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AEC RESEARCH AND DEVELOPMENT REPORT

SAVANNAH RIVER LABORATORY COBALT-60 POWER AND HEAT SOURCES

QUARTERLY PROGRESS REPORT

OCTOBER 1969

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Savannah River Laboratory

Aiken, South Carolina

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SAVANNAH RIVER LABORATORY
COBALT-60 POWER AND HEAT SOURCES

QUARTERLY PROGRESS REPORT

July - September 1969

Compiled by

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PREFACE

This report is one in a series on the applied aspects of ^{60}Co that are under study at the Savannah River Laboratory (SRL). 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 ^{60}Co fuel forms 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.

This report contains principally data from work performed during the report period. Previous reports are listed in the Publications section.

SUMMARY

Incentives and penalties were compared for using oxidized and alloyed forms of radioactive cobalt in heat sources. (p 1)

Melting temperatures were measured for CoO, $Co_{0.5}Mg_{0.5}O$, and $CoAl_2O_4$. (p 14)

Compatibility of the oxidized forms with rhenium was demonstrated at 1500°C. (p 15)

Capsules of "Inconel" 600 and "Hastelloy" C containing radioactive cobalt metal reached their goal exposure of 10,000 hr at 900°C. (p 19)

Tungsten capsules containing ^{60}Co can be handled for short times in air without excessive oxidation, as shown by tests at 500°C. (p 19)

In preliminary tests, corrosion of cobalt wafers in sea water was more rapid at defects in the nickel plating than rates reported for fully exposed cobalt surfaces. (p 22)

CONTENTS

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

High-melting forms of radioactive cobalt for heat sources are being considered for possible high-temperature applications, including thermionic converters (>1500°C), and for lower temperature applications for which they might provide increased safety over cobalt metal. The fuel forms include CoO, CoO-MgO solutions, $CoAl_2O_4$, and Co-Ru and Co-Re alloys.

Comparison of ^{60}Co Fuel Forms

The oxidized and alloyed forms of cobalt have higher melting temperatures than the base metal at the expense of decreased power density. These fuel forms are compared to cobalt metal in Table I. CoO and MgO form a continuous series of solid solutions, but for purposes of comparison CoO-MgO is listed at 50 mol % CoO. This composition has a high melting temperature (2230°C) and an acceptable power density (12 w/cc). The metallic alloys, Co-Ru and Co-Re, are listed at several compositions; alloys with cobalt concentrations less than 35 at. % have power densities less than $Co_{0.5}Mg_{0.5}O$, and Co-Ru alloys with cobalt concentrations greater than 50 at. % melt at nearly the same temperature as cobalt metal. $CoAl_2O_4$ has a higher melting temperature than CoO but its power density is the lowest of all the fuel forms compared. All of the fuel forms except Co-Re can be fabricated prior to irradiation; Co-Re must be alloyed after irradiation of cobalt because of the high neutron cross section of rhenium and its conversion to osmium, which forms a low-melting alloy with cobalt.

All of the fuels have advantages and disadvantages and none would be optimum for all applications. The intended application, with the corresponding fuel requirements for safety as well as for operation, must be defined to allow selection of a fuel form. Some general conclusions derived from comparing the fuels at ^{60}Co specific activity of 300 Ci/g are as follows:

TABLE I

Comparison of ^{60}Co Fuel Forms

Fuel Form	Co Atom %	Melting Point, °C	Theoretical Density, g/cc	Power Density,[a] w/cc	Advantages	Disadvantages
CoO	50	1800	6.4	23.5	Reasonable power density, fabricate before irradiation. Oxidation resistant	Low thermal conductivity
$Co_{0.5}Mg_{0.5}O$	25	2230	5.0	12.0	High melting temperature, more stable than CoO, fabricate before irradiation. Oxidation resistant	Lower power density than CoO, low thermal conductivity
$CoAl_2O_4$	14.3	1960	4.4	6.9	More stable than CoO, fabricate before irradiation. Oxidation resistant	Very low power density, low thermal conductivity
Co-Ru	20	1990	11.6	6.7	High thermal conductivity, fabricate before irradiation	Very low power density at high melting temperature, low melting temperature at high power density, oxidizes
Co-Ru	30	1860	11.3	10.3		
Co-Ru	40	1730	11.0	14.2		
Co-Ru	50	1600	10.7	18.2		
Co-Re	30	2400	18.1	10.1	High thermal conductivity, good power density	Fabricate after irradiation, oxidizes
Co-Re	40	2170	16.9	13.8		
Co-Re	50	1970	15.8	17.7		
Co-Re	60	1800	14.7	21.9		
Co-Re	70	1670	13.2	26.2		
Co Metal	100	1495	8.8	41.1	High power density, easily fabricated and handled. High thermal conductivity	Lowest melting temperature, oxidizes

[a] ^{60}Co specific activity 300 Ci/g for all forms

- Power density - The theoretical power density for 100% dense fuel, Figure 1, of CoO is greater than that for $Co_{0.5}Mg_{0.5}O$, $CoAl_2O_4$, and Co-Ru or Co-Re alloys containing up to 60 at. % cobalt. The power densities of the ruthenium and rhenium alloys are about the same over the full range of cobalt concentrations. The low power density of $CoAl_2O_4$ is a disadvantage that must be balanced against its potential excellent stability at high temperature.

- Melting temperature - Figure 2, summarizing the melting temperatures, shows $Co_xMg_{1-x}O$ and Co-Re to be the most refractory of the fuel forms. The low melting temperature of Co-Ru at reasonable power densities would probably preclude its use unless problems appear in the other fuel forms.

- Power density and melting temperature - Figure 3 shows the correlation of melting temperature and power density of the fuels. The mixed oxide $Co_xMg_{1-x}O$, CoO, and the alloy Co-Re are superior to the other fuels in this comparison.

- Operating temperature - Figures 4 and 5, comparing the calculated radial ΔT's in fuel cylinders, show the oxidized forms CoO, $CoAl_2O_4$, and $Co_{0.5}Mg_{0.5}O$ to have higher ΔT's than the metallic forms as expected. However, the ΔT is only 70°C or less for 1.2-cm-radius cylinders (typical of forms irradiated to date) because of the low internal gamma absorption. Figure 6 compares the maximum surface temperature that can be achieved in cylinders of various radii without melting in the center. $Co_{0.5}Mg_{0.5}O$ has the highest allowable surface temperature for cylinders through 3 cm in radius. CoO has a lower allowable surface temperature than cobalt metal for cylinders larger than 2 cm in radius.

- Heat block weight - For the purpose of comparison, fuel cylinders with length equal to diameter were assumed to be surrounded by a tungsten hot shield 2-cm thick to absorb 90% of the gamma radiation escaping from the fuel. Figure 7 shows the weight of the tungsten hot shield as a function of total energy; CoO requires the lightest hot shield because of its higher power density.

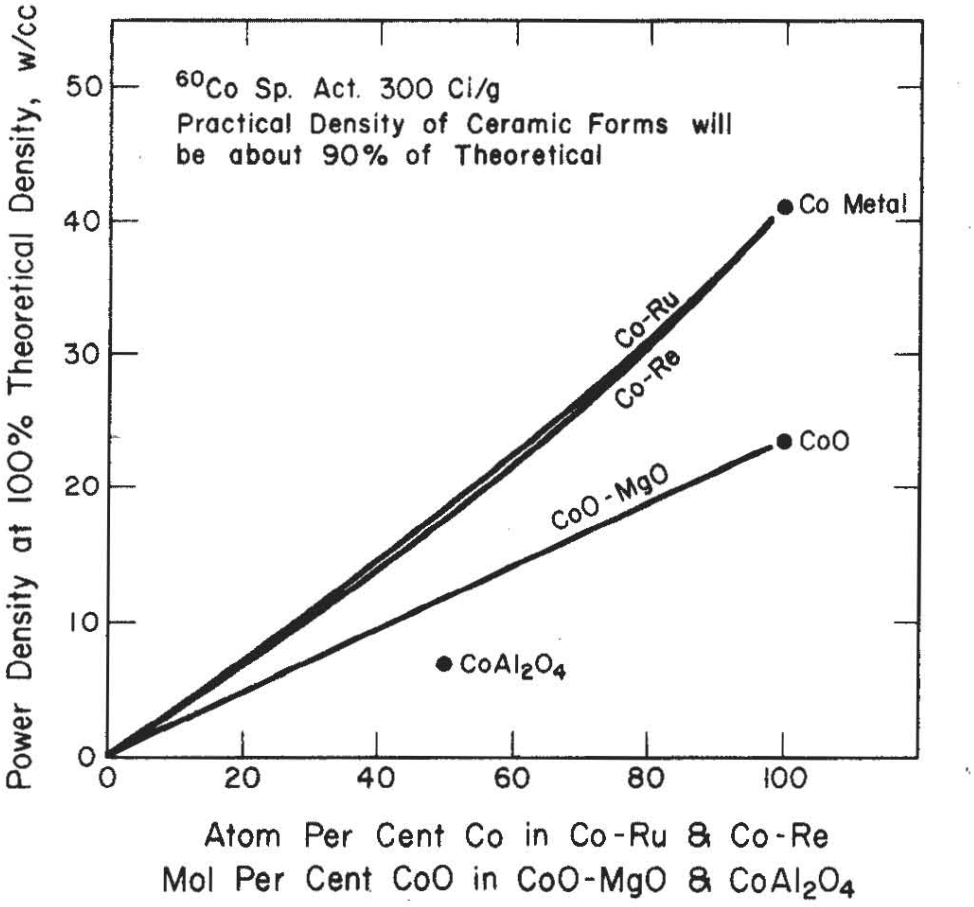

FIG. 1 THEORETICAL POWER DENSITY OF ^{60}Co FUEL FORMS

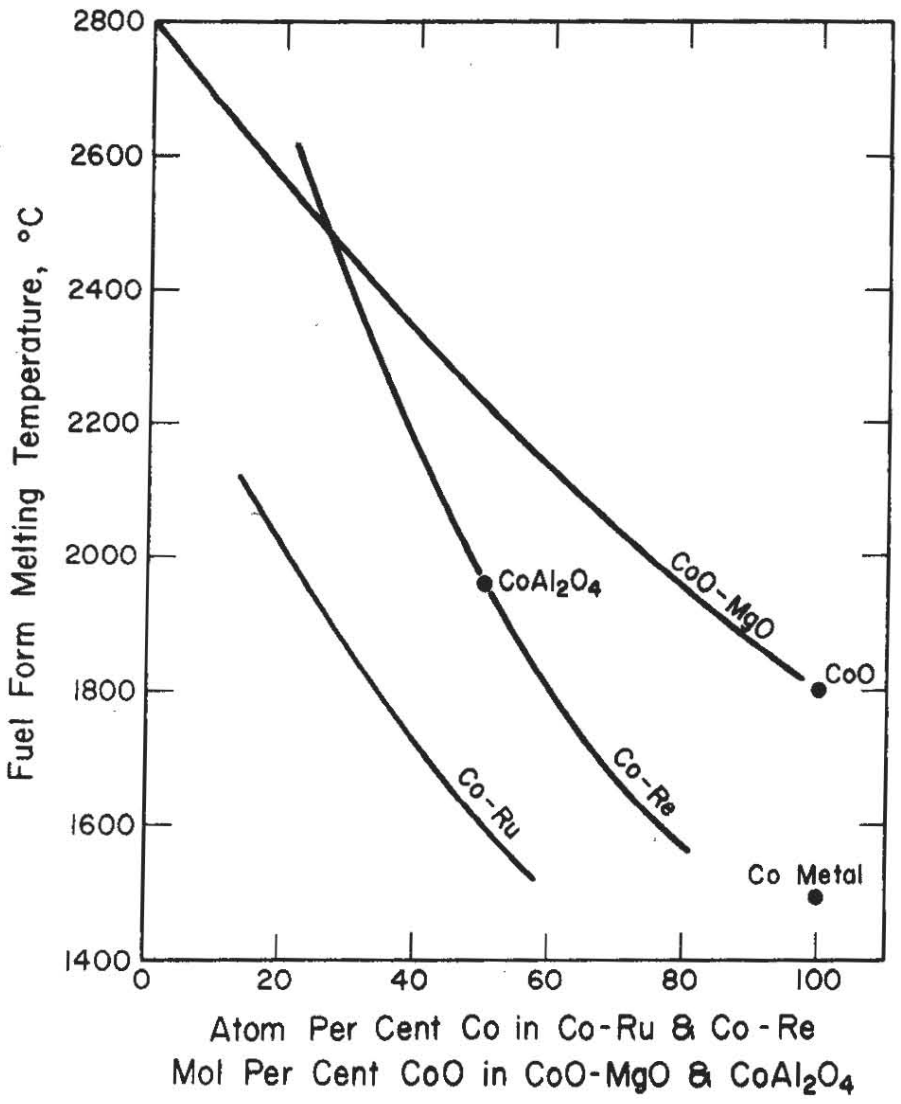

FIG. 2 MELTING TEMPERATURE OF ^{60}Co FUEL FORMS

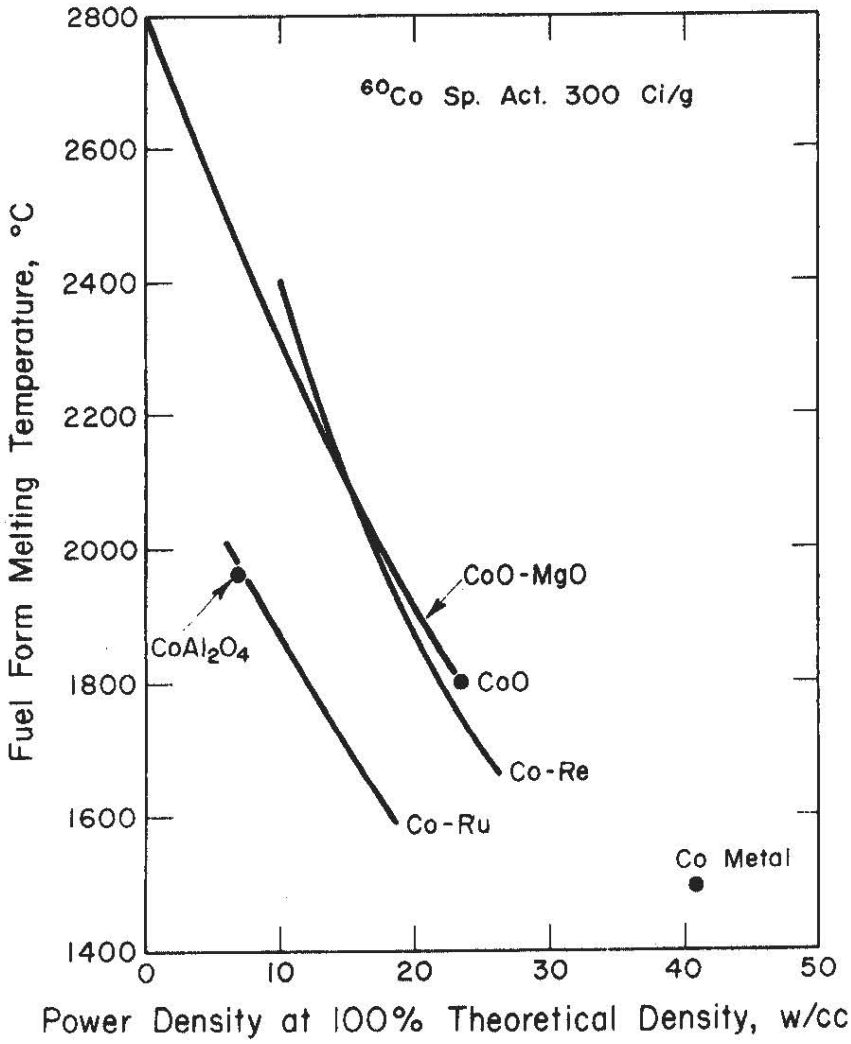

FIG. 3 CORRELATION OF POWER DENSITY AND MELTING TEMPERATURE OF ^{60}Co FUEL FORMS

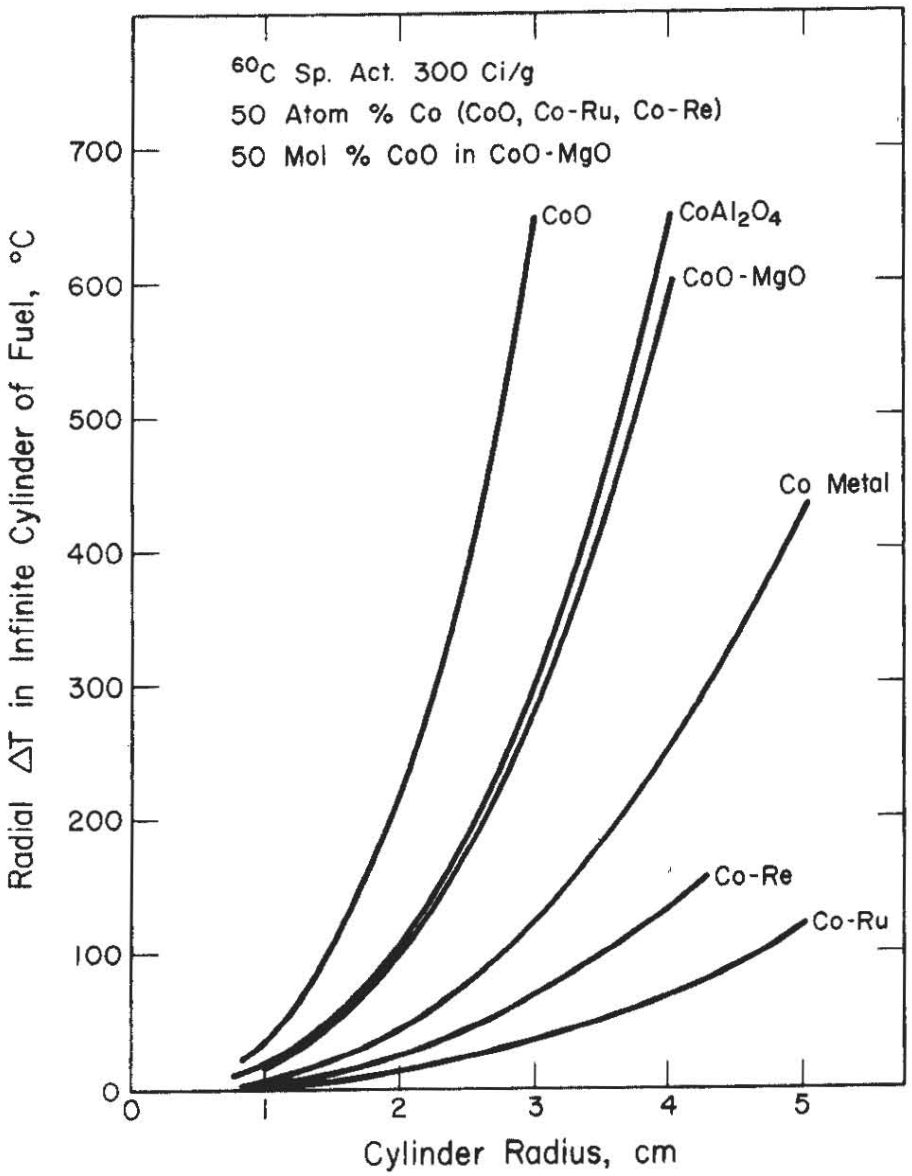

FIG. 4 RADIAL ΔT IN INFINITE CYLINDERS OF ^{60}Co FUEL FORMS

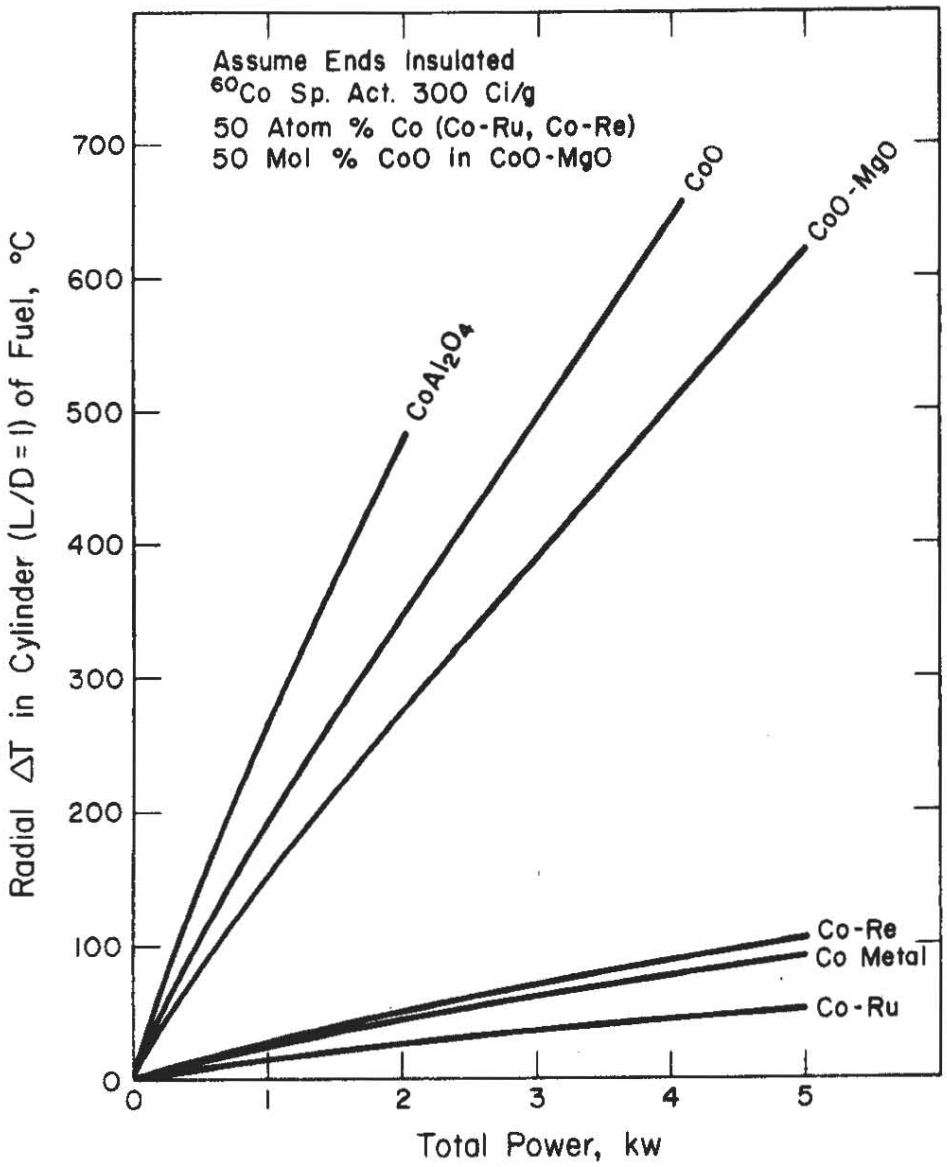

FIG. 5 RADIAL ΔT IN FUEL CYLINDERS WITH LENGTH EQUAL TO DIAMETER

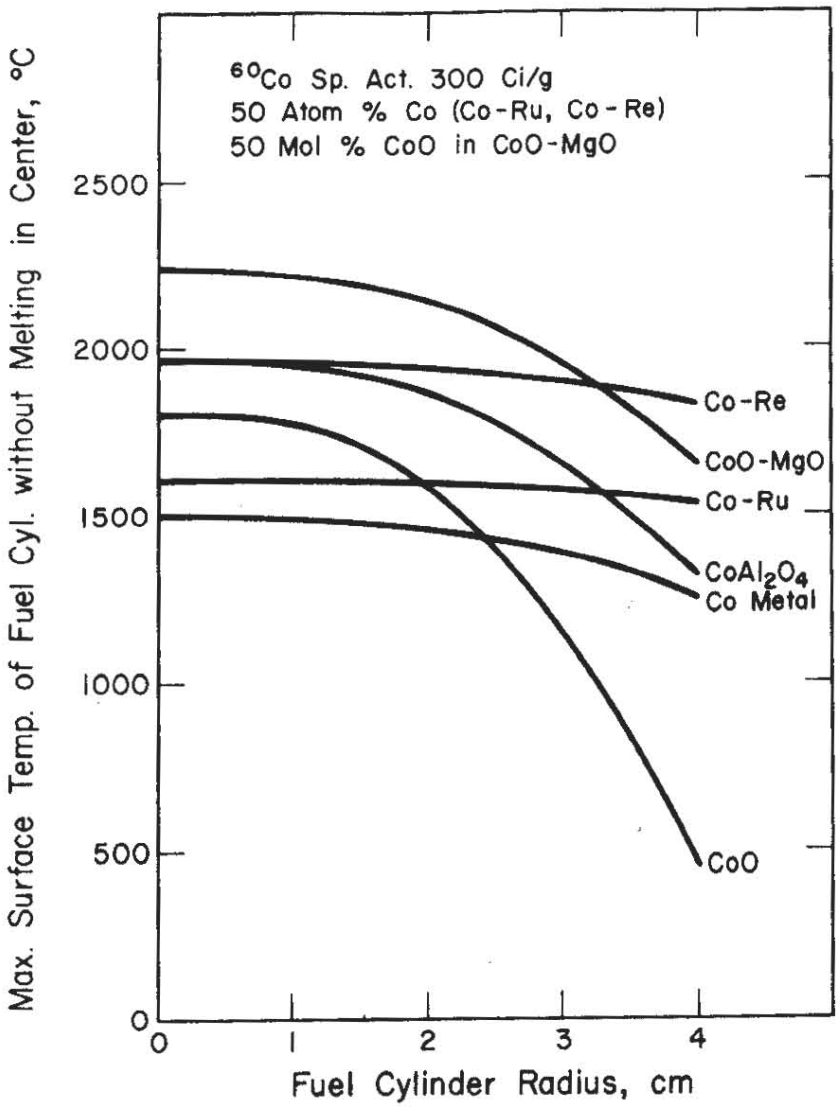

FIG. 6 MAXIMUM SURFACE TEMPERATURE OF FUEL CYLINDER WITHOUT MELTING IN CENTER

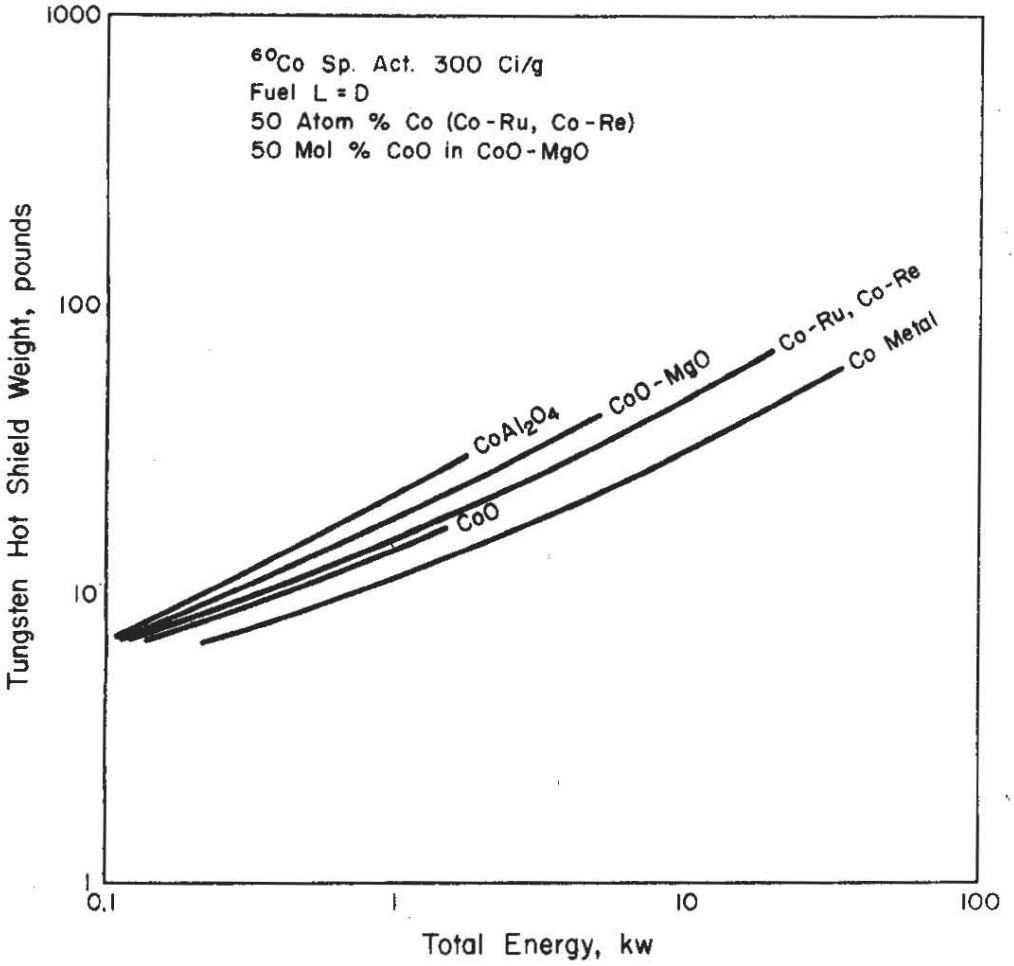

FIG. 7 TUNGSTEN HOT SHIELD TO ABSORB 90% OF GAMMA RADIATION ESCAPING FROM FUEL

- Overall consideration - From the foregoing conclusions, cobalt metal, CoO, $Co_{0.5}Mg_{0.5}O$ and Co-Re alloy are probably the best fuel forms for most applications. Final selection would depend on the specific application. SRL work to date has emphasized cobalt metal and the oxide forms because of the difficulty of alloying cobalt with rhenium, especially in a hot cell, and the potential oxidation of the alloy if exposed to air at high temperature.

Because the thermal conductivities of the oxides have not been measured, values for the calculations were estimated from elevated temperature data for similar oxides. A conductivity of 2 pcu/(hr)(ft^2)(°C/ft) was assumed for CoO and CoO-MgO and 1 pcu/(hr)(ft^2)(°C/ft) for $CoAl_2O_4$.

Fabrication of Oxide Wafers

Wafers of CoO, $Co_{0.5}Mg_{0.5}O$, and $CoAl_2O_4$ are being fabricated for irradiation in forthcoming high-flux reactor charges and subsequent development of refractory, oxidation resistant heat sources.[1-3] Estimated production in these reactor charges is given in Table II for the ceramic compounds and cobalt metal.

High-density wafers (0.700-in.-dia x 0.150-in.-thick) are fabricated by sintering powders obtained from hydroxide precipitation and decomposition.[4] The procedures followed for each oxide are basically similar and are summarized in the process flow sheet shown in Figure 8. CoO wafers are heated for an additional 16 hr in argon at 1000°C to decompose any Co_3O_4 that formed during cooling from the sintering temperature. Co_3O_4 is not formed in $Co_{0.5}Mg_{0.5}O$ or $CoAl_2O_4$.

Resistance to Thermal Shock

The oxide wafers were acceptably resistant to cracking when subjected to thermal shocks. Wafers of the three oxides did not crack when cooled from 1100°C to 400°C in 5 seconds. Wafers of CoO and $CoAl_2O_4$ did not crack, but wafers of $Co_{0.5}Mg_{0.5}O$ did, when cooled more rapidly by placing the 1100°C wafers on a polished aluminum disk that was immersed in ice water. Thermal gradients of several hundred degrees were established as the heat drained to the aluminum disk from one side of the wafer.

TABLE II

Planned Production of High Activity Cobalt in ^{252}Cf-Producing Charges

Cobalt Type[a]	Average Activity, Ci/g Co	No. of Pieces	Total Activity, MCi	Total Power, kw
Metal, 0.745-in.-dia wafers	450	1970	4.08	63.6
	300	1790	2.47	38.5
	200	4560	4.29	66.9
1.00-in.-dia half-wafers	450	396	.74	11.5
	300	286	.36	5.7
1.25-in.-dia half-wafers	450	252	.74	11.5
	300	182	.36	5.7
1.49-in.-dia half-wafers	450	360	1.48	23.0
	300	260	.72	11.4
CoO, 0.700-in.-dia wafers	450	108	.21	3.3
	300	12	.02	0.3
CoO-MgO solid solution, 0.700-in.-dia wafers	450	108	.11	1.7
	300	12	.01	0.2
$CoAl_2O_4$, 0.700-in.-dia wafers	450	108	.06	0.9
	300	12	.01	0.1
TOTAL			15.66	244.3

[a]Metal shapes 0.073-in.-thick
Ceramic shapes 0.150-in.-thick

HYDROXIDE PRECIPITATION

Excess 2.75M NaOH added at 25°C with stirring to:

0.25M $Co(NO_3)_2$ for CoO
0.25M $Co(NO_3)_2$ and 0.25M $Mg(NO_3)_2$ for $Co_{0.5}Mg_{0.5}O$
0.2M $Co(NO_3)_2$ and 0.4M $Al(NO_3)_3$ for $CoAl_2O_4$

WASH AND DRY

Wash and centrifuge until pH = 7
Dry at 120°C; screen - 100 mesh

CALCINE

4 hr in flowing argon at 835°C for CoO
4 hr in flowing argon at 750°C for $Co_{0.5}Mg_{0.5}O$
5 hr in flowing argon at 900°C for $CoAl_2O_4$

PRESS

Screen through 100 mesh; add binder
Press at 22,000 psi

SINTER

6 hr in air at 1650°C

MACHINE

Grind diameter and lap thickness to desired specifications

FIG. 8 PROCESS FLOWSHEET FOR FABRICATION OF COBALT OXIDE WAFERS

Melting Temperatures of Cobalt Oxides

The melting temperatures of the three oxides were measured by heating samples of the powders used in fabricating wafers for irradiation. Measured values of 1800°C for CoO, 2250°C for $Co_{0.5}Mg_{0.5}O$, and 1920°C for $CoAl_2O_4$ agree with values shown on published phase diagrams for the CoO-MgO and CoO-Al_2O_3 systems, Figures 9 and 10 respectively. CoO and MgO form a continuous series of ideal solid solutions with melting temperatures varying from 1800°C for pure CoO to 2800°C for pure MgO.[5] This system offers a range of power densities and corresponding melting temperatures. The system CoO-Al_2O_3 contains one compound, $CoAl_2O_4$, that melts congruently at 1960°C. The adjacent eutectic melting temperatures are estimated at 1910 and 1725°C, depending on whether the system is rich in Al_2O_3 or in CoO.

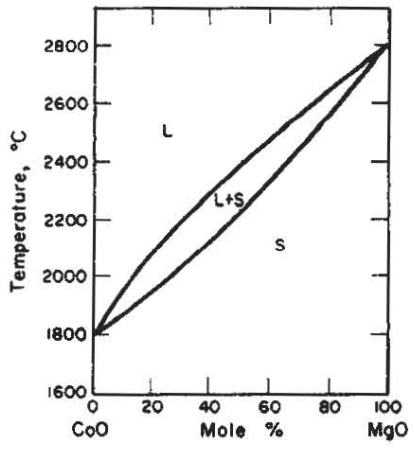

FIG. 9 CoO-MgO PHASE DIAGRAM (From Ref. 5)

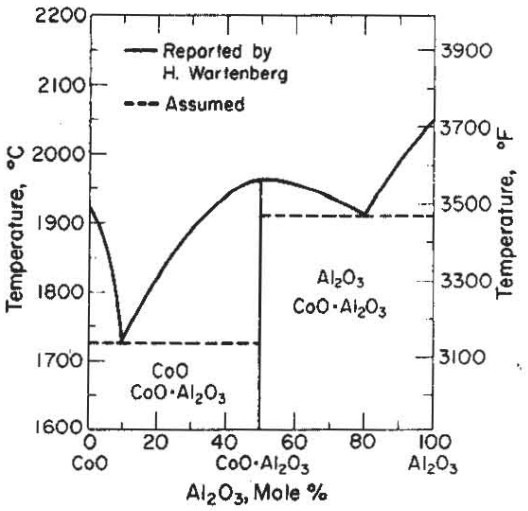

FIG. 10 CoO-Al_2O_3 PHASE DIAGRAM (From Ref. 6)

EVALUATION OF ENCAPSULATION MATERIALS FOR RADIOACTIVE COBALT

Compatibility of Cobalt Oxides with Capsule Materials

Thermodynamic analyses of the potential reaction between the three oxides and candidate high-temperature capsule materials indicated that rhenium and the noble metals would be compatible with the oxides.[1] Other refractory metals, such as tantalum, tungsten, and molybdenum, would reduce the oxides to cobalt metal and ternary compounds.

The predicted compatibility of rhenium and the oxides was demonstrated by heating capsules at 1500°C for 200 hr in a vacuum of 10^{-5} torr. Three rhenium capsules were spark-machined from powder-metallurgy rod. Each was loaded with alternating wafers of rhenium and one of the oxides and sealed by TIG welding. After the test, the capsules were intact and unchanged in appearance. Destructive examination showed a uniform black deposit (probably CoO) on the inside wall of each capsule and no gross reaction at the rhenium-oxide interfaces. Detailed metallographic examination and microprobe analyses are in progress.

Short-term compatibility of the oxides with iridium was demonstrated during the measurements of the melting temperature of the oxides. All three oxides melted on iridium filaments at the expected temperatures with no apparent reaction either during the fusion or in subsequent metallographic examination of the iridium filaments. In contrast, an attempt to measure the melting point of CoO on a tungsten filament failed when the tungsten reduced the oxide to cobalt metal at 1400°C; this cobalt then melted at 1495°C, the published value for cobalt metal.

CAPSULE FABRICATION AND TESTING

Development of Welding Techniques for Refractory Metals

Refractory metals and welding equipment are being procured for encapsulation of cobalt metal and cobalt oxides for applications above 1000°C. Capsules of tungsten, tungsten-25% rhenium, and rhenium are being purchased for welding development and long-term compatibility tests at ∿1200°C with ^{59}Co metal. Ten tungsten capsules and five tungsten-25 wt % rhenium alloy capsules have been delivered by the vendor.

Development of welding techniques for tungsten and W-25 wt % Re alloy capsules was delayed pending completion of the inert-gas welding station in the new Alpha Materials Facility (built to support Division of Production ^{244}Cm and ^{238}Pu programs). Sealing of the glove box to maintain a helium atmosphere with <10 ppm of O_2, N_2, and water vapor as impurities is nearly complete.

Heating Tests of Capsules Containing Unirradiated Cobalt Metal

Superalloy Capsules

Four capsules (one "Haynes"* 25, one "Hastelloy"* C, and two "Hastelloy"* X) attained goal exposures of 10,000 hr at 1000°C. Heating of all four capsules was resumed for an additional 10,000 hr because the capsules were intact (helium leak tests) and diameter increases were small, <0.002 in. for the "Hastelloy" capsules and 0.004 to 0.005 in. for the "Haynes" 25 capsule. The larger diameter increase for "Haynes" 25 had been observed in previous 10,000-hr tests. All three materials had demonstrated satisfactory resistance to oxidation and compatibility with cobalt in these earlier tests. These capsules are part of the continuing program to demonstrate structural integrity of superalloy capsules for >10,000 hr at typical temperatures of 850 to 1000°C, Table III.

Refractory Metal Capsules

A schedule was prepared for long-term heating tests of refractory metal capsules containing unirradiated cobalt metal; these tests will begin in November 1969, Table IV.

*Trademark of Union Carbide Corp.

TABLE III

Summary of ^{59}Co Capsule Heating Tests

Capsule Material	Heating Time, hr	Heating Temp, °C	Wall, mils	No. of Capsules	Approx. Starting Date	Approx. Completion Date	Remarks
"Inconel" 600 (m.p. 1370°C)	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	Capsule intact
	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 (m.p. 1270°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	
	20,000	1,000	95	1	5-68	9-70	
TD Nickel (m.p. 1450°C)	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	95	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 (m.p. 1430°C)	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 (m.p. 1330°C)	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	
	20,000	1,000	95	1	5-68	9-70	
"Hastelloy" X (m.p. 1260°C)	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	Capsule intact
	50,000	1,000	95	1	2-68	10-73	
	20,000	1,000	95	2	5-68	9-70	

[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.

TABLE IV

Summary of Planned ^{59}Co-Refractory Metal Capsule Heating Tests[a]

Capsule Material	Heating		Approx. Starting Date	Approx. Completion Date	Wafer Diameter, inch	Welding Technique
	Time, hr	Temp, °C				
Tungsten	1,000	1200	11-69	1-70	0.745	TIG
	1,000	1200	11-69	1-70	0.745	TIG
	5,000	1200	11-69	6-70	0.745	TIG
	5,000	1200	11-69	6-70	0.745	TIG
	5,000	1200	2-70	9-70	1.490	TIG
	5,000	1200	3-70	10-70	0.745	EB
	10,000	1200	11-69	1-71	0.745	TIG
	10,000[b]	1200	12-69	2-71[b]	0.745	TIG
	10,000	1200	2-70	4-71	1.490	TIG
	10,000	1200	2-70	4-71[b]	1.490	TIG
	10,000	1200	3-70	5-71	0.745	EB
	50,000	1200	12-69	8-75	0.745	TIG
Rhenium	1,000	1200	12-69	2-70	0.745	TIG
	5,000	1200	12-69	7-70	0.745	TIG
	5,000	1200	3-70	10-70	0.745	EB
	10,000	1200	1-70	3-71	0.745	TIG
	10,000[b]	1200	1-70	3-71[b]	0.745	TIG
	10,000	1200	3-70	5-71	0.745	EB
	50,000	1200	1-70	9-75	0.745	TIG
	1,000	1400	2-70	4-70	0.745	TIG
	5,000	1400	2-70	9-70	0.745	TIG
	10,000	1400	2-70	4-71	0.745	TIG
W-25 wt % Re	1,000	1200	11-69	1-70	0.745	TIG
	5,000	1200	12-69	7-70	0.745	TIG
	5,000	1200	3-70	10-70	0.745	EB
	10,000	1200	1-70	3-71	0.745	TIG
	10,000[b]	1200	1-70	3-71[b]	0.745	TIG
	10,000	1200	3-70	5-71	0.745	EB
	50,000	1200	1-70	9-75	0.745	TIG
	1,000	1400	2-70	4-70	0.745	TIG
	5,000	1400	2-70	9-70	0.745	TIG
	10,000	1400	2-70	4-71	0.745	TIG

[a]One capsule containing ~10 Co wafers 0.073-in.-thick and one 0.060-in.-thick spacer will be heated at each listed condition.

[b]Test time may be extended to 20,000 hr, or more, depending on results of other 10,000-hr tests.

Heating Tests of Capsules Containing Irradiated Cobalt Metal

Superalloy Capsules

Fifteen superalloy capsules, 13 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 V. An "Inconel"* 600 capsule containing 36,500 Ci and a "Hastelloy" C capsule containing 13,000 Ci achieved their goal exposures of 10,000 hr at 900°C and were removed from test. Another "Inconel" 600 capsule containing 13,700 Ci also reached 10,000 hr at 900°C; heating of this capsule will be continued for an additional 10,000 hr unless examination of the first "Inconel" capsule reveals unsatisfactory performance. Destructive examination of these two capsules and the three removed from test in April after 5000 hr at 1000°C are scheduled to begin in January 1970 after completion of the rearrangement of High Level Cave metallographic facilities.

Refractory Metal Capsules

A schedule was prepared for long-term heating tests of refractory metal capsules containing irradiated cobalt metal; these tests will begin in January 1971, Table VI.

Test Facilities

Design of a shielded capsule test facility for the Isotope Process Development Laboratory (IPDL) is continuing. Preparation of a Project Authorization Request for operating and process equipment for the facility was started.

Oxidation of Tungsten

A study of the oxidation of tungsten in air showed that relatively simple precautions will be required to prevent serious oxidation of tungsten capsules during fabrication and handling prior to the long-term heating tests in high-vacuum furnaces. Cooling the capsules below 500°C should provide adequate protection since the maximum depth of oxidation was <0.001 in. when a tungsten sample was heated in air at 500°C for 6 hr. Oxidation extended to a depth of ∿0.011 in. after heating for 2 hr at 900°C, the maximum calculated surface temperature for a 1-in.-dia x 1-in.-long capsule with an emissivity of 0.2 and containing 15,000 Ci of ^{60}Co or ∿250 w(t). Thermal cycling between room

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temperature and 900°C for 2 hr (20 min at 900°C/cycle) affected approximately twice this depth. Oxidation was uniform at both temperatures; no pitting or preferential attack occured at the grain boundaries. Additional tests will be made at ∿600°C, the expected capsule temperature (emissivity of 0.6).

TABLE V

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
(m.p. 1370°)	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	Begin destructive examination 1-70
	→20,000	900	95	1	288[c]	13,700	7-68	10-70	
	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	Begin destructive examination 1-70
	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
(m.p. 1270°C)	→10,000	900	95	1	276[c]	13,100	7-68	8-69	Begin destructive examination 1-70
	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	Begin destructive examination 1-70
(m.p. 1330°C)	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	Begin destructive examination 1-70
(m.p. 1260°C)	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.
→ New information reported

TABLE VI

Summary of Planned ^{60}Co-Refractory Metal Capsule Heating Tests[a]

Capsule Material	Heating Time, hr	Heating Temp, °C	Approx. Starting Time	Approx. Completion Date	Wafer Diameter, inch
Tungsten	1,000	1200	1-71	3-71	0.745
	1,000	1200	1-71	3-71	0.745
	5,000	1200	1-71	8-71	0.745
	5,000	1200	1-71	8-71	0.745
	5,000	1200	3-71	10-71	1.490
	10,000	1200	1-71	3-72	0.745
	10,000[b]	1200	1-71	3-72[b]	0.745
	10,000	1200	3-71	7-72	1.490
	10,000[b]	1200	3-71	7-72	1.490
	50,000	1200	1-71	9-76	0.745
Rhenium	5,000	1200	2-71	9-71	0.745
	10,000	1200	2-71	4-72	0.745
	10,000[b]	1200	2-71	4-72[b]	0.745
	50,000	1200	2-71	10-76	0.745
	5,000	1400	3-71	7-72	0.745
	10,000	1400	3-71	7-72	0.745
	10,000[b]	1400	3-71	7-72[b]	0.745
	50,000	1400	3-71	11-76	0.745
W-25 wt % Re	5,000	1200	2-71	9-71	0.745
	10,000	1200	2-71	4-72	0.745
	10,000[b]	1200	2-71	4-72[b]	0.745
	50,000	1200	2-71	10-76	0.745
	5,000	1400	3-71	10-71	0.745
	10,000	1400	3-71	7-72	0.745
	10,000[b]	1400	3-71	7-72[b]	0.745
	50,000	1400	3-71	11-76	0.745

[a]One capsule (ten wafers 0.073-in.-thick and one spacer 0.060-in.-thick) will be heated at each listed condition. Capsules with 0.745-in.-dia wafers will contain ∿250 watts, or 16,000 Ci of 350 Ci/g cobalt; capsules with larger wafers will produce up to 1000 watts.

[b]Test time may be extended to 20,000 hr, or more, depending on results of other 10,000-hr tests.

Safety Tests

Cobalt Release

Measurement of the release of ^{60}Co from an intentionally defected "Inconel" 600 capsule heated in flowing air at 1000°C is continuing.[2] The capsule has a 0.008-in.-dia hole drilled through the wall and contains a solid rod of unirradiated cobalt electroplated with ^{60}Co. Low levels of radioactivity (~10 c/m above background) become deposited each day on a filter located in the effluent air stream. Total test time to date is ~4000 hr.

Corrosion of Cobalt Metal in Sea Water

Six unirradiated, nickel-plated cobalt wafers were immersed for 865 hr in sea water (North Carolina coast) in preliminary tests to measure their corrosion resistance. The wafers, held 1/4-in. apart in a "Teflon"* rack, were suspended from a piling to a depth of 3 to 24 in., depending on tide level. Three of the wafers were mechanically dislodged by wave action and were lost.

Severe corrosion of the cobalt occurred wherever there was a defect in the nickel coating. The attack spread laterally around a defect, leaving the nickel coating intact over most of the corroded area; in two of the wafers the corrosion penetrated the 0.040-in.-thick wafers, Figure 11. Approximately 0.09 g of cobalt was lost from each wafer. This rate is apparently faster than the 0.0007 in./yr (0.1 g/yr/in^2 of surface area) that was observed after 4-yr tests with panels cut from cobalt electrodes.[7] A direct comparison of the two tests is difficult because the exposed surface area of the cobalt wafers was constantly changing. The small size of the defects in the nickel coating (<0.005-in.-dia) may have restricted the supply of oxygen to the corroding surfaces, leading to accelerated attack, as in crevice corrosion.

*Trademark of E. I. du Pont de Nemours & Co.

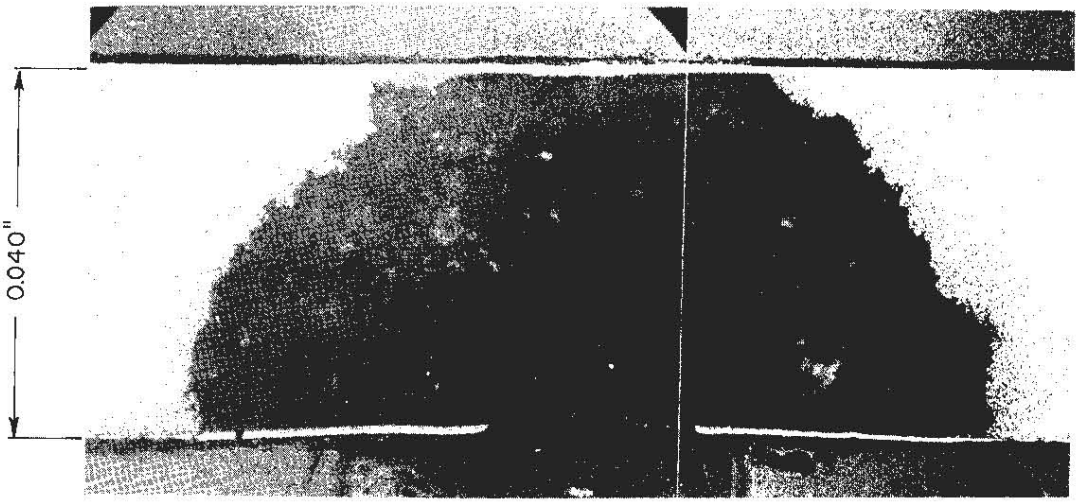

FIG. 11 SEAWATER CORROSION OF COBALT AT DEFECT IN NICKEL COATING

Recommended limits on release rates of radioactive wastes from nuclear powered ships during normal operation range from 0.01 to 100 Ci/day, depending on closeness to shore and ocean depth.[8] For a specific activity of 350 Ci/g, corrosion under the abnormal conditions of the current tests could release ∿1.0 Ci of ^{60}Co/day. Additional tests of the effects of sea water corrosion are being planned.

Compatibility of Molten Cobalt and Superalloys with Tungsten and Rhenium

Tests of compatibility of molten cobalt and superalloys with tungsten and rhenium will be continued to evaluate the consequences of temperature excursions above the melting points of the capsules (∿1500°C) that might occur under abnormal conditions, such as loss of coolant, re-entry from space, or burial in the earth.[2]

Five additional tests with tungsten capsules will be started as soon as the new welding facility is completed (see Development of Welding Techniques for Refractory Metals). These capsules will be heated for 4 hr at 1550 to 1750°C to evaluate the effects of a void space and superalloys inside the tungsten capsule and the effectiveness of a rhenium barrier between the cobalt and the tungsten.

HEAT SOURCE DEMONSTRATION TESTS

WANL 30 kw(t) Demonstration Unit

SRL is providing technical assistance as necessary in the program to design, develop, fabricate, and test an experimental ^{60}Co-fueled 30 kw(t) heat source. Westinghouse Astronuclear Laboratory (WANL) is the contractor in this program to demonstrate the principle of production of useful energy from a large ^{60}Co source.

Tests were planned to verify calculations of radiation streaming through ducts in the biological shield. These tests will be run at SRL, using an available 0.115 curie source of ^{60}Co and CaF_2 thermoluminescent detectors. The shield block for the test is scheduled for delivery in December.

Nickel was selected by WANL as the reference core material because of its high thermal conductivity, and nitrogen as the preferred coolant for compatibility at up to 900°C. Work is continuing to evaluate air as a possible coolant, and SRL is participating in a survey of coatings for oxidation protection of the nickel.

HIGH ACTIVITY ^{60}Co FOR HEAT SOURCE DEVELOPMENT

High activity cobalt that is being stored at Savannah River for experimental programs and heat source development is listed in Table VII. Estimated production in current charges is listed in Table II.

TABLE VII

^{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-in. Wafers				
Fuel form - cobalt metal (wrought)	310	2090	1.60	25.0
Fuel shape - wafers 0.745 ±0.001-in.-dia	270	912	0.62	9.6
0.040 ±0.003-in.-thick	250	1368	0.87	13.6
including 0.0005 to 0.001-in. Ni plate	230	1368	0.79	12.3
Cobalt density - 8.80 ±0.05 g/cm^3	240	152	0.09	1.4[b]
		5890	3.97	61.9
0.800-in. Wafers				
Fuel form - cobalt metal (sintered)	300	255	0.22	3.4
Fuel shape - wafers 0.800 ±0.001-in.-dia	320	136	0.13	2.0[c]
0.040 ±0.003-in.-thick	270	136	0.10	1.6[d]
including 0.0005 to 0.001-in. Ni plate		527	0.45	7.0
Cobalt density - 8.60 ±0.10 g/cm^3				
Nickel-Plated Slabs				
Fuel form - cobalt metal (wrought)	300	45	0.23	3.6
Fuel shape - slabs 3.00 ±0.03-in.-long	280	48	0.22	3.4
0.640 ±0.002-in.-wide		93	0.45	7.0
0.060 ±0.001-in.-thick				
including 0.0005 to 0.001-in. Ni plate				
Cobalt density - 8.80 ±0.05 g/cm^3				
Stainless Steel-Canned Slabs				
Fuel form - cobalt metal (wrought)	300	45	0.18	2.8
Fuel shape - slabs 2.96 ±0.03-in.-long	250	48	0.17	2.7
0.735 - 0.740-in.-wide		93	0.35	5.5
0.092 ±0.001-in.-thick				
sheath thickness 0.015-in. min				
cobalt dimensions same as nickel-plated slabs above except 2.44-in.-long				
Cobalt density - 8.80 ±0.05 g/cm^3				
Stainless Steel-Coextruded Slabs				
Fuel form - cobalt metal (wrought)	300	60	0.21	3.3
Fuel shape - slabs 3.00 ±0.03-in.-long	280	64	0.21	3.3
0.740 ±0.002-in.-wide		124	0.42	6.6
0.072 ±0.002-in.-thick				
SST thickness 0.015-in. min				
Cobalt dimensions: 2.75-in.-long				
0.71-in.-wide				
0.042-in.-thick				
Cobalt density - 8.80 ±0.05 g/cm^3				
Grand Total			5.64 MCi	88.0 kw
Average Activity	280 Ci/g			

[a] Activity and power as of June 30, 1969.
[b] Wafers have central hole of 0.070-in.-dia.
[c] 110 wafers have experimental compositions, to be used at SRL.
[d] 68 wafers have central hole of 0.070-in.-dia.

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

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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
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