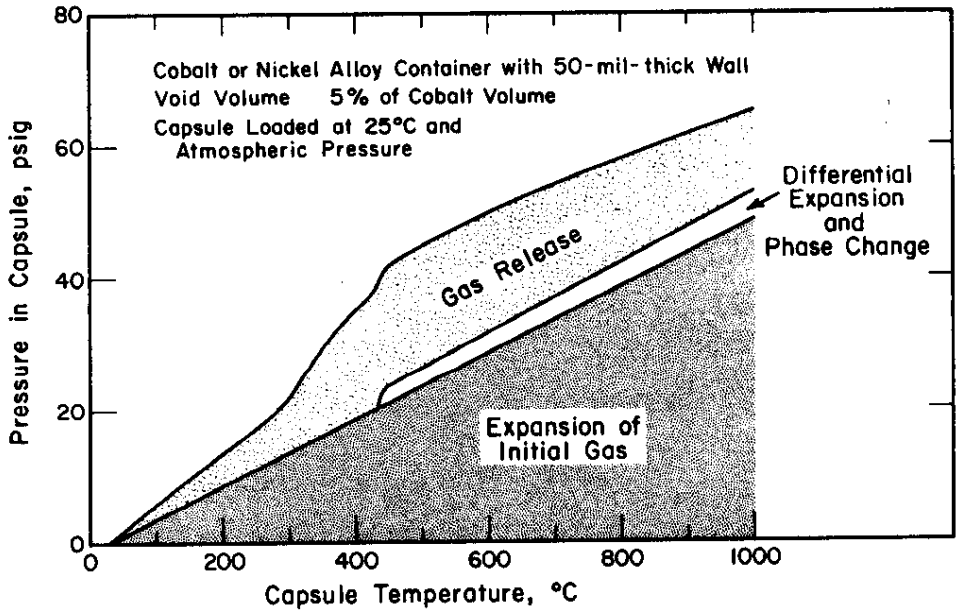



FIG. 8 CAPSULE PRESSURE INCREASE WITH TEMPERATURE

Drying of all capsule components is essential to prevent high vapor pressure from residual liquids.

Failure of a capsule due to internal pressure at long times will be by either excessive creep or stress rupture. For a diameter of 1 inch, a diametral creep of about 0.5% (5 mils) should be acceptable. This value will be in the range of loading clearances between the capsule and supporting structures. If the capsule were used in coolant channels of reasonable size, a 5-mil increase in diameter would not increase the coolant pressure loss unduly. For service of 5 years, a creep of 0.5% corresponds to about $1 \times 10^{-5}$% per hour.

## HEAT GENERATION IN CAPSULES

Experimental $^{60}Co$ capsules will enclose stacks of 0.75-inch-diameter wafers or a square array of slabs each 0.64 inch wide by 0.06 inch thick. Concentrating the cobalt pieces increases the internal heat generation to about 26% of the total energy produced and decreases the heat transfer area per piece. Consequently, the surface heat flux of a 0.75-inch-diameter cobalt stack may exceed 20,000 Btu/(hr)(ft$^2$) (Table VI), and the cobalt

TABLE VI

Heat Transfer from Stacked Cobalt Wafers (or Capsules)

(cobalt diameter: 0.8 inch)

| | | |
|---|---|---|
| Specific activity, Ci/g | 200 | 400 |
| Total activity per inch of column, Ci | 14,500 | 29,000 |
| Total heat generated per inch of column, watts | 230 | 460 |
| Internal heat generated per inch of single column, watts[a] | 60 | 120 |
| Adiabatic heating rate, °C/min | 103 | 205 |
| Maximum heat flux, Btu/(hr)(ft$^2$)[b] | 11,300 | 22,600 |
| Maximum ΔT, °C | | |
| Cobalt to air | 650 | 770 |
| Cobalt to water | 44 | 80 |
| Center of cobalt to surface of cobalt[c] | 4 | 7 |
| Water temperature at which surface boiling occurs, °C | 82 | 47 |

(a) 26% internal absorption.
(b) Based on cylindrical area.
(c) See Figures 9 and 10 for ΔT across gas gap between $^{60}Co$ and capsule.

temperature could exceed 800°C in air. Although this temperature is well below the cobalt melting point of 1495°C, the cobalt will oxidize rapidly. In practice, capsules are cooled during loading and welding by natural convection from finned aluminum collets that can be further cooled by forced air or helium flow. Advantages and problems of handling larger diameter capsules are being evaluated.

Encapsulation increases the cobalt temperature by an amount that depends on the size and nature of any gaps between the cobalt and the capsule wall. However, the temperature drop across reasonable size gaps, even if filled with air, is not excessive (Figure 9).

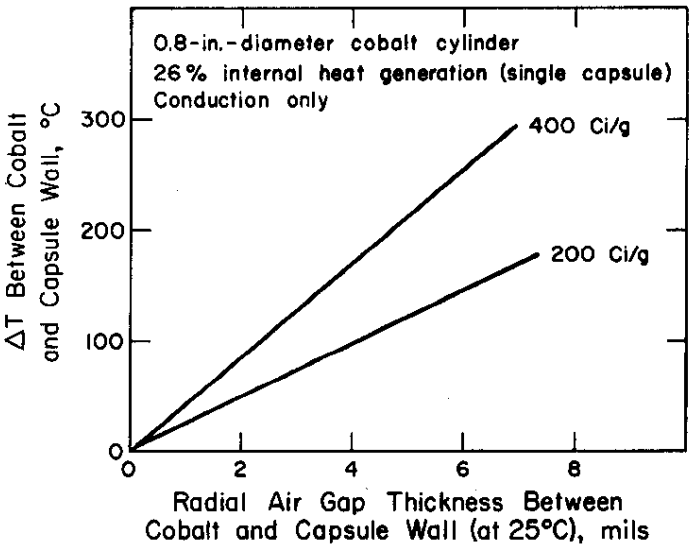



FIG. 9 GAP ΔT IN AIR-FILLED CAPSULES

In large heat sources, the absorption of gamma rays from surrounding capsules effectively increases the heat generation in the center capsules to 80% or more of the total energy generated by the cobalt in that capsule. Figure 10 shows that even with this increased heat load the cobalt temperatures will not be excessively greater than the capsule wall temperature if relatively pure (>90%) helium fills the small radial clearances.

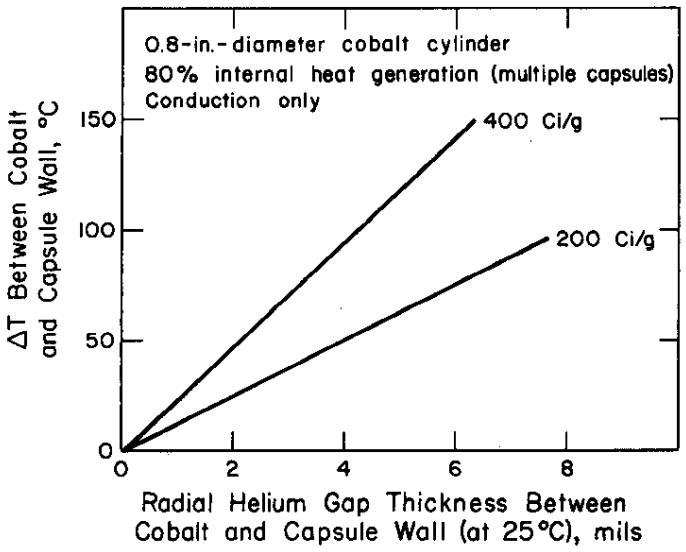



FIG. 10 GAP ΔT IN HELIUM-FILLED CAPSULES

## DESIGN OF TYPICAL $^{60}Co$ CAPSULES

Figure 11 shows experimental capsule designs. Walls 50 mils thick are being used for preliminary evaluations with $^{60}Co$ wafers. The nominal wafer diameter is 0.75 inch. The height of cobalt varies with the specific activity and the desired power, and ranges from 1 to 3 inches. A 60-mil-thick beveled spacer is placed beneath the cobalt to ensure that the wafers are parallel to the caps and to allow the cap to be cut off if desired. A void space equal to 5 to 10% of the cobalt volume is provided to allow allotropic expansion of the cobalt and differential thermal expansion of the metals, and to avoid excessive pressure from released gases.

The inside diameter of the capsule wall is chosen to prevent interference due to differential thermal expansion between the cobalt and the wall. For most oxidation-resistant encapsulating materials, the minimum radial clearance is about 2-1/2 mils at room temperature.

The top closure is by tungsten-inert-gas (TIG) weld, and weld penetration in the thinner container is equal to the wall thickness. A screw closure may be specified to provide additional strength for thicker-walled capsules.

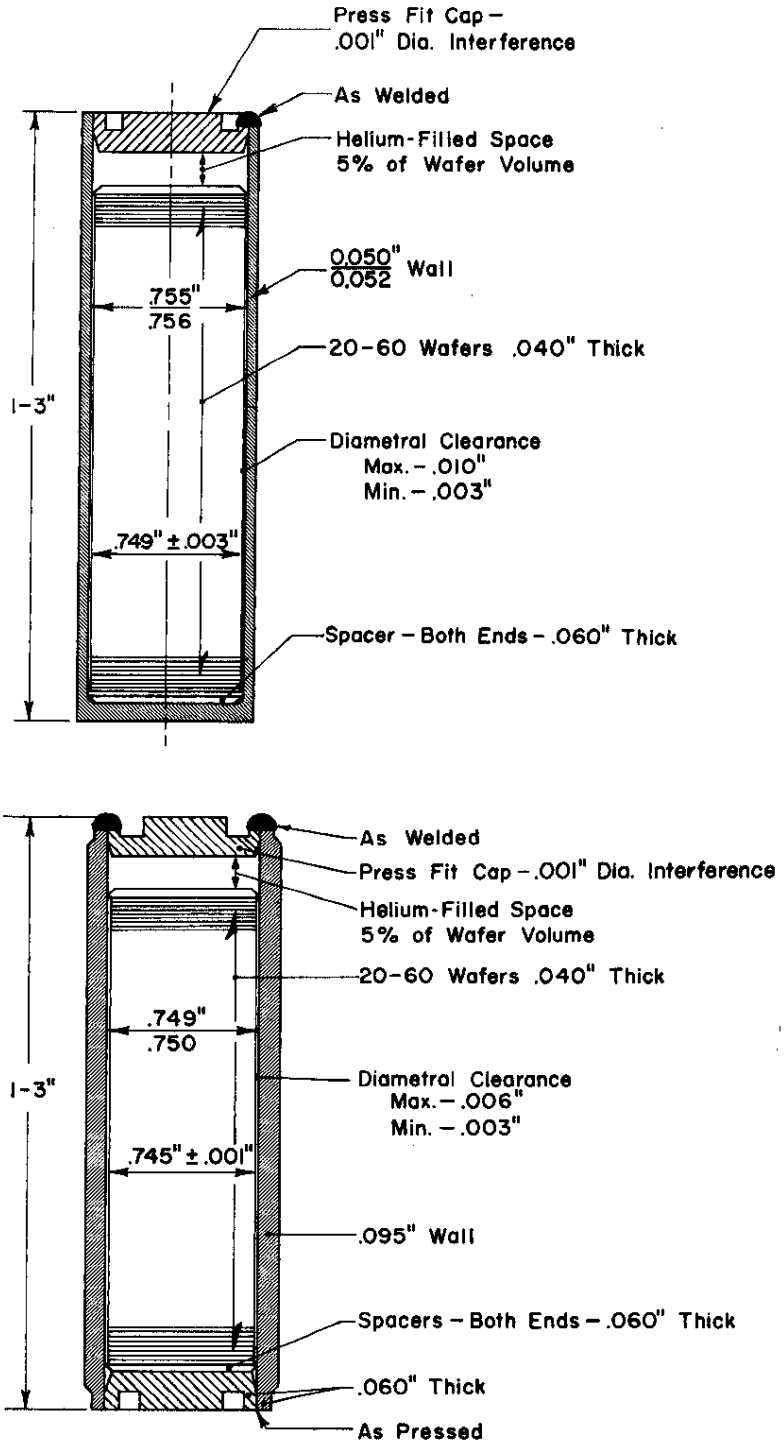



FIG. 11 EXPERIMENTAL CAPSULE DESIGNS

## PROPERTIES OF ENCAPSULATING MATERIALS

Temperature limits for encapsulating materials (Table VII) are being defined by evaluating three basic factors: compatibility with cobalt metal, strength over at least one half-life of $^{60}Co$ at temperature, and resistance to oxidation. In these evaluations, expected operating temperatures and effects of higher temperatures that might be caused by interrupted coolant flow are considered.

### STRENGTH

Two aspects of mechanical behavior of particular concern are: (1) the stress that the capsule wall can withstand without rupturing at elevated temperatures for times up to 5 yr (44,000 hr), and (2) the amount of strain that will be produced by a given stress at these temperatures and times. The mechanical properties that measure these factors are the rupture and creep strengths, respectively. A survey of the literature showed that these properties were the most desirable for TD Nickel, "Haynes"* 152, "Haynes" 25, and "Inconel" 600.

#### Stress Rupture

Variations in rupture strength with temperature for various alloys, obtained from the literature[9] and shown in Figure 12, indicated two types of behavior. Age-hardened alloys, such as "Udimet"** 700, have relatively high rupture strength at temperatures below 800°C, but their strengths decrease sharply with increasing temperature due to agglomeration of the precipitated phases. In contrast, solid-solution-hardened alloys, such as "Inconel", and dispersion-hardened alloys, such as TD Nickel, have lower rupture strengths below 800°C, but their strengths decrease less with increasing temperature. TD Nickel retains significant strength until near its melting point of 1450°C.

Variations in rupture strength with time at 1000°C, obtained by extrapolating stress-rupture curves for selected alloys using the Manson-Haferd parameter,[10] are shown in Figure 13. The two alloys with the highest rupture strength are TD Nickel and "Haynes" 152. The strengths of "Haynes" 25 and "Inconel" 600 are somewhat lower, but decrease little with time. Rupture strengths of "Hastelloy" C and "Hastelloy" X are very low (<700 psi) after only 1000 hr.

* Trademark of Union Carbide Corp.
** Trademark of Kelsey-Hayes Co.

TABLE VII

Compositions and Melting Points of Oxidation-Resistant Alloys

| Alloy | Source | Composition, wt % | | | | | | | | | | | Melting Point, °C |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Ni | Cr | Fe | Co | Ti | Al | Mo | Mn | Si | W | Cb | |
| **Nickel-based** | | | | | | | | | | | | | |
| "Incoloy" 825 | International Nickel Co. | 42 | 21 | 30 | - | 1 | 0.2 | 3 | 1 | 0.5 | - | - | 1360 |
| "Inconel" | " " " | 72 | 15.5 | 8 | - | - | - | - | 1 | - | - | - | 1395 |
| "Inconel" 600 | " " " | 77 | 15.5 | 7 | - | - | - | - | 0.2 | 0.2 | - | | 1370 |
| "Rene" 41 | General Electric Co. | 55 | 19 | - | 11 | 3 | 1.5 | 10 | - | - | - | - | 1315 |
| "Udimet" 700 | Kelsey-Hayes Co. | 53 | 15 | 1 | 18.5 | 3 | 4 | 5 | - | - | - | - | 1220 |
| "Hastelloy" N | Union Carbide Corp. | 69 | 7 | 5 | - | 0.5 | - | 16.5 | 1 | 1 | 0.5 | - | 1326 |
| "Hastelloy" X | " " " | 47 | 22 | 18 | 1.5 | - | - | 9 | 1 | 1 | 1 | - | 1260 |
| "Hastelloy" C | " " " | 55 | 15.5 | 5.5 | 2.5 | - | - | 16 | 1 | 1 | 4 | - | 1270 |
| TD Nickel (2% $ThO_2$) | Du Pont Co. | 98 | - | - | - | - | - | - | - | - | - | - | 1450 |
| TD Nickel Chromium | " " " | 78 | 20 | - | - | - | - | - | - | - | - | - | 1430 |
| "RA-333" | Rolled Alloys, Inc. | 45 | 25 | 18 | 3 | - | - | 3 | 2 | 1 | 3 | - | 1300 |
| **Cobalt-based** | | | | | | | | | | | | | |
| "Haynes" 25 | Union Carbide Corp. | 10 | 20 | 3 | 50 | - | - | - | 1.5 | 1 | 15 | - | 1330 |
| "Haynes" 152 | " " " | 1 | 21 | 1 | 64 | - | - | 4 | - | - | 11 | 2 | 1315 |
| "Haynes Stellite" 21 | " " " | 2.5 | 27.5 | 2 | 61 | - | - | 5.5 | 1 | 1 | - | - | 1350 |

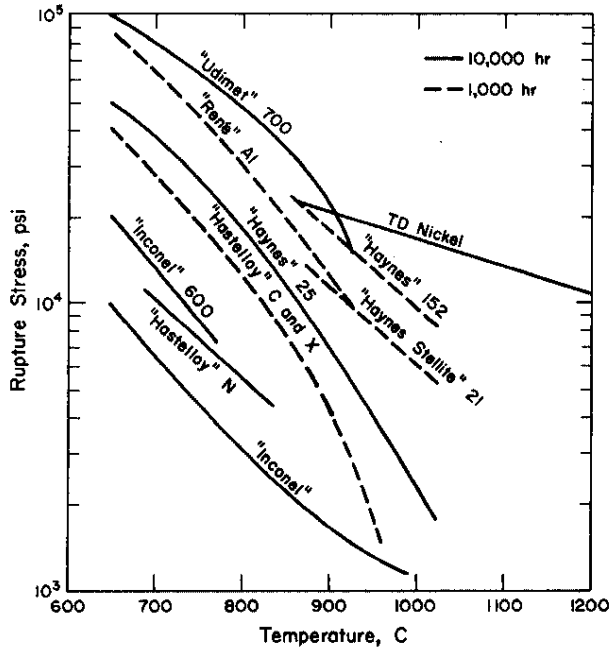



FIG. 12 STRESS RUPTURE PROPERTIES OF CAPSULE ALLOYS
(Data from Reference 9)

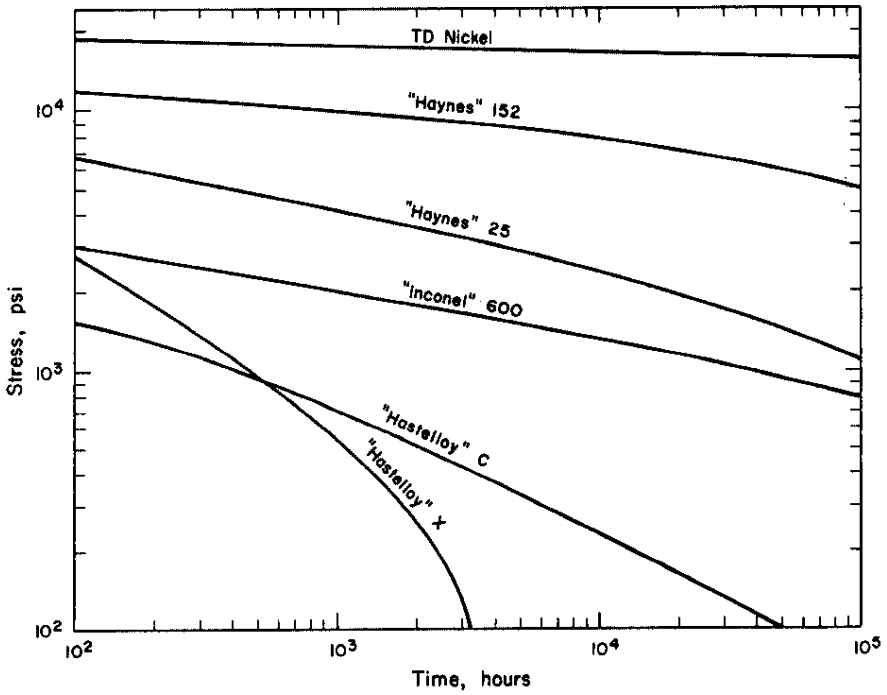



FIG. 13 1000°C STRESS RUPTURE DATA EXTRAPOLATED THROUGH ONE HALF-LIFE OF $^{60}Co$ (Original data from Reference 7)

## Creep

Creep properties (secondary creep) of selected alloys are summarized in Figures 14 and 15.[7] TD Nickel and "Haynes" 25 maintain their creep strengths at temperatures above 800°C. The effect of very long times could not be adequately assessed from existing data. In general, age-hardened alloys should be less stable during creep than solid-solution alloys because of the dissolution of the second-phase particles that are responsible for the strengthening.

Short-term tensile properties of the alloys are shown in Table VIII for reference.[9] Although some of the alloys have relatively high ultimate and yield strengths, these properties are not considered to be as important as long-term rupture and creep properties.

TABLE VIII

Tensile Properties of Encapsulating Materials[9]

| | Short-Term Tensile Properties | | | |
|---|---|---|---|---|
| Alloy | Temp, °C | Ultimate Tensile Strength, kpsi | Yield Strength, kpsi | Elongation, % |
| "Inconel" 600 | 760 | 28 | 17 | 46 |
| | 982 | 8 | 4 | 118 |
| "Hastelloy" C | 815 | 51 | - | 36 |
| | 1037 | 14 | - | 36 |
| "Hastelloy" X | 871 | 36 | 25 | 51 |
| | 1093 | 13 | 8 | 40 |
| | 1204 | 5 | 4 | 31 |
| "Haynes" 25 | 815 | 50 | - | 15 |
| | 982 | 23 | - | 16 |
| | 1260 | 5 | - | 19 |
| "Haynes" 152 | 760 | 94 | 49 | 10 |
| | 1037 | 26 | 17 | - |
| | 1149 | 15 | 10 | - |
| "Haynes Stellite" 21 | 760 | 66 | - | 11 |
| | 982 | 32 | - | 36 |
| TD Nickel | 871 | 23 | 21 | 0.6 |
| | 1037 | 16 | 15 | 0.5 |
| | 1315 | 11 | 10 | 0.4 |

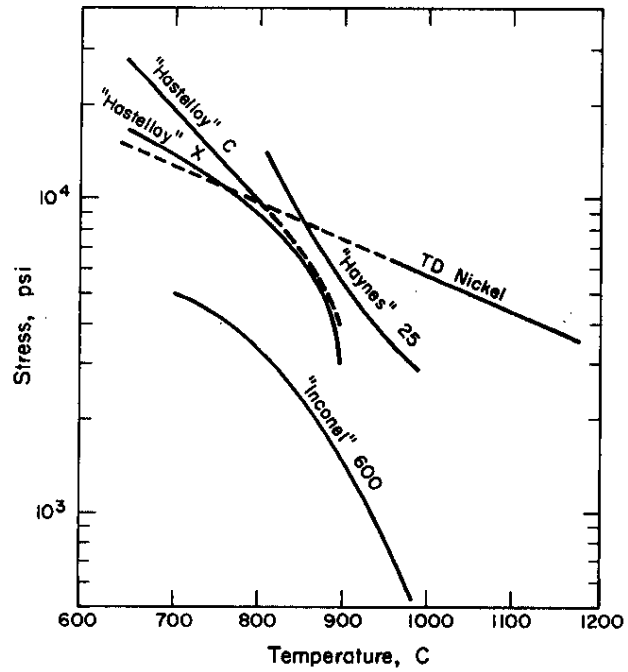



FIG. 14 CONDITIONS FOR SECONDARY CREEP RATE OF $10^{-4}$%/hr
(Data from Reference 7)

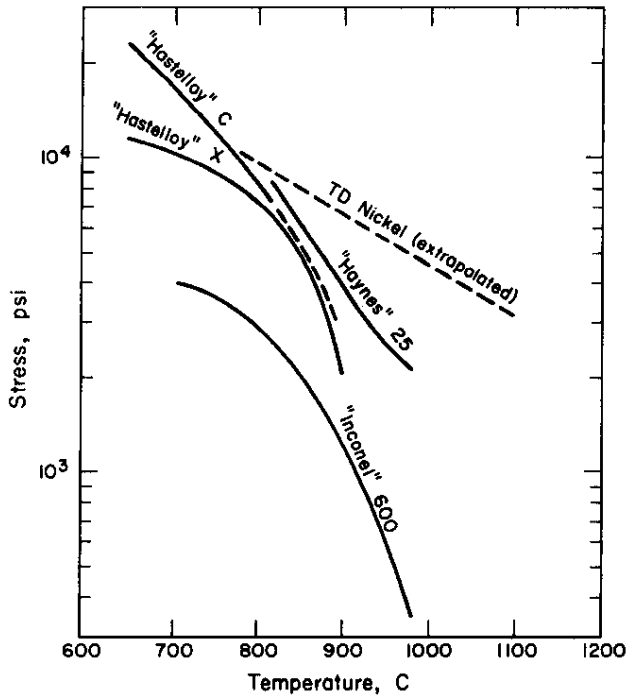



FIG. 15 CONDITIONS FOR SECONDARY CREEP RATE OF $10^{-5}$%/hr
(Data from Reference 7)

## COMPATIBILITY

During operation of a heat source, interdiffusion will occur if the cobalt is in close contact with the capsule wall. Two possible consequences of this diffusion are: (1) the formation of a solid-solution zone or intermetallic compounds may result in a reduction of the wall strength, and (2) $^{60}Co$ may diffuse through the wall. A given encapsulating material is said to be compatible with the cobalt if neither of these consequences is detrimental to operation of the heat source. In recent experiments, "Inconel" 600 and TD Nickel had the best compatibility with cobalt.

### Intermetallic Layer Formation

Preliminary studies of the formation of intermetallic compounds at interfaces were conducted using multilayered diffusion couples of several nickel- and cobalt-based alloys, tantalum, and tungsten. The latter two metals were chosen because their high strengths above 1000°C might be required for capsule integrity. They in turn would have to be clad with an oxidation-resistant alloy for use in air.

Couples of unirradiated cobalt and these materials were annealed for 168 hr at 800, 1000, or 1200°C. The contacting surfaces were ground and clamped together immediately before the anneal. This test permitted identification of any intermetallic layers, measurement of their rate of growth, and characterization of effects of temperature on the structure of the encapsulating alloy.

Two zones were formed at the interfaces between the cobalt and the nickel- and cobalt-based alloys. The first zone, which spanned the original interface, was a continuous solid solution composed of all the elements present, as indicated by electron microprobe analyses (Figure 16). Any second-phase particles originally present in the alloys were dissolved in this zone. The second zone contained voids that formed because diffusion into the cobalt was faster than diffusion into the alloy (the Kirkendall effect).[11] Formation of the voids depended on the temperature. At 800°C, none of the alloys had a void zone. At 1000°C, the nickel-based alloys had voids, but the cobalt-based alloys did not. At 1200°C, some voids were present in all the alloys (Figure 17).

Two intermetallic layers formed at the interfaces between cobalt and tungsten. The Co-W phase diagram indicates that these two compounds are $Co_3W_2$ and CoW. The original interface corresponded to the boundary between these two layers.

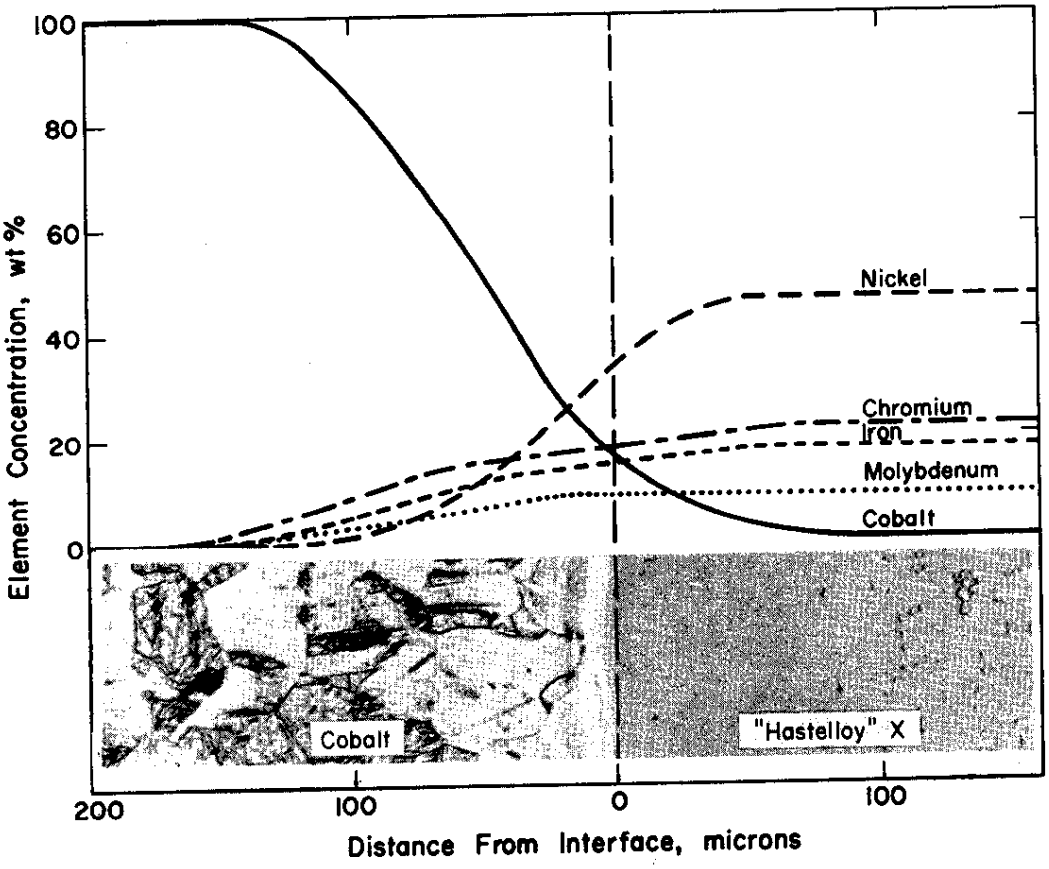



FIG. 16 ELEMENTAL DISTRIBUTION IN COBALT-"HASTELLOY" X DIFFUSION COUPLE
168 hr at 1000°C

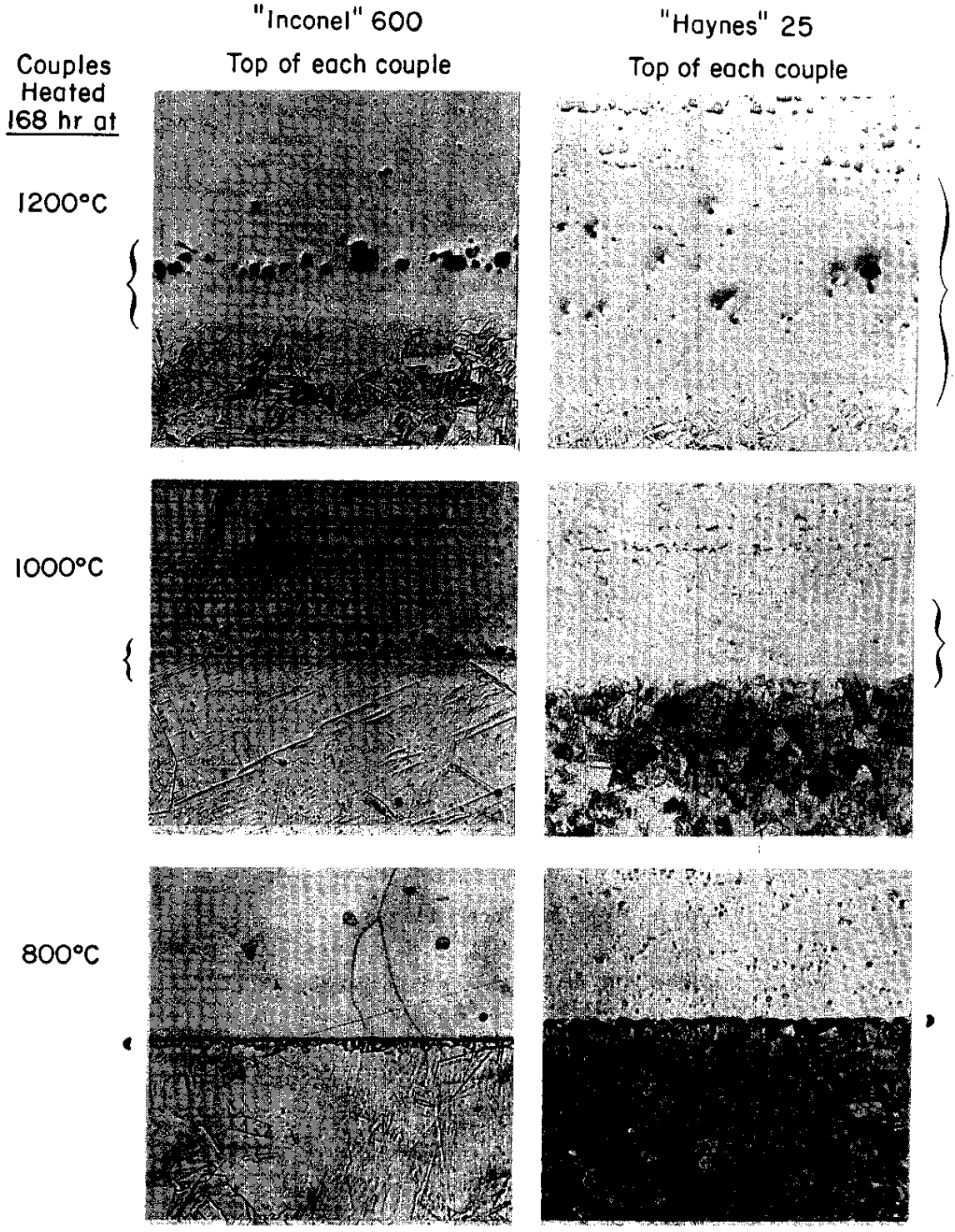



FIG. 17 DIFFUSION-ZONE VOIDS IN ENCAPSULATING MATERIALS, 100X
Cobalt at bottom of each couple. Diffusion zones indicated by braces.

No intermetallic layers were formed at the cobalt-tantalum interface during the anneal at 1000°C, but melting occurred at the interface during the anneal at 1200°C. This melting resulted from the formation of the eutectic mixture at 30 wt % tantalum, and would limit the use of tantalum to heat sources in which the temperature would be below 1200°C.

Growth of intermetallic layers during diffusion usually follows the parabolic relationship $x^2 = kt$, where x is the width of the layer, k is a rate factor that is related to temperature by an Arrhenius-type equation, and t is the time.[12] Expected widths of diffusion zones after one half-life at temperature were calculated from this relationship. Because the extrapolation of one-week diffusion tests to 5-year predictions contains a recognized risk, the extrapolated values given in this report will be confirmed by a continuing program of long-term diffusion studies. The variation in width with temperature (Figure 18) provided an assessment of the relative compatibility of the alloys at a given temperature. "Inconel" 600 and TD Nickel were the most compatible of the nickel- and cobalt-based alloys at all temperatures.

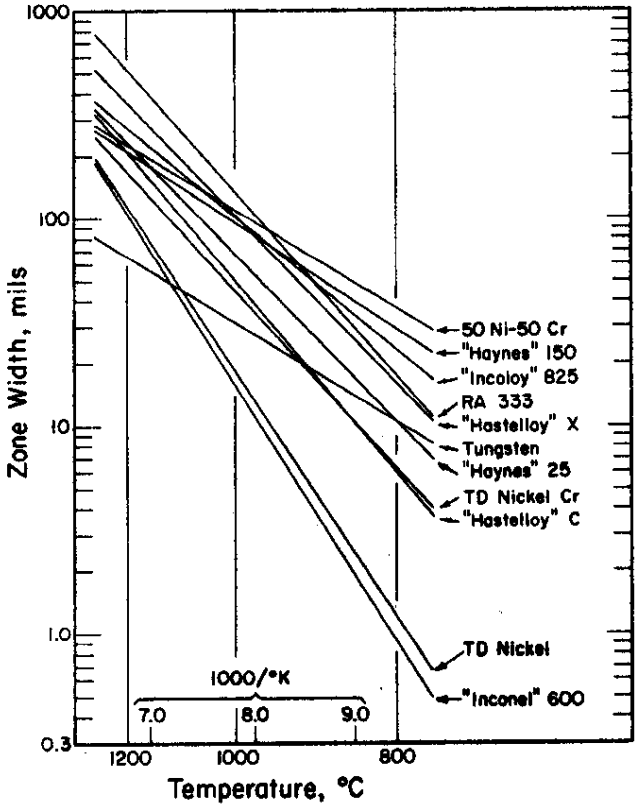



FIG. 18 CALCULATED DIFFUSION ZONE WIDTHS AFTER ONE HALF-LIFE OF $^{60}Co$ (Based on 168 hr test)

Similar compatibility tests will be conducted on other potentially suitable alloys. These alloys include TD Nickel Chromium (high strength and oxidation resistance), Fe-Cr-Al-Y alloys (oxidation resistance), pure nickel (high thermal conductivity), and rhenium and tungsten-rhenium alloys (only feasible refractory metals other than tungsten).

### Diffusion of $^{60}Co$

The rates of volume and grain boundary diffusion of $^{60}Co$ through encapsulating materials are being measured by standard radioactive tracer techniques. Cobalt is electrodeposited onto the sample from a solution containing $^{60}Co$, and the plated samples are annealed for 4, 40, or 100 hr at 800, 1000, or 1200°C. Residual activity is counted after removal of successive layers by electrochemical dissolution of the alloy sample. Preliminary results obtained at 1000°C indicate that the rate of volume diffusion in the nickel alloys is similar to that reported in the literature for pure nickel,[13] but the rate of grain boundary diffusion is faster in some alloys than in others. This increased rate may produce significant amounts of $^{60}Co$ near the surface of a capsule after 5 years at temperature. The practical significance of this diffusion is being investigated further.

## OXIDATION RESISTANCE

The depth of oxidation penetration of prospective encapsulating materials is being evaluated by reviews of published data and confirmatory tests. Oxidation data beyond 1000 hours at high temperature are not available for most materials, and unless the mechanism of oxidation is understood fully, extrapolations based on a parabolic rate law may be erroneous. Data for "Hastelloy" X, "Inconel" 600, and "Inconel" 702 at 1000°C indicate that changes in both the mechanism and rate of oxidation in air occur between 5000 and 7500 hours,[14] and the trend of the data indicates that after 5 years the depth of oxidation penetration may exceed the depth of interaction layers between cobalt and the capsule wall.

Oxidation resistance of potential alloys is being measured in tests at 1000°C in still air for up to 10,000 hours. Data for 1000 hours (Table IX) indicate that TD Nickel Chromium is most resistant to high-temperature oxidation. Similar examinations will be made after exposures to 5000 hr (March 1967) and 10,000 hr (September 1967).

TABLE IX

Oxidation Resistance of Encapsulating Materials

(1000 hr in air at 1000°C)

| Material | Thickness of Oxide Scale, mils | Depth of Intergranular Oxidation, mils | Total |
|---|---|---|---|
| TD Nickel Chromium | <0.1 | None | <0.1 |
| "Inconel" 600 | 1.1 | 0.8 | 1.9 |
| "Hastelloy" C | 0.4 | 1.9 | 2.3 |
| "Hastelloy" X | 0.6 | 1.7 | 2.3 |
| "Incoloy" 825 | 1.1 | 1.9 | 3.0 |
| TD Nickel | 10.8 | None | 10.8 |
| 99.97% Nickel | 11.1 | Complete* | Complete* |

* 60-mil-thick samples

## SELECTION OF ENCAPSULATING MATERIALS

Preliminary comparisons of design criteria and material properties indicate that the most promising alloys are "Inconel" 600, TD Nickel, and TD Nickel Chromium because of their high melting points (1370 to 1430°C), retention of reasonable creep and rupture properties, low rate of interaction with cobalt, and relatively low oxidation rate up to 1 year (Figure 19).

Figures 19a and 19b show comparative wall thicknesses for capsules to resist creep and stress rupture for 5 years. Creep data are based on the maximum wall stress and the assumption that secondary creep continues throughout the 5-year period; this assumption should be valid for the low stresses encountered. At 800 to 900°C, prospective encapsulating materials have sufficient strength to withstand the 60-psig internal pressure with thin walls. Above ~900°C, "Hastelloy" C and "Inconel" 600 may have excessive creep rates.

Comparative depths of diffusion layers calculated at various temperatures after 5 years are shown in Figure 19c. Penetration into the wall is assumed to be 2/3 the total region of interaction given in Figure 18, as indicated by the position of the original interface relative to the boundaries of the diffusion zone. Cobalt-wall interactions are satisfactorily slow at 800 to 900°C. Above ~900°C, diffusion zone widths for "Hastelloy" C and "Haynes" 25 may be excessive.

Oxidation depths after 1 year, derived from 10,000-hour tests[14,15] or by extrapolating 1000-hour data, are shown in Figure 19d. The rate of oxidation at 1000°C is not excessive for materials under consideration, with the possible exception of TD Nickel.

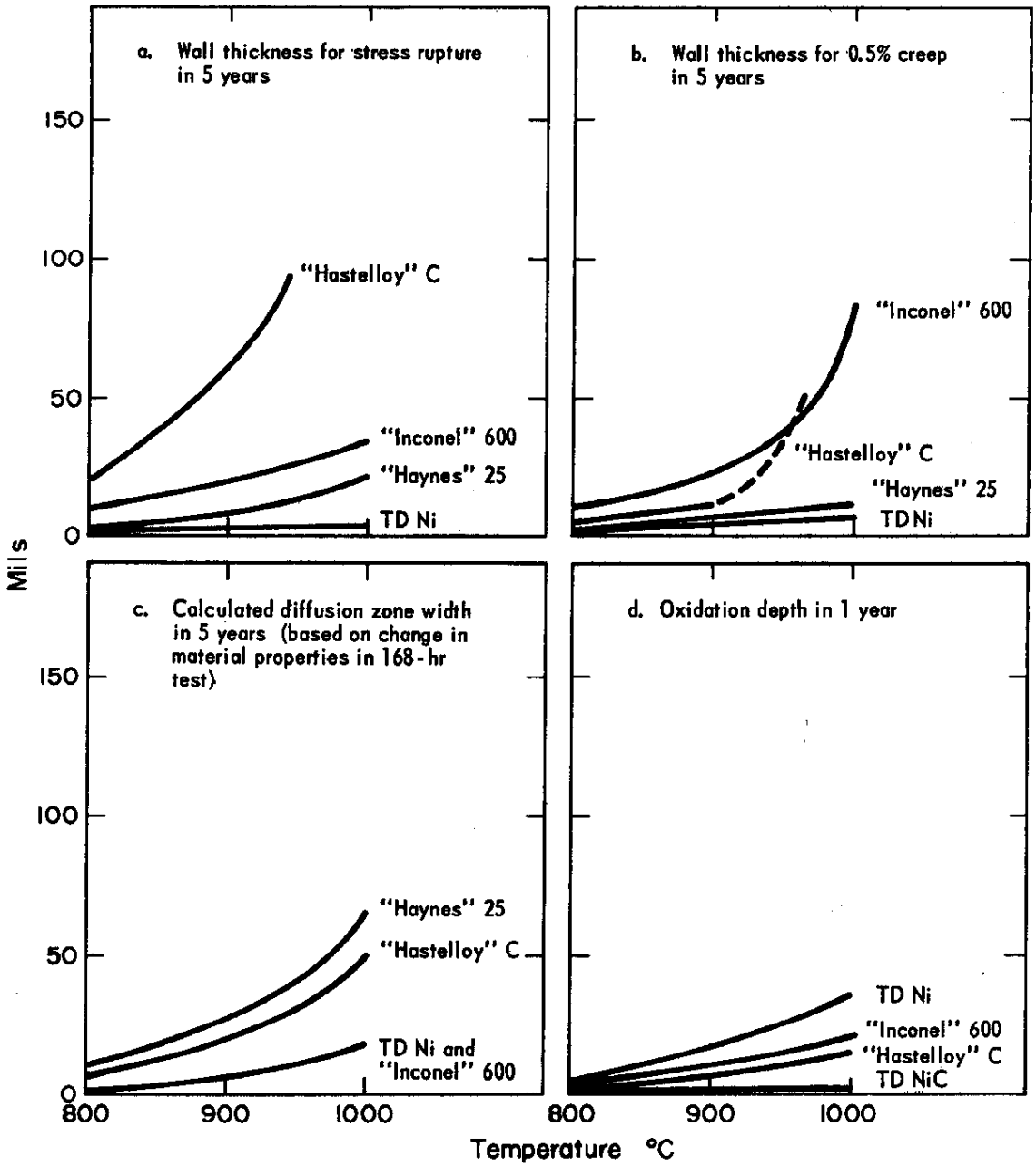



FIG. 19 CALCULATED FACTORS AFFECTING CAPSULE WALL THICKNESS

# CAPSULE FABRICATION AND TESTING

## FABRICATION PROCESSES

Encapsulation processes are being developed to define the problems associated with disassembly of target elements, cleaning of cobalt wafers, and fabrication of capsules. Experimental capsules are being fabricated for engineering and metallurgical tests of performance at typical heat source operating conditions. $^{60}Co$ encapsulation includes the following steps:

1. The aluminum can is dissolved from reactor elements with sodium hydroxide, and residual corrosion products are removed at a cleaning station.

2. The nickel-plated wafers are rinsed, hot-air dried, and gaged to ensure that they will fit into the container.

3. Wafers are individually loaded into a container that has been hot-air dried and placed in a locally shielded loading fixture. The container is evacuated and back-filled with helium several times before the cap is pressed in.

4. The closure weld is made in a helium atmosphere with conventional TIG welding equipment and an automatic welding head.

5. Final inspection steps include measurement of dimensions, surface inspection, helium leak test, and a short heating test.

Welding tests have shown no weld blowouts in circumferential welds when caps are pressed into the container with a 1-mil interference fit. Figure 20 shows a typical weld closure. The requirement for weld penetration is a depth equal to or greater than the 50-mil wall thickness.

Encapsulation equipment installed in the SRL High Level Caves was tested by fabrication of dummy capsules, some of which contained heaters to simulate the temperature rise caused by the $^{60}Co$. Shown in Figures 21 and 22 is equipment for wafer handling and cleaning, wafer measurement, wafer loading, helium backfilling, welding, leak detection, and preliminary heating tests.

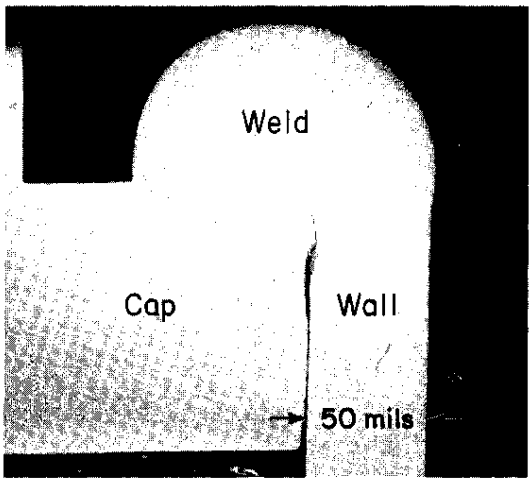
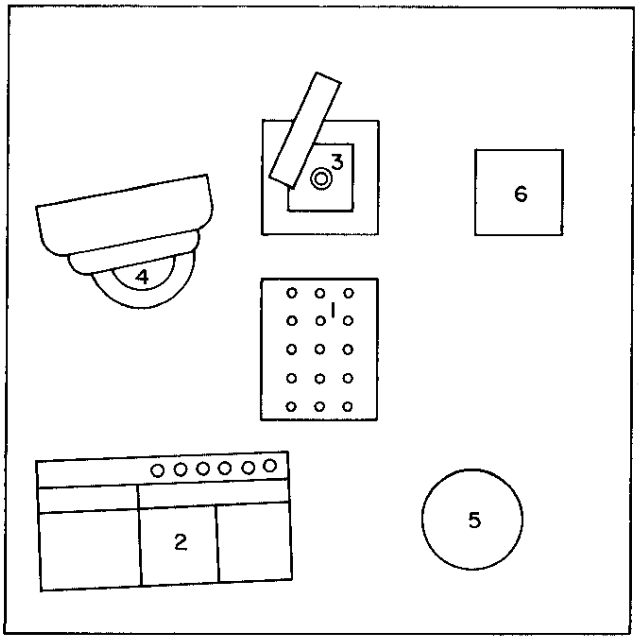
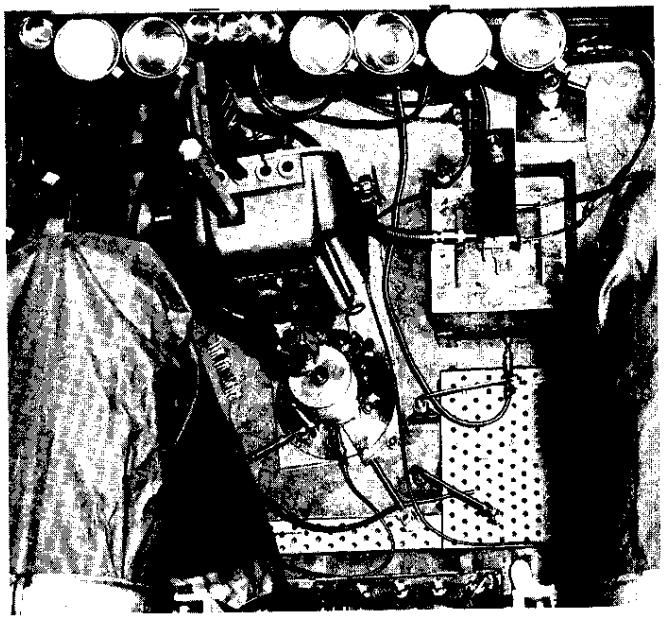



FIG. 20 CROSS SECTION OF CLOSURE WELD

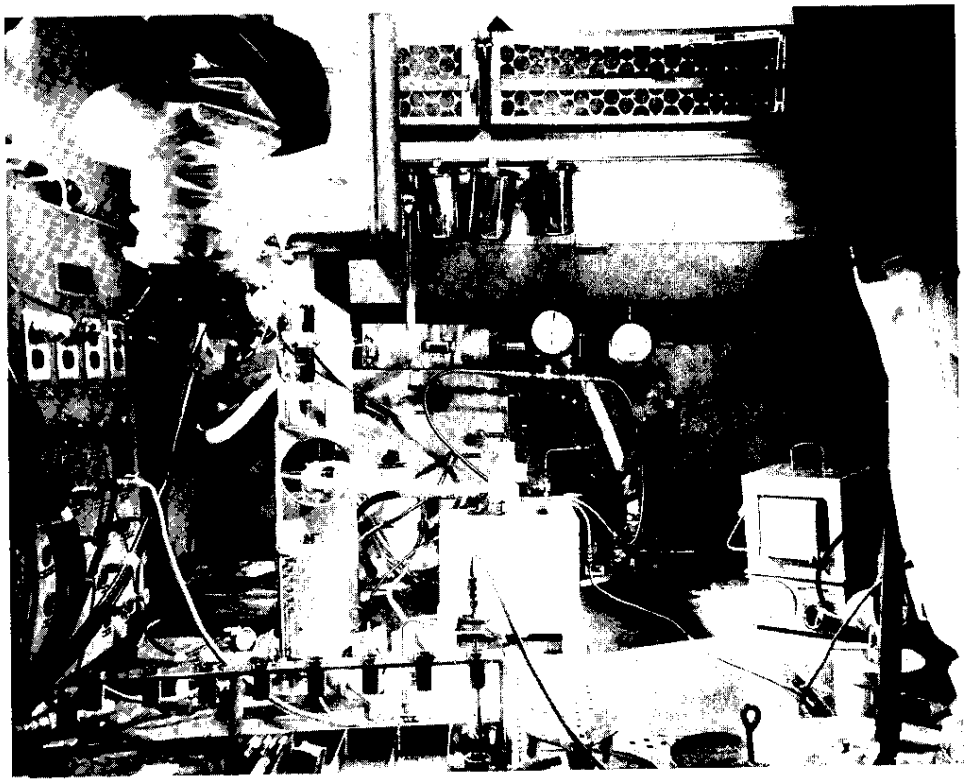



1- Wafer cleaning well
2- Wafer gaging and work tray
3- Capsule loading and helium backfill
4- Welding station
5- Leak detection chamber
6- Oven

FIG. 21 ARRANGEMENT OF $^{60}Co$ ENCAPSULATION EQUIPMENT



Top View



Front View through Cell Window

FIG. 22 $^{60}Co$ ENCAPSULATION EQUIPMENT IN CELL

An experimental air-cooled storage cask (Figure 23), designed to attenuate 99% of the gamma radiation from capsules containing up to 100,000 Ci of $^{60}Co$, was fabricated and tested out-of-cell using a 1500-watt cartridge heater. Maximum capsule temperature was about 550°C. In-cell tests with a 17,000-Ci source (270 watts) produced a capsule surface temperature of ~120°C. Cask heat dissipation tests will continue as higher activity capsules are produced.

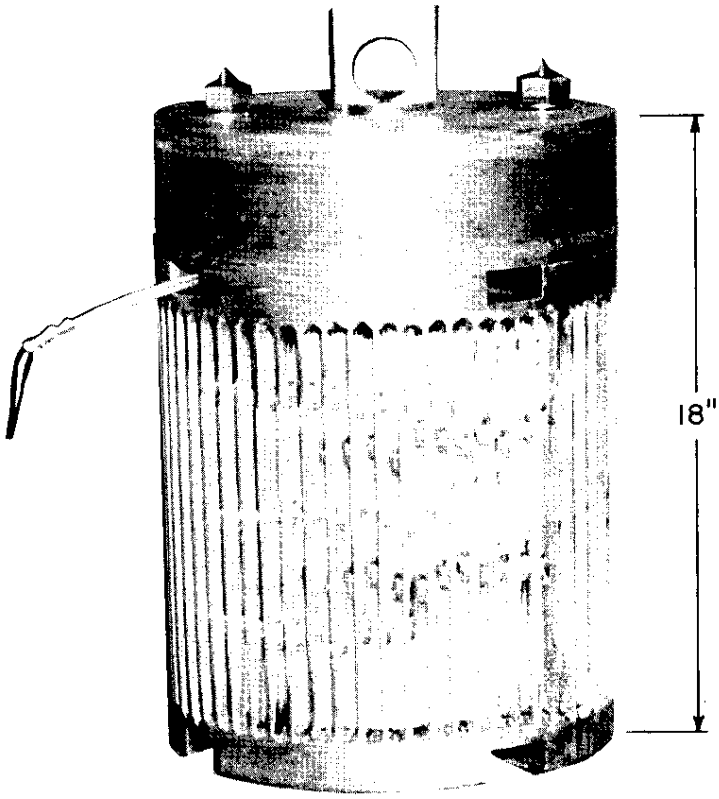



FIG. 23 EXPERIMENTAL CAPSULE STORAGE CASK

## CAPSULE TESTING

Reliability and durability of capsules are being demonstrated by heating capsules filled with cobalt wafers for 1000 to 10,000 hr at temperatures up to 1000°C in still air. These tests provide measurement of the compatibility over longer times than the multi-layered diffusion couples, measurement of oxidation resistance, and evaluation of the encapsulation procedures. Comparison of similar tests of inactive and radioactive capsules will measure the effects of the radiation field on oxidation and compatibility.

**Inactive Capsules**

Eight nonradioactive capsules fabricated with 50-mil-thick walls from "Inconel" 600, "Hastelloy" C, and "Incoloy" 825 have been heated at 1000°C in three separate tests (Table X). In the first and third tests, capsules were intact after 1000 hr; in the second test, under essentially the same conditions as the first, one capsule of each material oxidized catastrophically after 500 hr at temperature due to an interaction with the firebrick on which the capsules rested. In the third test, capsules were heated in contact with "Inconel" 600. In the first two tests, capsules were loaded with unirradiated cobalt wafers in air; in the third test, a helium-filled void space was provided at the top of the capsule and type 304 stainless steel spacers were included at the top and bottom of the stack of cobalt wafers. None of the wafers could be removed after the heating tests because the wafers bonded to the wall and to each other.

TABLE X

Reaction Zone Widths in Heated Nonradioactive Capsules

(1000 hr at 1000°C; initial wall thickness 50 mils)

| | | Maximum Width of Zone, mils | | | | |
|---|---|---|---|---|---|---|
| | | Surface Oxidation | Intergranular Oxidation | | Compositional Change | Unaffected |
| | | | Outside | Inside | | |
| "Inconel" 600 | | | | | | |
| Air-filled | 1 | 1.6 | 3.5 | 7.5 | 5.0 | 37.4 |
| " " | 2[a] | 1.5 | 2.0[b] | 14.0 | 5.0 | 32.5 |
| Helium-filled | 3 | 14.0[c] | 4.0 | 2.5 | 5.0 | 24.5 |
| "Hastelloy" C | | | | | | |
| Air-filled | 1 | 1.2 | 1.2 | 4.0 | 8.0 | 39.6 |
| " " | 2[a] | 50 | - | - | - | - |
| Helium-filled | 3 | 16.0[c] | 2.0 | 4.0 | 12.0 | 16.0 |
| "Incoloy" 825 | | | | | | |
| Air-filled | 1 | 1.2 | 6.0 | 10.0 | 25.0 | 13.8 |
| " " | 2[a] | 50 | - | - | - | - |

(a) Capsules oxidized catastrophically after 500 hr.
(b) On side away from firebrick.
(c) These capsules were cooled to room temperature and reheated to 1000°C five times during test because of furnace control problems.

These heating tests showed that "Inconel" 600 was the best of the three encapsulating materials tested, as indicated by the larger wall thickness that was unaffected by oxidation or changes in alloy composition (Table X). The change in alloy composition due to diffusion affected the largest portion of the wall thickness. This could possibly reduce the strength and perhaps result

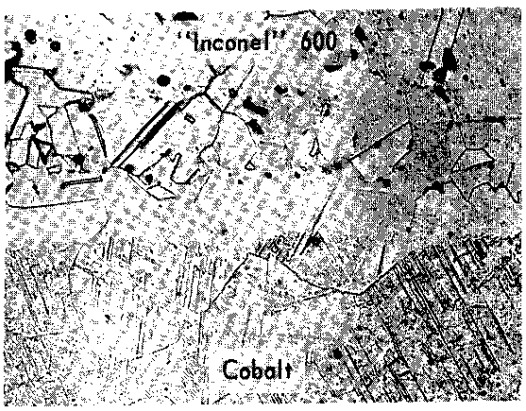
in a change in oxidation resistance when this compositional change extended to the external surface. Resistance of the "Incoloy" 825 to intergranular oxidation was inadequate. The presence of helium in the void space apparently reduced the intergranular oxidation on the internal surface of the "Inconel" 600 to an acceptable value. Typical microstructures from the heated capsules are shown in Figures 24 and 25.

Tests at 1000°C for 1000 hr are being made on similar capsules fabricated from alloys, such as TD Nickel, TD Nickel Chromium, "Hastelloy" X, and "Haynes" 25, that were selected on the basis of good compatibility or resistance to oxidation. Heating of "Inconel" 600 capsules at 850°C for up to 10,000 hr has begun; these are companions to capsules filled with $^{60}Co$ that are being tested similarly.

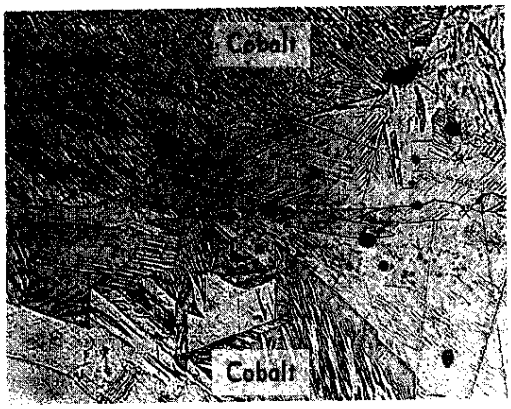
**Radioactive Capsules**

Two capsules, one of "Hastelloy" C containing 9,000 Ci of $^{60}Co$ (140 watts) and one of "Inconel" 600 containing 16,000 Ci (250 watts), have been heated for over 100 hr at 850°C in still air. No leaks were detected before or after the heating tests, and no significant changes occurred in the appearance of the capsules. Metallographic examination of these capsules is in progress.

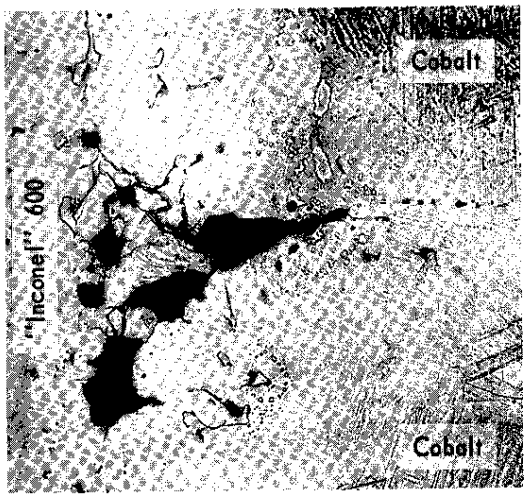
Cobalt in these capsules consisted of wafers 0.75 inch in diameter by 40 mils thick having a specific activity of about 100 Ci/g. Capsule wall thickness was 50 mils. Additional capsules of "Inconel" 600 are being heated at 850°C for 1000, 5000, and 10,000 hr.



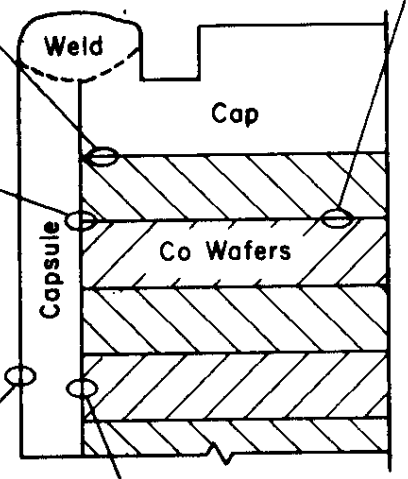

a. Cobalt-to-"Inconel" diffusion bond

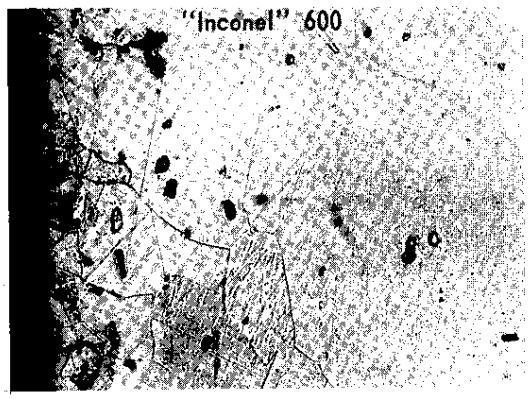



b. Cobalt-to-cobalt bond

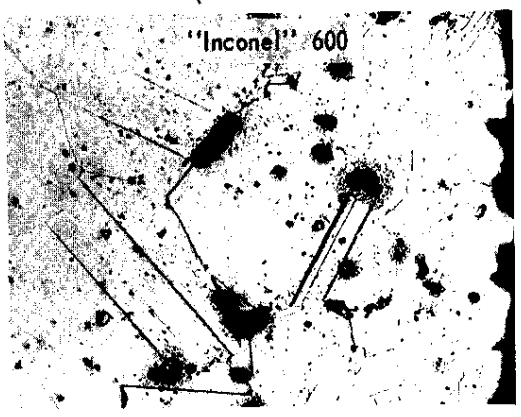



c. Bonding between "Inconel" wall and cobalt wafers



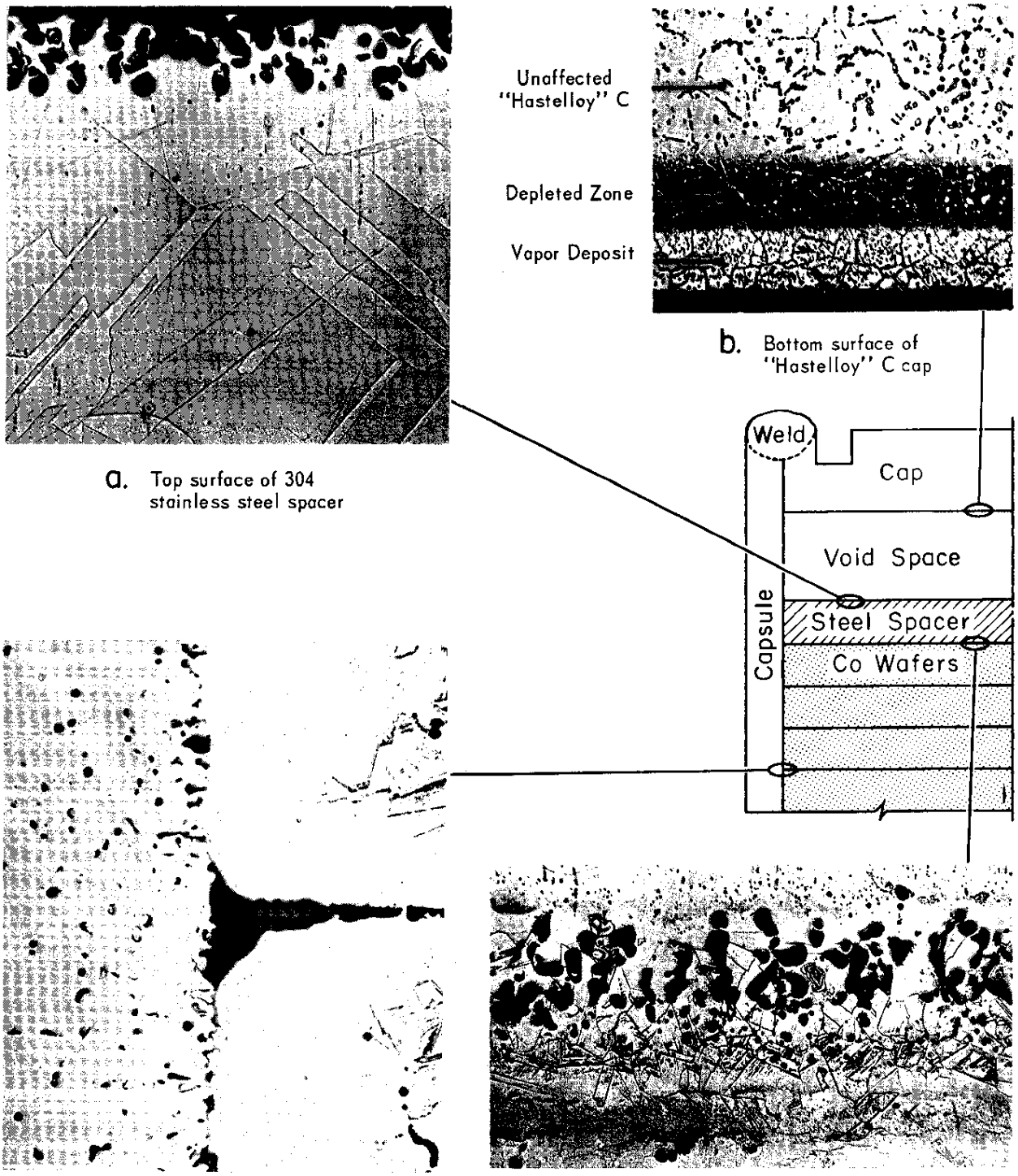




d. Subsurface oxidation at exterior of capsule




e. Subsurface oxidation at interior of capsule

FIG. 24 DIFFUSION AND OXIDATION OF HEATED "INCONEL" 600 CAPSULE
1000 hr at 1000°C




a. Top surface of 304 stainless steel spacer

b. Bottom surface of "Hastelloy" C cap

c. Bonding between capsule and cobalt wafers

d. Bonding between steel spacer and cobalt wafer

FIG. 25 DIFFUSION AND OXIDATION OF HEATED "HASTELLOY" C CAPSULE
1000 hr at 1000°C

## REFERENCES


1. J. W. Joseph, Jr., H. F. Allen, C. L. Angerman, and A. H. Dexter. Radioactive Cobalt for Heat Sources. USAEC Report DP-1012, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1965).

2. W. C. Windley. Properties of $^{60}$Co and Cobalt Metal Fuel Forms. USAEC Report DP-1051 (Rev. 1), E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1966).

3. H. S. Hilborn (compiler). Savannah River Laboratory Isotopic Power and Heat Sources - Quarterly Progress Report - July-September 1966. USAEC Report DP-1088, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1966)(Confidential).

4. H. S. Hilborn (compiler). Savannah River Laboratory Isotopic Power and Heat Sources - Quarterly Progress Report, October-December 1966. USAEC Report DP-1094, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1967).

5. D. T. Goldman. Chart of the Nuclides. Seventh Edition. General Electric Company, Schenectady, N. Y. (1964).

6. E. D. Arnold. Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine, and Space Applications. USAEC Report ORNL-3576, Oak Ridge National Laboratory, Tenn. (1966).

7. F. D. King and C. L. Angerman. "Large Scale Cobalt-60 Heat Sources." Large Scale Production and Application of Radioisotopes. USAEC Report DP-1066, Vol. II, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1966).

8. M. L. Storm, H. Hurwitz, and G. M. Roe. Gamma-Ray Absorption Distribution for Plane, Spherical, and Cylindrical Geometrics. USAEC Report KAPL-783, General Electric Co., Knolls Atomic Power Laboratory, Schenectady, New York (1952)(declassified 1957).

9. "Alloy Digest." Engineering Alloys Digest, Inc., Upper Montclair, N. J. (Data sheets on individual alloys revised periodically to reflect additional data and newly developed alloys.)

10. G. E. Dieter, Jr. Mechanical Metallurgy. McGraw-Hill Book Company, Inc., New York (1961).

11. W. Seith and T. Heumann. Diffusion of Metals: Exchange Reactions. Report AEC-tr-4506 (January 1962).

12. B. Lustman and R. F. Mehl. "Rate of Growth of Intermediate Alloy Layers in Structurally Analogous Systems." Trans AIME 147, 369 (1942).

13. J. Askill. A Bibliography on Tracer Diffusion in Metals. Part I - Self-Diffusion in Pure Metals; Part II - Impurity Diffusion in Pure Metals. USAEC Report ORNL-3795, Oak Ridge National Laboratory, Tenn. (1966).

14. J. S. Brunhouse and G. W. Titus. Evaluation of Long Term Gas Corrosion of Heat Resistant Alloys for Use as Gas-Cooled Reactor Fuel Cladding. USAEC Report IDO-28605, p 34, Aerojet-General Nucleonics, San Ramon, Calif. (1963).

15. High Temperature Materials Program Progress Report No. 55, Part A. USAEC Report GEMP-55-A, General Electric Co., Nuclear Materials and Propulsion Operation, Cincinnati, Ohio (1966).