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AEC RESEARCH AND DEVELOPMENT REPORT

SAVANNAH RIVER LABORATORY ISOTOPIC POWER AND HEAT SOURCES

QUARTERLY PROGRESS REPORT

JANUARY - MARCH 1969

PART I - COBALT-60

DU PONT
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Savannah River Laboratory

Aiken, South Carolina

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ISOTOPIC POWER AND HEAT SOURCES

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PART I - COBALT-60

H. S. Hilborn, Compiler

April 1969

E. I. DU PONT DE NEMOURS & COMPANY
SAVANNAH RIVER LABORATORY
AIKEN, S. C. 29801

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PREFACE

This report is one in a series on the applied aspects of isotopes that are under study at the Savannah River Laboratory (SRL), and that are of interest as isotopic heat source materials. Principal emphasis is on isotopes that are produced by neutron addition, since these are the materials for which the production capabilities of the Savannah River Plant (SRP) reactors and other facilities can be used effectively. Data for other materials will be included if pertinent -- such as the isotopic or chemical composition of fission products that can be recovered from Savannah River process wastes.

These reports are intended to present data that are useful to system designers and also to potential or active user agencies. The reports thus deal with the following subject areas of SRL programs:

1. Properties and reactions of isotopes useful or potentially useful as heat sources.
2. Information on the irradiation and postirradiation processing of these materials, when the information is relevant to their use as heat sources and is not in a sensitive area of production technology.
3. Development of design data directed toward the use of and manufacturing capability for isotopic heat sources.

The report is issued in two parts: Part I includes only information on cobalt; Part II includes information on the other isotopic heat source materials. Both parts contain principally data from work performed during the report period. Previous reports are listed in the Publications section.

SUMMARY

Fabrication procedures are being developed to provide high density wafers of CoO, (Co,Mg)O, and $CoAl_2O_4$ for irradiation tests and evaluation as high melting, oxidation resistant heat source fuels. (p 1)

Evaluations to date indicate that "Haynes" 25 will be the preferred capsule alloy for most uses of ^{60}Co at temperatures to 1000°C. (p 6)

Inactive capsules of "Haynes" 25 increased uniformly in diameter by 0.006 and 0.008 inch in 10,000 hour tests at 1000°C. This increase is consistent with the density change associated with the cobalt phase transformation above 417°C. (p 12)

A preliminary scope of work was completed for a new facility for ^{60}Co capsule testing. (p 15)

Initial tests were run to measure the compatibility of molten cobalt with tungsten. (p 17)

Selection of techniques and equipment for welding refractory alloy capsules for SRL tests was begun. (p 20)

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PROGRAM

The purpose of the Savannah River Laboratory (SRL) program on ^{60}Co is to provide data that will be required for designing, fabricating, and operating ^{60}Co heat sources. Primary emphasis is on selecting materials for encapsulating cobalt fuel forms and establishing temperature limits for long-term operation of capsules. Development of specific heat source concepts is not at present included in the scope of the SRL program.

MATERIALS TECHNOLOGY AND DEVELOPMENT

HIGH-TEMPERATURE FUEL CANDIDATES

Alloys and compounds of cobalt that have higher melting temperatures than cobalt metal are being investigated for use in ^{60}Co heat sources, to provide increased safety and possible fuel forms for thermionic converters. Literature data indicate that cobalt oxide (CoO), cobalt oxide-magnesium oxide solid solutions ((Co,Mg)O), cobalt aluminate ($CoAl_2O_4$), and cobalt-rhenium alloys (Co-Re) would have high melting temperatures and, except for cobalt aluminate, sufficiently high power densities for thermionic converters.(1) The ceramic compounds are the preferred candidates because they are resistant to oxidation and could be fabricated before irradiation. The Co-Re alloys would have to be fabricated after irradiation because of the high cross-section of rhenium and its conversion to osmium, which forms a lower-melting alloy with cobalt.

Fabrication of Oxide Wafers

Fabrication procedures are being developed to provide high-density wafers of CoO, (Co,Mg)O, and $CoAl_2O_4$ for use as refractory, oxidation-resistant heat sources.(1) Wafers of these materials, with densities of 70-82% of the theoretical value, have been fabricated by cold pressing and sintering powders obtained from precipitated hydroxides. A (Co,Mg)O wafer, with a density >95% of theoretical, was produced by hot pressing, an alternative fabrication process. The cold-pressing and sintering technique is being refined to reduce porosity in sintered wafers and to provide wafers, 0.70-inch diameter by 0.15-inch thick, for irradiation tests. Half-scale wafers (0.5-inch diameter x 0.1-inch thick) are used during process development.

Cobalt Oxide

Half-scale CoO wafers were sintered for four hours in air at ~1675°C to 82% of theoretical density. X-ray analysis indicated some Co_3O_4 formed during cooling; this oxidation could be eliminated by an inert gas quench. Powder derived from decomposition of $Co(OH)_2$ at 1000°C was used to produce these wafers. The decomposition temperature will be reduced to about 600°C to obtain a smaller particle size powder that may be sintered to higher densities.

Cobalt Oxide - Magnesium Oxide Solid Solutions

Half-scale wafers of MgO-42 at. % CoO were sintered for two hours in helium at 1600°C to 80% of theoretical density. No reduction of the CoO content of the wafer by preferential vaporization was observed after sintering. Coprecipitation of the mixed hydroxides with NaOH led to more consistent compositional control than that attained with NH_4OH. However, more thorough washing was required to ensure complete removal of sodium from the precipitate.

Production-scale wafers of CoO-MgO solid solution were sintered for four hours in air at 1600°C to 77% of theoretical density. The wafers were generally acceptable for irradiation.

One (Co,Mg)O wafer of 95% density was produced by vacuum hot-pressing for 15 minutes at 1260°C under a pressure of 4000 psi. A platinum-40% rhodium die liner and spacers were used to prevent contact between the powder and graphite die. Some cobalt metal, formed by reduction of CoO with CO, was observed near the pellet edges. Although hot pressing produces high-density wafers, only a few wafers will be made by this technique because of low production rate and cobalt metal formation.

Cobalt Aluminate

Cobalt aluminate ($CoAl_2O_4$) powder was obtained by coprecipitation of $Co(OH)_2$ and $Al(OH)_3$ and subsequent reaction during calcination for two hours in air at 1100°C. X-ray and metallographic analysis indicated only $CoAl_2O_4$ was present. Half-scale wafers of this material were sintered for two hours in helium at 1600°C to about 70% of theoretical density. These low-density wafers were fragile compared to CoO and (Co,Mg)O wafers.

EVALUATION OF ENCAPSULATION MATERIALS FOR IRRADIATED COBALT METAL

The materials evaluation program is designed to select the most promising alloys for encapsulating ^{60}Co, to define the limiting operating conditions of these alloys, and to demonstrate capsule integrity at conditions typical of heat source operation. The kinetics of cobalt-capsule compatibility reactions,(2,3) diffusion of ^{60}Co,(3-7) and oxidation,(5,7,8) are being measured using short-term (<500 hours) laboratory tests. Selection of the most promising alloys is based on extrapolation of these results to the expected service life (1 to 5 years). Limiting operating conditions are defined by the time and temperature dependency of each of the reactions. Published data on mechanical properties such as creep have been evaluated. The predicted behavior of the materials is being verified by 1000-, 5000-, 10,000-, and 50,000-hour (5.7-year) heating tests of experimental capsules, Tables I and II. Tests of companion capsules containing unirradiated or irradiated cobalt measure any effects of the radiation field and the increased nickel content (from radioactive decay of the cobalt) on the performance of the capsule materials.

Additional tests are performed with unirradiated cobalt encapsulated in the most promising materials to provide data for the safety analyses of ^{60}Co heat sources. These tests are designed to evaluate the response of cobalt and capsule materials to environments common to several potential accidents and are restricted to those conditions that are not critically affected by capsule geometry.

TABLE I

SUMMARY OF ^{59}Co CAPSULE HEATING TESTS

Capsule Material	Heating		Wall, mils	No. of Capsules	Approx. Starting Date	Approx. Completion Date	Remarks
	Time, hr	Temp, °C					
"Inconel" 600	1,000	850	50	1	12-66	2-67	Capsule intact
	5,000	850	50	1	12-66	7-67	Capsule intact
	10,000	850	50	1	12-66	1-68	Capsule intact
	10,000	850	95	1	7-67	9-68	Capsule intact
	50,000	850	95	1	7-67	3-73	
	→ 1,000	900	95	1	11-68	12-68	Capsule intact
	5,000	900	95	1	11-68	6-69	
	→ 5,000(e)	900	95	1	3-69	10-69	
	10,000	900	95	1	11-68	1-70	
	10,000	900	95	1	11-68	1-70	
	→ 10,000(e)	900	95	1	3-69	5-70	
	50,000	900	95	1	11-68	7-74	
	1,000	1,000	50	4	8-66	10-66	3 capsules intact; 1 capsule oxidized(b)
	5,000	1,000	50	1	4-67	11-67	Capsule intact
	1,000(a)	1,000	95	2	7-67	9-67	No severe oxidation of Co
	1,000(d)	1,000	95	1	2-68	4-68	No oxidation of Co or capsule
	5,000(d)	1,000	95	1	2-68	9-68	No oxidation of Co or capsule
	5,000	1,000	95	1	8-67	2-68	Capsule intact
	10,000	1,000	95	1	8-67	10-68	Capsule intact
	50,000	1,000	95	1	10-67	6-73	
	10,000(d)	1,000	95	1	11-68	1-70	
"Hastelloy" C	1,000	1,000	50	4	8-66	10-66	3 capsules intact; 1 capsule oxidized(b)
	5,000	1,000	95	1	10-67	5-68	Capsule intact
	→ 10,000	1,000	95	1	10-67	12-68	Capsule intact
	→ 50,000	1,000	95	1	10-67	6-73	
	10,000+	1,000	95	1	5-68	7-69+	
TD Nickel	1,000	850(c)	95	1	10-67	12-67	Capsule intact
	5,000	850	95	1	10-67	5-68	Capsule intact
	→ 10,000	850	95	1	10-67	12-68	Capsule intact
	→ 50,000	850	95	1	10-67	6-73	
	1,000	1,000	50	1	12-66	2-67	Capsule intact
	1,000(a)	1,000	95	2	10-67	12-67	No severe oxidation of Co
TD Nickel Chromium	1,000(a)	1,000	95	2	10-67	12-67	Co near pinhole oxidized
	1,000	1,000	95	1	10-67	12-67	Capsule intact
	5,000	1,000	95	1	10-67	5-68	Capsule intact
	→ 10,000	1,000	95	1	10-67	12-68	Capsule intact
	→ 50,000	1,000	95	1	10-67	6-73	
"Haynes" 25	10,000	850	95	1	11-68	1-70	
	1,000	1,000	95	1	10-67	12-67	Capsule intact
	5,000	1,000	95	1	10-67	5-68	Capsule intact
	→ 5,000	1,000	95	1	5-68	12-68	Capsule intact
	→ 10,000	1,000	95	1	10-67	12-68	Capsule intact
	→ 50,000	1,000	95	1	10-67	6-73	
	10,000+	1,000	95	1	5-68	7-69+	
"Hastelloy" X	1,000	1,000	50	1	4-67	6-67	Capsule intact
	5,000	1,000	50	1	4-67	11-67	Capsule intact
	5,000	1,000	95	2	2-68	9-68	Capsules intact
	10,000	1,000	95	1	2-68	4-69	
	10,000+	1,000	95	1	2-68	4-69+	
	10,000+	1,000	95	2	5-68	7-69+	

(a) Two capsules, one not welded and one with drilled hole in wall, to test effects of capsule defects.
(b) Capsules reacted with fire-brick. See DP-1094, "SRL Isotopic Power and Heat·Sources - Quarterly Progress Report," October-December 1966.
(c) Tests of TD Nickel at 850°C in flowing argon.
(d) Internal atmosphere air instead of helium.
(e) Caustic residue on wafers.
→ New information reported.

TABLE II

SUMMARY OF ^{60}Co CAPSULE HEATING TESTS

Capsule Material	Heating Time, hr	Heating Temp, °C	Wall, mils	No. of Capsules	Activity Spec. Ci/g	Activity Total, Ci	Approx. Starting Date	Approx. Completion Date	Remarks
"Inconel" 600	130	850[a]	50	1	120	16,000	2-67	2-67	Swelled due to overheating
	1,000	~900	50	1	100	5,000	4-67	6-67	Capsule intact
	5,000	~900	50	1	150[b]	15,000	4-67	10-67	Capsule intact
	10,000	~900	50	1	150[b]	15,000	4-67	6-68	Increased Co/capsule reaction
	10,000	~900	50	1	150[b]	9,000	5-67	10-68	Increased Co/capsule reaction
	10,000	900	95	1	255[c]	36,500	2-68	8-69	
	10,000+	900	95	1	288[c]	13,700	7-68	8-69+	
	50,000	900	95	1	282[c]	13,400	7-68	3-74	
	5,000	1,000	95	1	295[c]	14,000	9-68	4-69	
	10,000	1,000	95	1	288[c]	13,700	9-68	11-69	
	10,000+	1,000	95	1	263[c]	12,500	9-68	11-69+	
	50,000	1,000	95	1	255[c]	12,100	9-68	5-74	
	10,000	850	95	1	(d)	-	9-68	11-69	
"Hastelloy" C	100	850	50	1	120	9,000	1-67	1-67	Capsule intact
	10,000	900	95	1	276[c]	13,100	7-68	8-69	
	10,000	1,000	95	1	282[c]	13,400	9-68	11-69	
	50,000	1,000	95	1	270[c]	12,800	9-68	5-74	
"Haynes" 25	5,000	1,000	95	1	263[c]	12,500	9-68	4-69	
	10,000	1,000	95	1	288[c]	13,700	9-68	11-69	
	10,000+	1,000	95	1	282[c]	13,400	9-68	11-69+	
	50,000	1,000	95	1	295[c]	14,000	9-68	5-74	
	10,000	850	95	1	(d)	-	9-68	11-69	
"Hastelloy" X	5,000	1,000	95	1	250[c]	11,900	9-68	4-69	
	10,000	1,000	95	1	263[c]	12,500	9-68	11-69	
	10,000+	1,000	95	1	263[c]	12,500	9-68	11-69+	
	50,000	1,000	95	1	301[c]	14,300	9-68	5-74	

(a) Excursion to >1100°C for 3-6 hr.
(b) Activity as of 2-67.
(c) Activity as of 6-68.
(d) Capsule contains ^{59}Co but is being heated along with ^{60}Co capsules.

Selection of Materials

Evaluations to date indicate that "Haynes" 25 is the best capsule alloy for most applications of ^{60}Co metal at temperatures up to 1000°C. "Inconel" 600, "Hastelloy" X, and "Hastelloy" C are also promising capsule materials. Selection of the best alloy for any particular application will be based on the final data from continuing tests of ^{60}Co capsules, Table II. Properties of the four alloys are compared in Tables III and IV and in Figure 1.

"Haynes" 25 appears to be suitable for more applications than the other alloys because of high strength, compatibility with cobalt, and resistance to diffusion of ^{60}Co. "Inconel" 600 is considered because of high melting temperature and compatibility with cobalt, but has marginal resistance to diffusion of cobalt and marginal rupture stress at 1000°C. "Hastelloy" X is considered because of oxidation resistance, but has marginal melting temperature and rupture stress at 1000°C. Each of the four alloys is expected to be suitable for long-term operation at 900°C.

TABLE III

MELTING TEMPERATURES OF ALLOYS

	Minimum Melting Temp, °C
"Haynes" 25	1330
"Inconel" 600	1370
"Hastelloy" X	1260
"Hastelloy" C	1270
Cobalt-nickel alloy representing high-activity ^{60}Co heat source	1485

TABLE IV

CALCULATED DIFFUSION OF ^{60}Co IN ALLOYS AT 1000°C

Data from DP-1155-I

	Depth at which ^{60}Co Concentration is ~1 ppm, mils	
	1 yr	5 yr
"Haynes" 25	37	67
"Inconel" 600	~100	>100
"Hastelloy" X	35	77
"Hastelloy" C	45	76

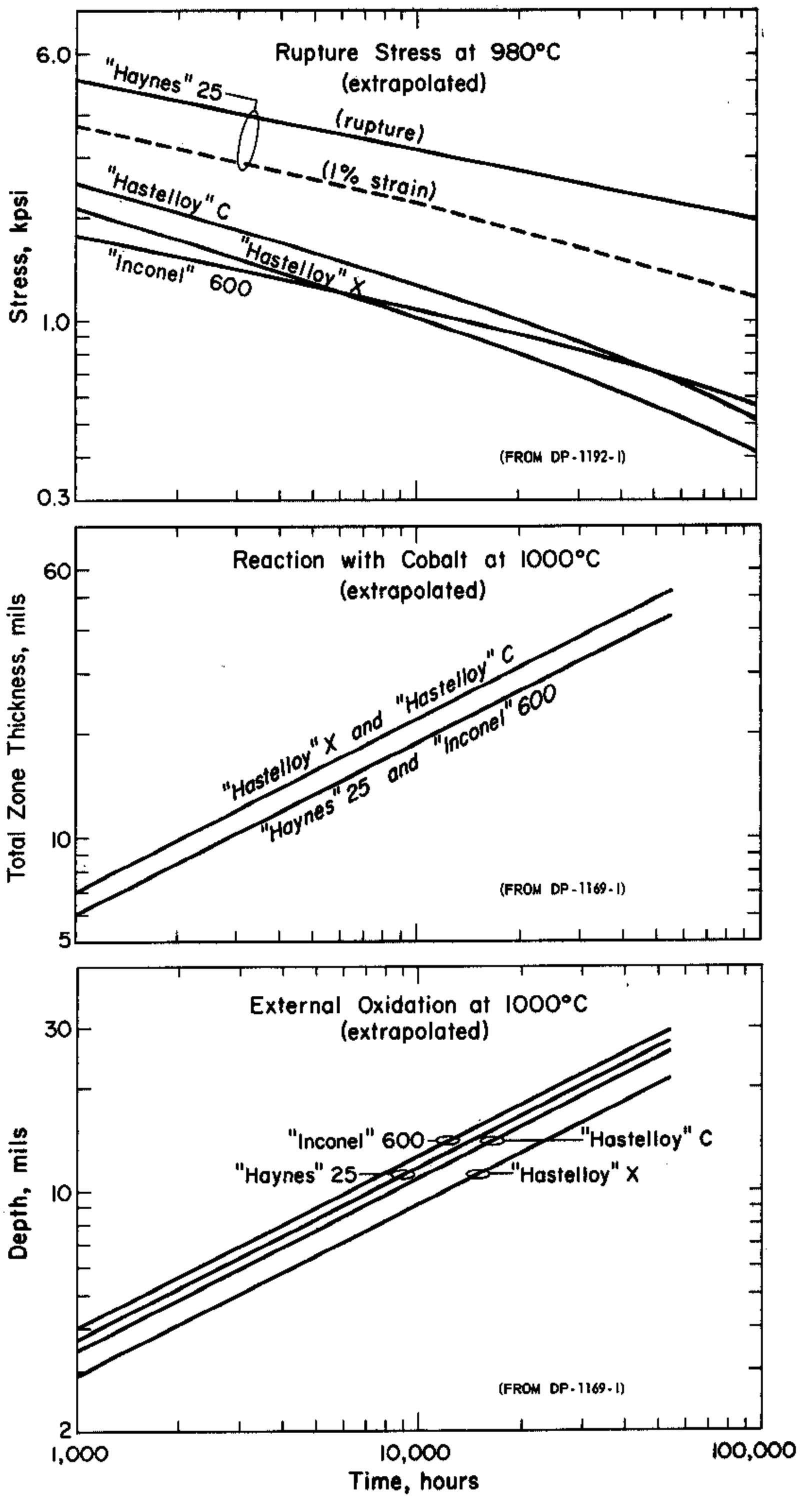

FIG. 1 PROPERTIES OF CAPSULE MATERIALS

Diffusion of ^{60}Co

Measurements were started on the depth of penetration of ^{60}Co in "Haynes"* 25 and "Hastelloy"* C after 5000 hours and after 10,000 hours at 1000°C. The samples are spacers that were plated on one face with ^{60}Co and heated in capsules as part of the ^{59}Co capsule tests. The measured penetrations will be compared with those predicted from 100-hour tests at 1000°C.

Correlation of diffusion coefficients (measured at 800 to 1200°C for up to 100 hours) with time, temperature, alloy composition, and grain size was deferred due to emphasis on preliminary design of a capsule testing facility in the Isotope Process Development Laboratory (IPDL).

Mechanical Properties

Analysis of expected stresses in ^{60}Co heat source capsules indicates that the four best alloys ("Haynes" 25, "Inconel"** 600, "Hastelloy" X, and "Hastelloy" C) will perform satisfactorily at 1000°C for about five years; "Haynes" 25 is best where creep resistance is a prime consideration, although it may increase slightly in volume because of a phase change (see next section). No creep rupture is predicted for the ^{60}Co capsules in the current test, Table II. Some swelling is predicted for capsules of "Hastelloy" X after about two years, and possibly for "Hastelloy" C and "Inconel" 600.

Stresses in ^{60}Co capsules remain low during normal operation because no gas is generated. However, the alloys also have low rupture strength at predicted heat source temperatures.

Wall stresses were calculated on the basis of predicted thinning of the wall by reaction with cobalt and with air, Figure 2. About 80% of the total reaction with cobalt, Figure 1, is assumed to occur in the original capsule wall and the reaction zone is assumed to have zero strength. The latter assumption is probably conservative because the reaction zone has been shown to have high strength after 1000 hours of diffusion; however, this strength may decrease markedly at longer exposures because of void formation. Ring sections from aged capsules will be tested for residual strength.

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* Trademark of Union Carbide Corp.

** Trademark of International Nickel Co.

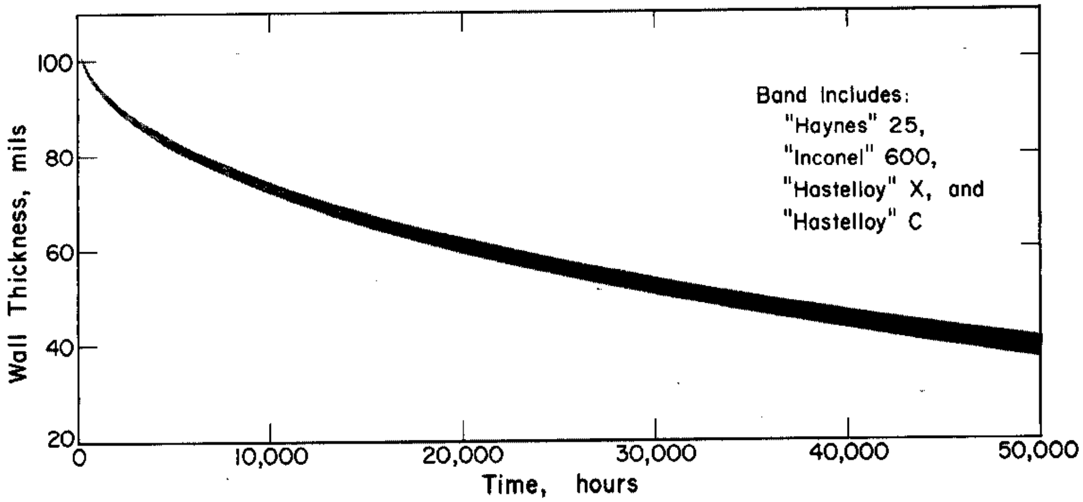

FIG. 2 THINNING OF CAPSULE WALL AT 1000°C

Pressure in the capsule is assumed to be 100 psia at 1000°C.[2] This is probably conservative because the capsule is assumed to be loaded at atmospheric pressure and low temperature without outgassing; the capsule could be outgassed and the fill gas partially evacuated.

The stress in a capsule wall is compared to the extrapolated rupture stress at 980°C (1800°F) for each of the four selected alloys in Figure 3; for "Haynes" 25 the extrapolated creep curve (1% strain) is included. The rupture and creep curves were obtained by time extrapolation of published data by the Manson-Haford time-temperature parameter.[9,10] The "Hastelloy" C data were also temperature-extrapolated, above 870°C (1600°F).

Except for "Haynes" 25, the capsule stress curves cross the stress rupture curves between 30,000 and 40,000 hours. An approximate method was used to estimate the rupture time on the basis of percent of life. Rupture is assumed to be probable when

$$\sum t/t_f = 1.0$$

where

t = time at specified stress

t_f = time to rupture at specified stress

The rupture times so estimated are indicated in Figure 3. They are:

	Rupture Time	
	hr	yr
"Haynes" 25	>50,000	>5.7
"Inconel" 600	46,000	5.2
"Hastelloy" X	38,000	4.3
"Hastelloy" C	45,000	5.1

Because of the foregoing conservative bases for the calculations, no ruptures are expected in the 0.095-inch-wall capsules now being tested at 1000°C. Some swelling is expected in "Hastelloy" X, and may also occur in "Inconel" 600 and "Hastelloy" C after ~20,000 hours (2.3 years).

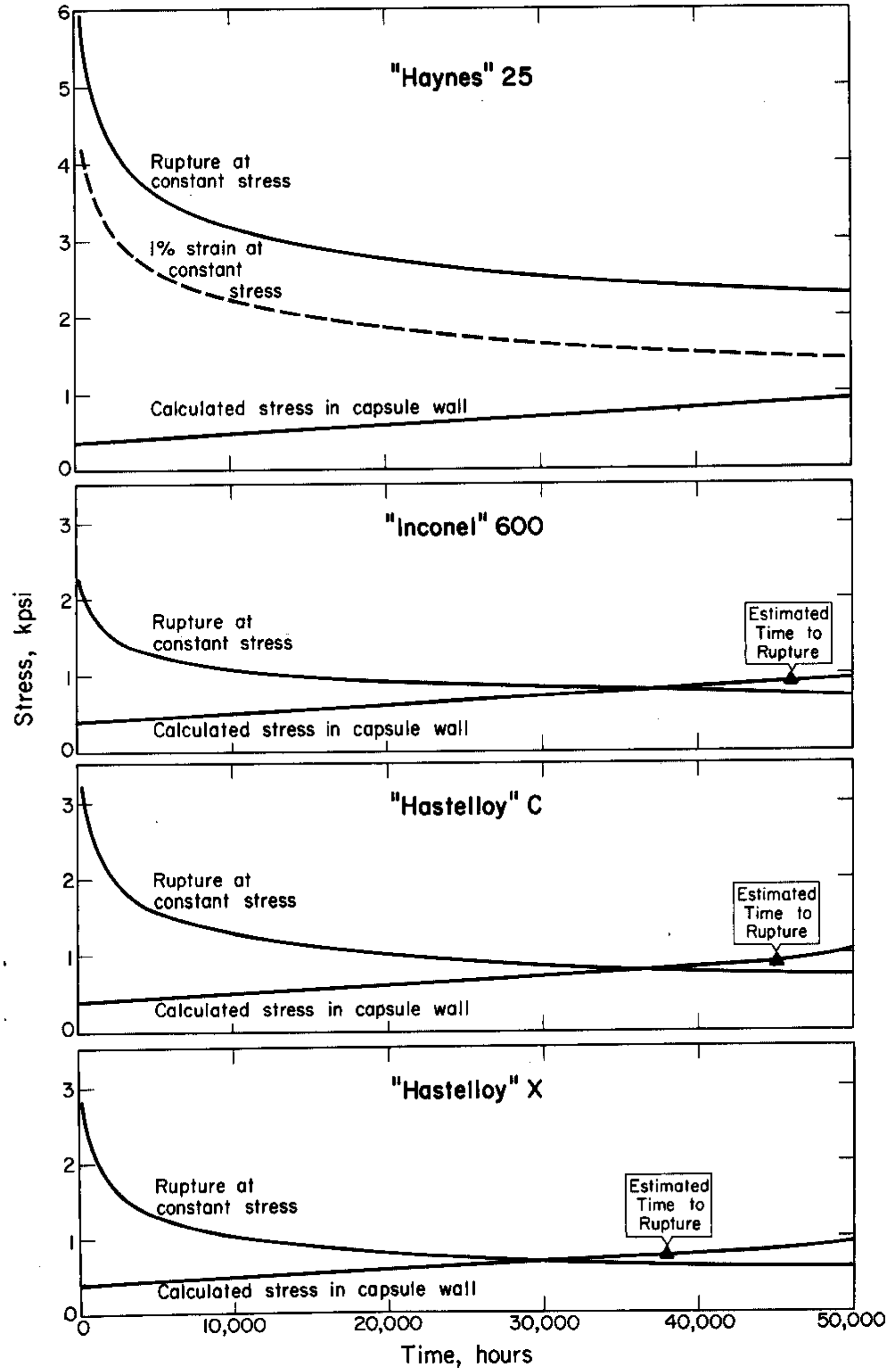

FIG. 3 CAPSULE STRESS AT 980°C

Initial Wall = 0.100 inch – I.D. = 0.75 inch – Internal Pressure = 100 psia

CAPSULE FABRICATION AND TESTING

Heating Tests of Capsules Containing Unirradiated Cobalt

Long-Term Tests

Satisfactory 10,000-hours durability was demonstrated in air at 1000°C with duplicate capsules of "Inconel" 600, "Hastelloy" C, "Haynes" 25, and TD Nickel Chromium* and in argon at 850°C with capsules of TD Nickel*. Since destructive examination of one capsule of each material revealed satisfactory performance, the companion capsule was returned to the furnace for additional heating up to 50,000 hours. These tests are part of the continuing program to demonstrate structural integrity of cobalt heat sources for >10,000 hours at typical temperatures of 850 to 1000°C, Table I.

Integrity was maintained in all capsules and diameter increases were 0.002 inch or less, except for the two "Haynes" 25 capsules in which increases of 0.006 and 0.008 inch were measured. These increases occurred uniformly over the 1.3-inch capsule length (at ends over the end-caps as well as in the center over the cobalt) indicating that the density of the "Haynes" 25 decreased. Creep resulting from a high internal pressure should have caused increases only in the center. "Haynes" 25 is more creep resistant than the other capsule materials.(1)

The decrease in density may be associated with the phase transformation in the cobalt matrix of the "Haynes" 25, a 50Co-20Cr-15W-10Ni-3Fe alloy. The density of pure cobalt decreases ~0.6% when it transforms from a hexagonal-close-packed (hcp) crystal structure to a face-centered-cubic (fcc) structure on heating above 417°C. Additions of 20 wt % Cr and 15 wt % W raise the transformation temperature to 900°C. The phase transformation in pure cobalt is sluggish and strongly influenced by prior cold working. The density and crystal structure will be determined on a sample from this 10,000-hour capsule and compared with similar measurements on samples from a 5000-hour capsule and from as-received material.

Destructive examinations of one capsule of each material showed that the depths affected by the cobalt-capsule compatibility reaction and by oxidation of the exterior capsule walls agree in general with previous capsule and screening tests. For all materials the reaction zone thicknesses are proportional to the square root of the heating time, Figure 4. Data for "Hastelloy" X through 5000 hours at 1000°C and for "Inconel" 600 through 10,000 hours at 1000°C are also shown in Figure 4 for comparison. Additional tests with "Hastelloy" X for 10,000 hours or more are in progress, Table I.

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* Product of Fansteel Metallurgical Corp.

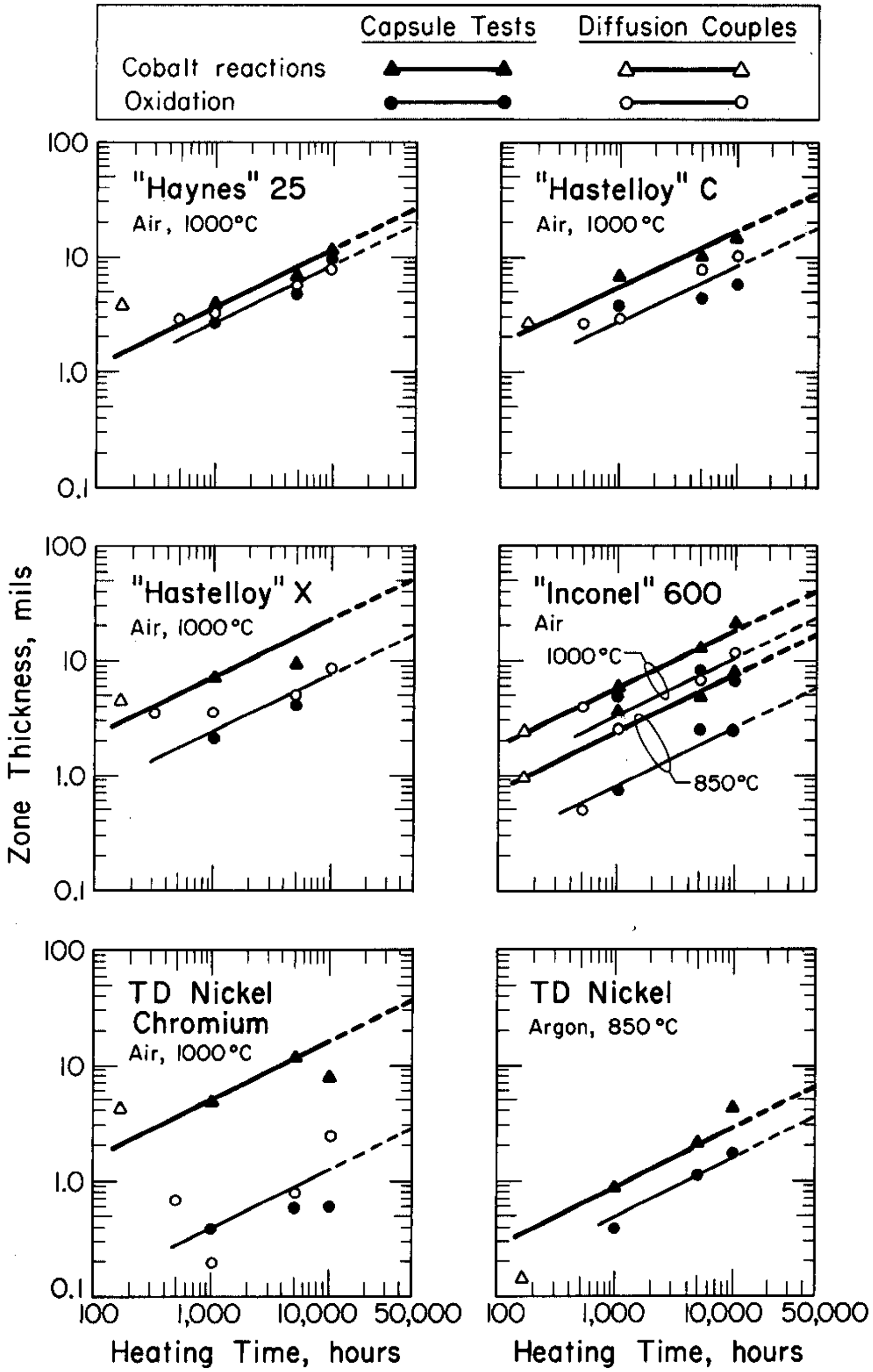


FIG. 4 GROWTH OF COBALT-CAPSULE COMPATIBILITY AND CAPSULE OXIDATION ZONES (Unirradiated Cobalt)

Although TD Nickel Chromium and TD Nickel would not be recommended for actual heat sources, capsule tests of these materials are being continued to provide further proof of the parabolic relationship between the heating time and the compatibility and oxidation reactions. These materials are unacceptable because the diffusivity of ^{60}Co is too fast, and they are difficult to weld. Tests with TD Nickel are done in flowing argon because of the high oxidation rate in air.(5)

Effects of Component Cleanliness on Compatibility

Heating tests were started to measure the effects of residues from the caustic dissolution of the aluminum cladding of the target slugs on the compatibility of cobalt and "Inconel" 600. The cladding of two unirradiated target slugs was dissolved in caustic by the same procedure as that for irradiated cobalt target slugs. The cobalt wafers were removed from the caustic and allowed to dry, leaving a film of reaction products. These wafers were encapsulated, without any additional cleaning, in two "Inconel" 600 capsules that are being heated at 900°C for 5000 and 10,000 hours, respectively. These residues may have been the cause of the increased cobalt-"Inconel" 600 reaction observed in two capsules that contained irradiated cobalt and were heated for 10,000 hours at 900°C.(1)

Effects of Thermal Gradients on Capsule Performance

Vapor transport of "Inconel" 600 and cobalt was produced by heating a 3-inch-long "Inconel" 600 capsule for 500 hours in a thermal gradient. One end was held at 800°C and the other at 950°C. Material was deposited between the "Inconel" 600 spacer and the first cobalt wafer and between successive cobalt wafers at the cooler (800°C) end of the capsule. Microprobe analyses showed that material between the spacer and the first wafer had essentially the same composition as "Inconel" 600, but enriched in cobalt. In contrast, deposits between the cobalt wafers were primarily cobalt with small amounts of chromium, nickel, and iron, the constituents of "Inconel" 600. Similar vapor transport is believed to have been the cause of the peninsular grains present between the irradiated cobalt wafers contained in the two "Inconel" 600 capsules heated 10,000 hours at 900°C.(1)

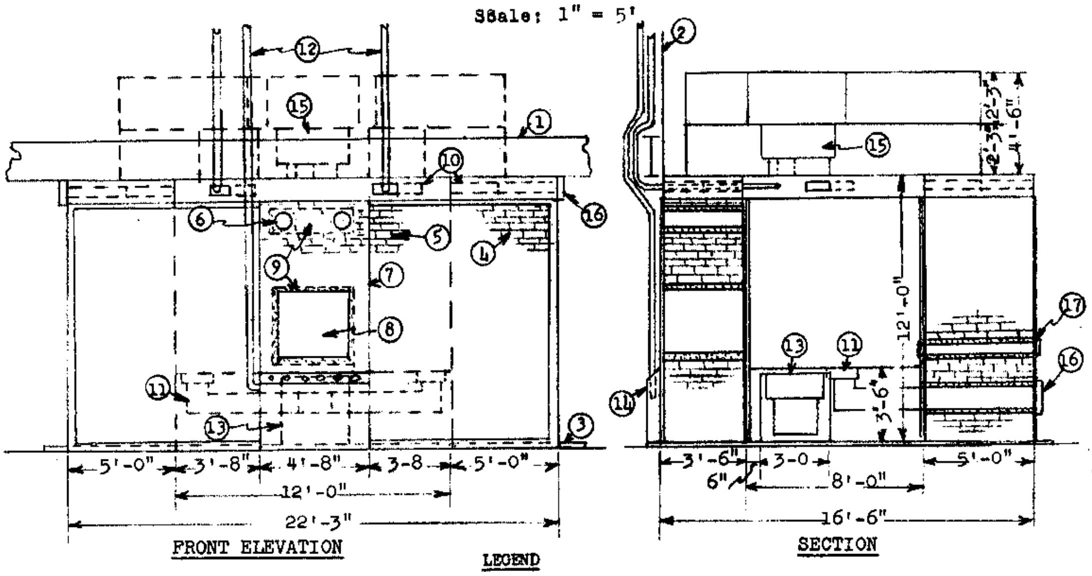
Heating Tests of Capsules Containing Irradiated Cobalt

Long-Term Tests

Twenty superalloy capsules, 18 containing irradiated and 2 containing unirradiated cobalt metal are being heated in air at 900 to 1000°C in the High Level Caves (HLC) to demonstrate capsule performance at typical heat source conditions, Table II. In April, one capsule each of "Inconel" 600, "Hastelloy" X, and "Haynes" 25 will attain goal exposures of 5000 hours at 1000°C. Destructive examination of these capsules will be deferred until FY-1970 because of conflicts with other isotope programs for cave space.

Test Facilities

A preliminary scope of work was completed for a shielded cell to be located in the Isotope Process Development Laboratory (IPDL) and to be used for long-term heating tests of capsules containing ^{60}Co, Figure 5. The cell walls, constructed of magnetite and ordinary concrete blocks, will provide shielding for 500,000 curies of ^{60}Co. The stainless steel containment box has a working area 12-feet long by 8-feet wide to accommodate seven furnaces for the heating tests. Three muffle furnaces will be used to heat superalloy capsules containing cobalt metal at 800 to 1000°C. Two high-vacuum furnaces will be used to heat refractory metal capsules containing cobalt metal at about 1200°C and two similar furnaces will be used to heat refractory or noble metal capsules containing cobalt compounds at 1200°C and higher. Transfer of capsules between the IPDL cell and the High Level Caves, where the capsules are fabricated and finally destructively examined, will be done with a bottom-loading cask through transfer ports in the cell roofs. Nondestructive tests, such as dimensional measurements and helium leak tests, will also be done in the new IPDL cell. A water-cooled storage cask underneath the box floor provides shielding for 500,000 curies in case the cell roof has to be removed for replacement of a defective furnace.



1-Existing beam
2-Existing containment wall
3-Steel pad
4-Ordinary concrete block; end and rear walls
5-Magnetite concrete block; 12' span of front wall
6-Manipulator ports
7-Steel plate supporting manipulators and window
8-Window; EBR-II type
9-Cast concrete fill
10-Service and ventilation ports
11-Ventilation ducts under box floor
12-Service piping
13-Storage cask
14-Service control panel
15-Capsule transfer port
16-Filter boxes
17-Vacuum piping

FIG. 5 ^{60}Co CELL FOR IPDL

Safety Tests

Cobalt Release

A test was started to measure the amount of ^{60}Co released through a pinhole in an "Inconel" 600 capsule during heating at 1000°C. A solid rod of unirradiated cobalt metal was plated with ^{60}Co and encapsulated in an "Inconel" 600 capsule that had a 0.008-inch-diameter hole drilled through the capsule wall. This capsule is being heated at 1000°C in a stream of air that is filtered and monitored daily for radioactivity from the ^{60}Co. The amount of cobalt released from the capsule will be calculated from the measured activity.

Compatibility of Molten Cobalt with Tungsten and Rhenium

Initial tests were run to measure the compatibility of liquid cobalt with tungsten. Penetration of a pure tungsten capsule with 0.100-inch-thick walls occurred in only one localized area during heating for four hours at 1550°C. Similar, but more extensive, penetrations occurred in a "Kennertium"* capsule of the same dimensions. Penetrations up to 0.055-inch deep occurred in both tungsten and "Kennertium" capsules that had 0.375-inch-thick walls; however, a portion of the molten cobalt flowed out of the capsules through cracks in the girth welds. Continuing tests of the compatibility of molten cobalt with tungsten will evaluate the consequences of temperature excursions to above the melting point of cobalt (1485°C) that might occur under abnormal conditions such as loss of coolant, re-entry from space, or after burial in the earth.

Two designs of capsules that had different relative amounts of cobalt and tungsten or "Kennertium" were used in the compatibility tests. Type A capsule (1.3-inch long by 1.0-inch in diameter with 0.100-inch-thick walls) contained a stack of 0.750-inch-diameter by 0.965-inch-long cobalt wafers. Type B capsule (2.5-inch long by 1.0-inch in diameter with 0.375-inch-thick walls) contained a 0.24-inch diameter by 0.46-inch long cobalt rod. Type A capsule contained 30% Co by wt and Type B capsule contained 0.5% Co by wt. The two designs were selected because the phase diagram indicated that the eventual extent of reaction is dependent upon the relative masses of cobalt and tungsten. Type A capsule was sealed by a circumferential TIG weld at one end; Type B capsule was sealed by a girth weld at the center of the capsule (see Development of Welding Techniques below).

Complete penetration through the 0.100-inch wall occurred in both the tungsten and the "Kennertium" Type A capsules, Figure 6. The attack of the tungsten capsule was confined to one localized area in contrast to the general attack of the "Kennertium" capsule. In the area of penetration, the pure tungsten dissolved uniformly in the molten cobalt; no preferential dissolution at the tungsten grain boundaries was visible. In other areas no change was observed in the wall thickness of the capsule. Penetration of the "Kennertium" progressed along the grain boundaries that were rich in the copper and nickel alloying elements so that each original particle of tungsten became isolated and then dissolved.

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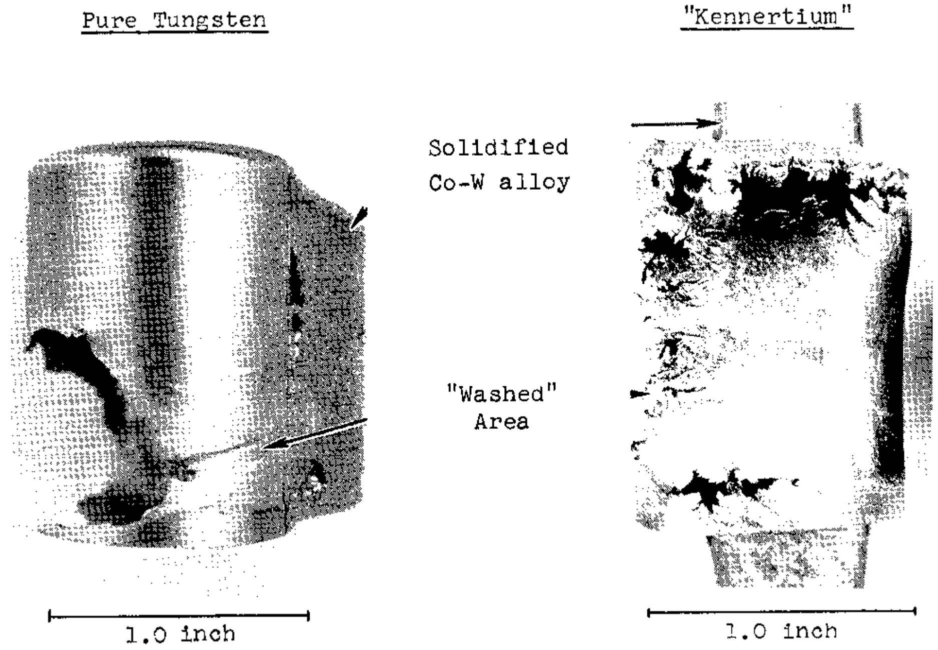
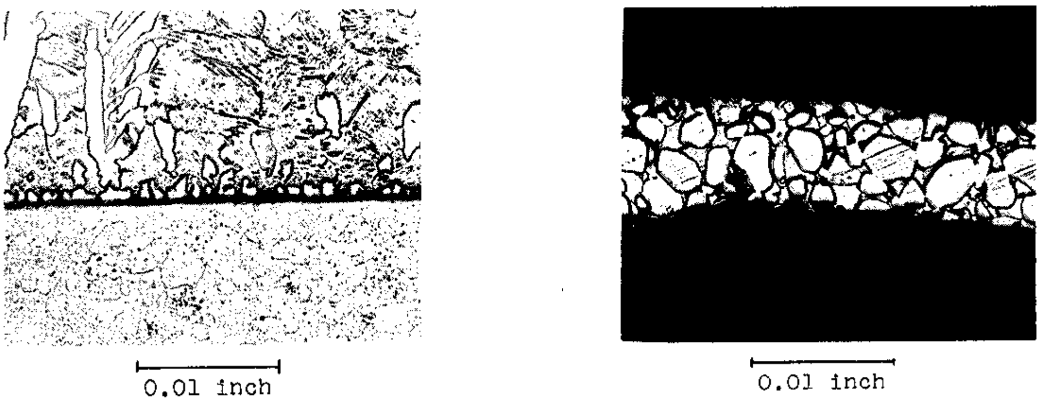
* Trademark of Kennametal, Inc. ("Kennertium" is a machinable W-2 wt % (Cu + Ni) alloy)



General capsule appearance

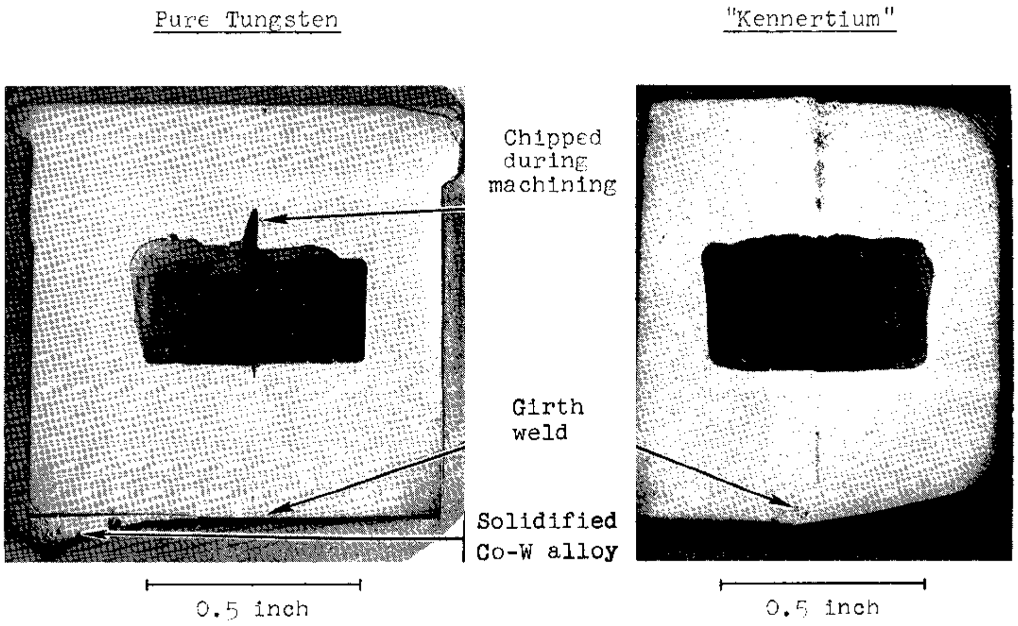


Interface between Co-W alloy (top) and capsule wall

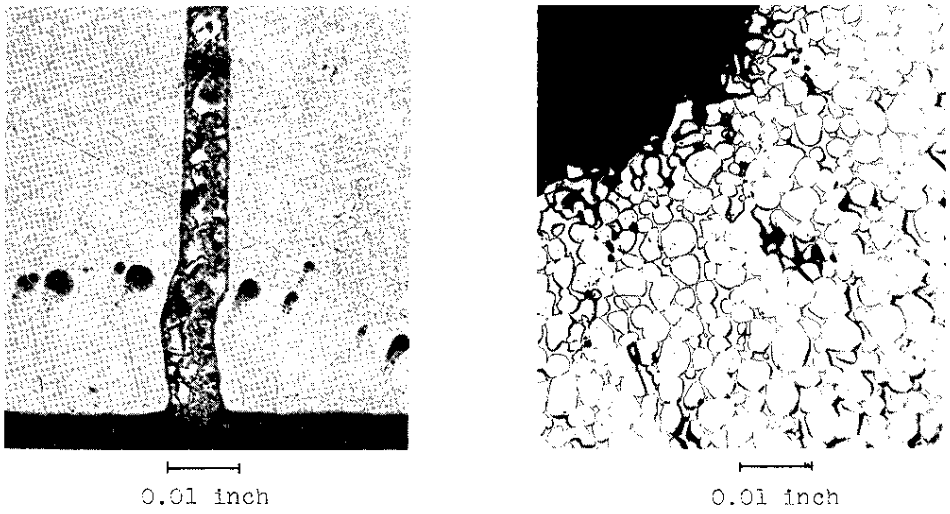
Residual capsule wall

FIG. 6 TYPE A TUNGSTEN AND "KENNERTIUM" CAPSULES AFTER HEATING 4 HOURS AT 1550°C

Penetrations up to 0.055-inch deep occurred in both tungsten and "Kennertium" Type B capsules, Figure 7. Most of the molten cobalt-tungsten (Co-W) alloy flowed out of the tungsten capsule through cracks in the girth welds. No Co-W alloy was visible in the "Kennertium" capsule. The cobalt diffused into the capsule partly on the inside and partly on the outside after it flowed through cracks in the girth welds.



General capsule appearance



Penetration of Co-W alloy through crack in girth weld

Bottom inside corner of capsule

FIG. 7 TYPE B TUNGSTEN AND "KENNERTIUM" CAPSULES AFTER HEATING 4 HOURS AT 1550°C

In each of the four capsules the penetration occurred in the upper portion of the capsule as it lay in the horizontal position in an alumina boat. The location of the attack may be associated with vaporization of cobalt or impurities in the void space that accommodated the volume expansion of the cobalt during melting. If the cause of the localized dissolution of tungsten can be controlled, containment of liquid cobalt may be possible.

Additional compatibility tests will be made with Type B capsules of pure tungsten that have been redesigned to eliminate the girth weld and void space. Similar tests will be made with rhenium capsules and with capsules that contain superalloys as well as cobalt.

Refractory Metal Capsules

Development of Welding Techniques

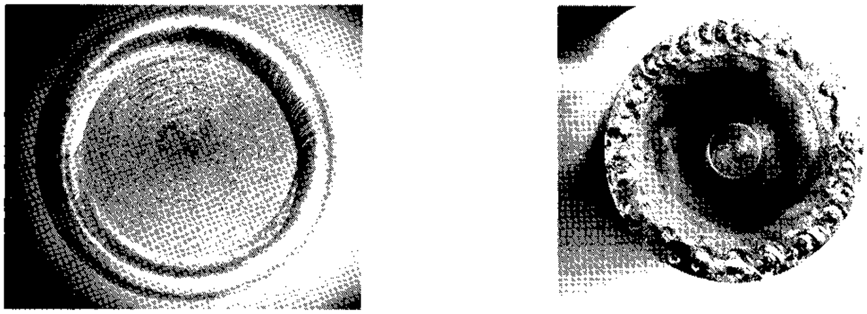
Development was begun on techniques for welding refractory metal capsules to be heated at about 1200°C. Initial TIG welding of powder-metallurgy tungsten and "Kennertium" capsules was only partially successful. A tungsten capsule of a design similar to that proposed for encapsulating ^{60}Co metal and oxide was welded with promising results. Cap welds in "Kennertium" and girth welds in both tungsten and "Kennertium" were either cracked or pitted, or both.

The initial welding was done on capsules that were used to test the compatibility of the capsule materials with liquid cobalt, Figure 8. Cap welds were made by welding a 0.9-inch-diameter disk recessed into a 1.0-inch-diameter capsule. Girth welds joined the two halves of a 2.5-inch-long by 1.0-inch-diameter capsule.

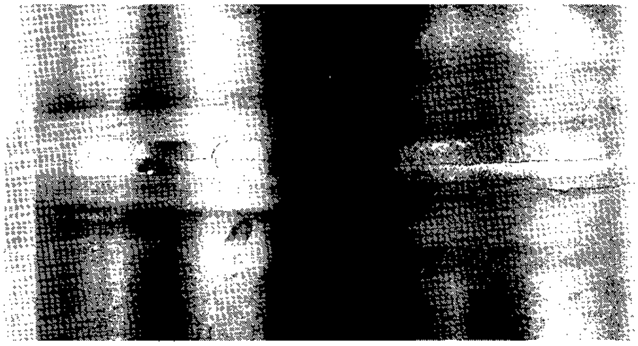
The tungsten cap weld, Figure 8a, appeared smooth and sound from the outside, except for two small cavities on the surface. Sectioning of the weld revealed porosity within the weld that is typical of weldments of power-metallurgy tungsten. Careful cleaning of all components may help to minimize porosity in future welds.

"Kennertium" is an alloy for which welding is not recommended but which machines well. During welding the molten region emits sparks, smoke, and liquid metal droplets. The resulting weld is pocked and does not appear sound, Figure 8b.

Girth welds in tungsten and "Kennertium" were cracked around the circumference of the capsules, Figure 8c. The cracks are due to stresses caused by thermal contraction of the capsules during or just subsequent to welding. Special cooling blocks could minimize cracking.



a. Tungsten cap weld b. "Kennertium" cap weld



c. Tungsten (left) and "Kennertium" (right) girth welds

Note circumferential cracks in the weld centers of both capsules and in the heat affected zone of the "Kennertium" capsule.

FIG. 8 WELDS IN REFRACTORY METAL CAPSULES

Development of techniques and equipment for welding refractory metals will continue. Small capsules of tungsten, rhenium, W-25 Re, and TZM alloy are being procured for weld development and for encapsulation of cobalt metal and oxide.

HIGH ACTIVITY ^{60}Co FOR HEAT SOURCE DEVELOPMENT

High activity cobalt that is being stored at Savannah River for experimental programs and heat source demonstrations is listed in Table V.

TABLE V

^{60}Co for Heat Sources[a]

Cobalt Shape	Average Activity, Ci/g	No. of Pieces	Total Activity, 10^6 Ci	Total Power, kw
0.745-inch Wafers				
Fuel form - cobalt metal (wrought)	340	2090	1.77	27.6
Fuel shape - wafers 0.745 ±0.001-inch diameter	300	912	0.68	10.6
0.040 ±0.003-inch thick	280	1368	0.96	14.9
including 0.0005 to 0.001-inch Ni plate	255	1368	0.87	13.6
Cobalt density - 8.80 ±0.05 g/cm^3	270	152	0.10	1.6[b]
		5890	4.38	68.3
0.800-inch Wafers				
Fuel form - cobalt metal (sintered)	330	255	0.24	3.7
Fuel shape - wafers 0.800 ±0.001-inch diameter	350	136	0.14	2.2[c]
0.040 ±0.003-inch thick	300	136	0.11	1.7[d]
including 0.0005 to 0.001-inch Ni plate		527	0.49	7.6
Cobalt density - 8.60 ±0.10 g/cm^3				
Nickel-Plated Slabs				
Fuel form - cobalt metal (wrought)	330	45	0.25	3.9
Fuel shape - slabs 3.00 ±0.03-inch long	305	48	0.25	3.9
0.640 ±0.002-inch wide		93	0.50	7.8
0.060 ±0.001-inch thick				
including 0.0005 to 0.001-inch Ni plate				
Cobalt density - 8.80 ±0.05 g/cm^3				
Stainless Steel-Canned Slabs				
Fuel form - cobalt metal (wrought)	330	45	0.20	3.1
Fuel shape - slabs 2.96 ±0.03-inch long	280	48	0.19	3.0
0.735 - 0.740-inch wide		93	0.39	6.1
0.092 ±0.001-inch thick				
sheath thickness 0.015-inch min				
cobalt dimensions same as nickel-plated slabs above except 2.44-inch long				
Cobalt density - 8.80 ±0.05 g/cm^3				
Stainless Steel-Coextruded Slabs				
Fuel form - cobalt metal (wrought)	330	60	0.23	3.6
Fuel shape - slabs 3.00 ±0.03-inch long	305	64	0.23	3.6
0.740 ±0.002-inch wide		124	0.46	7.2
0.072 ±0.002-inch thick				
SST thickness 0.015-inch min				
Cobalt dimensions: 2.75-inch long				
0.71-inch wide				
0.042-inch thick				
Cobalt density - 8.80 ±0.05 g/cm^3				
		Grand Total	6.22 MCi	97.0 kw

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[a] Activity and power as of September 30, 1968.

[b] Wafers have central hole of 0.070-inch diameter.

[c] 110 wafers have experimental compositions, to be used at SRL.

[d] 68 wafers have central hole of 0.070-inch diameter.

SAVANNAH RIVER LABORATORY ^{60}Co PUBLICATIONS

Quarterly Progress Reports

"Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report," compiled by H. S. Hilborn

DP-1088	July - September 1966
DP-1094	October - December 1966
DP-1105-I	January - March 1967, Part I - Cobalt
DP-1120-I	April - June 1967, Part I - Cobalt
DP-1129-I	July - September 1967, Part I - Cobalt
DP-1143-I	October - December 1967, Part I - Cobalt
DP-1155-I	January - March 1968, Part I - Cobalt
DP-1169-I	April - June 1968, Part I - Cobalt
DP-1177-I	July - September 1968, Part I - Cobalt
DP-1192-I	October - December 1968, Part I - Cobalt

Topical Reports

DP-974	"^{60}Co Heat Sources for 10-60 kw(e) Generators" by A. H. Dexter, July 1965.
DP-1012	"Radioactive Cobalt for Heat Sources" by J. W. Joseph, H. F. Allen, C. L. Angerman, and A. H. Dexter, October 1965.
DP-1051 (Rev. 2)	"Properties of ^{60}Co and Cobalt Metal Fuel Forms", June 1968.
DP-1096	"Development of ^{60}Co Capsules for Heat Sources" by C. P. Ross, C. L. Angerman, and F. D. R. King, June 1967.
DP-1145	"Experimental ^{60}Co Heat Source Capsules" by J. P. Faraci, May 1968

Journal Articles

A. H. Dexter, W. R. Cornman, and E. J. Hennelly. "The Advantages of ^{60}Co for Heat and Radiation Sources", Nucl. Appl. 2(2), 99-101 (1966).

C. P. Ross. "Cobalt-60 for Power Sources", Isotopes and Radiation Technology, 5(3), 185-94 (1968).

C. L. Angerman, F. D. R. King, J. P. Faraci, and A. E. Symonds. "^{60}Co Heat Source Encapsulation", Nucl. Appl. 4(2), 88-95 (1968).

C. L. Angerman and J. P. Faraci. "Heating Tests of Encapsulated Cobalt Heat Sources", presented at AIME Nuclear Metallurgy Symposium on Materials for Radioisotope Heat Sources, Gatlinburg, October 1968.

REFERENCES

1. H. S. Hilborn (Compiler). Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, October-December 1968, Part I - Cobalt 60. USAEC Report DP-1192-I, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1969).

2. C. P. Ross, C. L. Angerman, and F. D. R. King. Development of ^{60}Co Capsules for Heat Sources. USAEC Report DP-1096, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1967).

3. H. S. Hilborn (Compiler). Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, April-June 1967, Part I - Cobalt-60. USAEC Report DP-1120-I, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1967).

4. H. S. Hilborn (Compiler). Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, July-September 1967, Part I - Cobalt-60. USAEC Report DP-1129-I, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1967).

5. H. S. Hilborn (Compiler). Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, October-December 1967, Part I - Cobalt-60. USAEC Report DP-1143-I, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1968).

6. H. S. Hilborn (Compiler). Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, January-March 1968, Part I - Cobalt-60. USAEC Report DP-1155-I, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1968).

7. H. S. Hilborn (Compiler). Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, April-June 1968, Part I - Cobalt-60. USAEC Report DP-1169-I, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1968).

8. H. S. Hilborn (Compiler). Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, January-March 1967, Part I - Cobalt-60. USAEC Report, DP-1105, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1967).

9. R. Widmer, J. M. Dhosi, A. Mullendore, and H. F. Grant. Mechanisms Associated with Long-Time Creep Phenomena. AFML-TR-65-181 Parts I and II. New England Materials Lab., Inc., Medford, Mass. (1965).

10. S. C. Manson. Design Considerations for Long Life at Elevated Temperatures. James Clayton Lecture, Joint International Creep Conference, London, 1963.