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AEC RESEARCH AND DEVELOPMENT REPORT

SAVANNAH RIVER LABORATORY COBALT-60 POWER AND HEAT SOURCES

QUARTERLY PROGRESS REPORT

OCTOBER-DECEMBER 1969

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Savannah River Laboratory

Aiken, South Carolina

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October - December 1969

Compiled by

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March 1970

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SAVANNAH RIVER LABORATORY
AIKEN, S. C. 29801

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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

High reaction rates between cobalt metal and the capsule walls observed in two capsules containing radioactive cobalt metal were caused by NaOH residue on the cobalt wafers. (p 2)

Heating for 10,000 hr at 1000°C was completed on radioactive capsules of "Inconel" 600, "Hastelloy" C, and "Hastelloy" X. (p 7)

Minor amounts of ^{60}Co were released when a capsule with an intentional defect was heated for 5000 hr at 1000°C. (p 12)

Fabrication processes were established for wafers of CoO, $Co_{0.5}Mg_{0.5}O$, and $CoAl_2O_4$, and irradiation of 120 wafers of each compound was begun. (p 14)

High activity cobalt metal is being produced for experimental purposes and the AEC's ^{60}Co loan program. (p 21)

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 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

CAPSULE MATERIALS FOR RADIOACTIVE COBALT

Oxidation of Superalloys

Microprobe analyses were begun on the oxide scale and subsurface alloy-depletion zones of selected specimens from the long-term oxidation tests.[1] In contrast to most literature data, manganese (a minor constituent in most superalloys) is present in the outer oxide layer of "Inconel"* 600, "Hastelloy"** C and "Hastelloy" X. This layer is believed to be largely responsible for the excellent resistance to oxidation of the superalloys. Chromium was the major constituent in the second layer of the scale, in agreement with previous X-ray diffraction identification of Cr_2O_3 as a major part of the scale.[1] The zone beneath the scale was correspondingly depleted in these alloying elements. A third, innermost layer rich in tungsten and silicon was found on "Hastelloy" C. A topical report on the results of the oxidation tests, including the microprobe data, is being prepared.

Compatibility of Cobalt Compounds with Capsule Materials

Early results of short-term tests confirm the predicted compatibility of the cobalt oxides with rhenium and their incompatibility with other refractory metals.[2] CoO and $CoAl_2O_4$ reacted extensively with their tungsten capsules while at 1620°C for 260 hr. Identical tests have been completed with three TZM*** alloy capsules each containing one of the oxides CoO, $Co_{0.5}Mg_{0.5}O$ and $CoAl_2O_4$. These capsules will be examined metallographically to determine the extent of reaction.

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* Trademark of International Nickel Co.
** Trademark of Union Carbide Corp.
*** Product of Climax Molybdenum Co.

CAPSULE FABRICATION AND TESTING

Adaptation of Welding Techniques for Refractory Metal Capsules

Refractory metal capsules and welding equipment are being procured for encapsulation of cobalt metal and cobalt oxides for applications between 1000 and 1850°C. Twenty tungsten capsules and ten tungsten-25 wt % rhenium capsules for compatibility tests with ^{59}Co metal, and 12 rhenium capsules for compatibility tests with ^{59}Co oxides, have been procured.

Installation of a tungsten-inert-gas (TIG) welding station was completed in the Alpha Materials Facility (AMF). Capsules of tungsten, rhenium, and TZM alloy were welded in a high-purity helium atmosphere. No leaks were detected by helium leak tests, although some cracking was observed in one tungsten capsule. Weld development is continuing to establish optimum parameters and to eliminate cracking of tungsten. A more satisfactory means for heat removal from the weld area will also be developed to prevent overheating of the capsule contents.

An inert gas welding station is being designed for installation in the High Level Caves (HLC) for encapsulating ^{60}Co metals and oxides.

Heating Tests of Capsules Containing Unirradiated Cobalt Metal

Superalloy Capsules

A test showed that the rapid reaction between cobalt and "Inconel" 600 that was observed in two radioactive capsules after 10,000 hr at 900°C[2] was due to a caustic residue on the wafers. This reaction was duplicated with unirradiated wafers intentionally coated with a caustic residue and heated in an "Inconel" 600 capsule for 5000 hr at 900°C.[3] The residue was produced by dissolving an unirradiated aluminum target slug in 4M NaOH solution, as is done after irradiation to dejacket the wafers, Figure 1. The wafers were removed from the solution and allowed to dry in air without the rinsing or cleaning in oxalic acid that is normally used for irradiated wafers. At the time the two ^{60}Co capsules exhibiting the extensive reaction were fabricated, the wafers remained in the NaOH solution until all the aluminum was dissolved. Wafers encapsulated since then remained in the solution only until the cladding on the slug was dissolved and the D-bars could be separated to free the wafers for removal from the caustic.

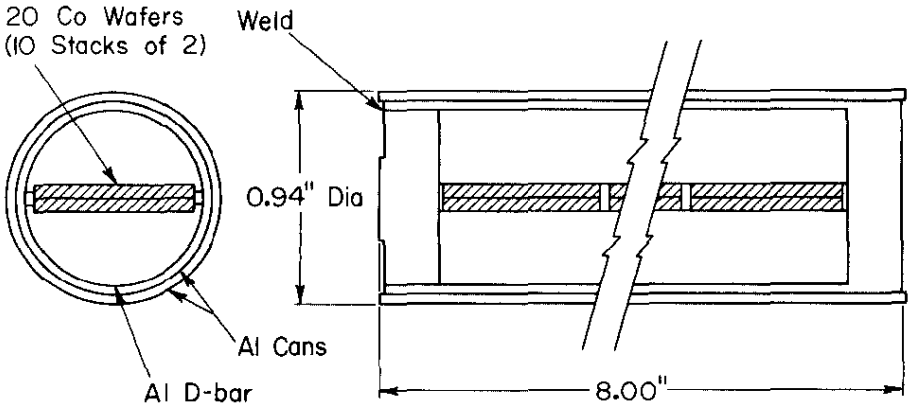


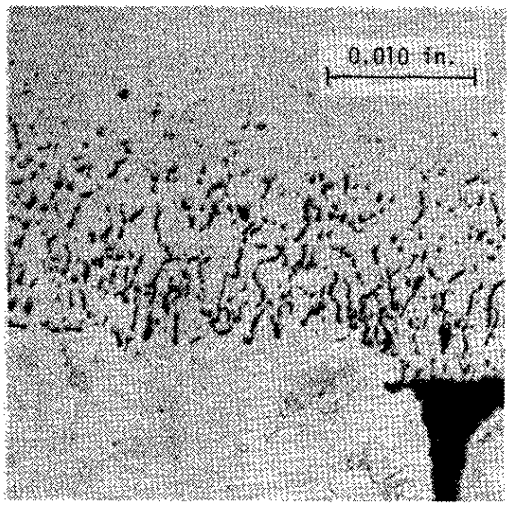
FIG. 1 ALUMINUM TARGET SLUG FOR COBALT METAL WAFERS

The appearance of the reaction zone in the unirradiated capsule was the same as that in the irradiated capsules, Figure 2. The average width of the reaction zone in the ^{59}Co capsule was 0.016 in. compared to the width of 0.018 in. observed in the ^{60}Co capsules after 10,000 hr. In contrast reaction zones 0.008- to 0.010-in. wide were observed in previous 5000-hr tests with clean ^{59}Co wafers and clean ^{60}Co wafers. The zones in the capsules with the NaOH residue contained voids and oxide particles along the grain boundaries of the "Inconel" 600. Microprobe analyses showed aluminum was concentrated in these grain boundaries. Aluminum is present as an impurity in both the cobalt and the "Inconel" 600, but not in sufficient concentrations to account for the aluminum observed in the reaction zone.

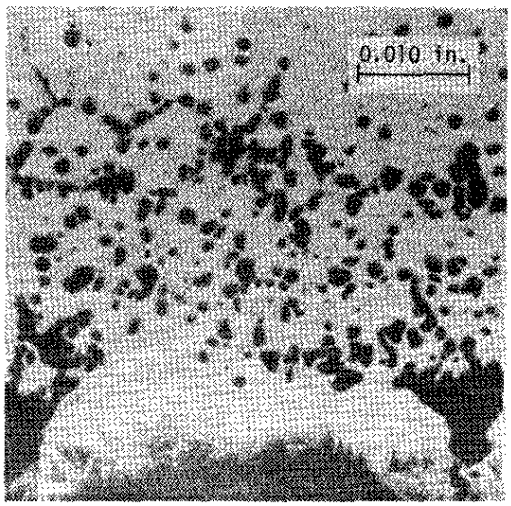
Heating of a similar capsule containing unirradiated, coated wafers for 10,000 hr at 900°C is continuing. Heating of 16 other capsules at 900 and 1000°C to demonstrate their performance is also continuing, Table I.

Refractory Metal Capsules

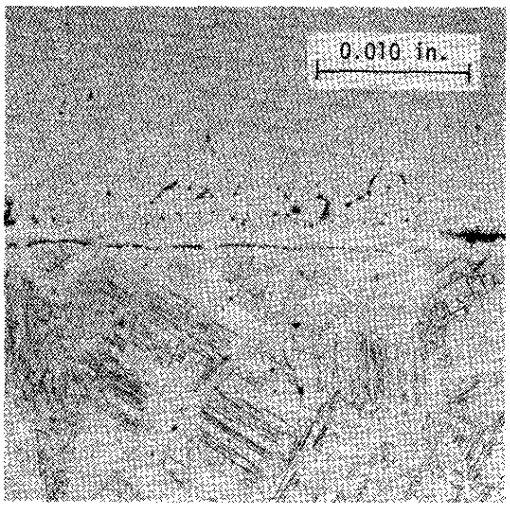
Tests with unirradiated cobalt in tungsten and W-Re alloy will be started in the coming quarter, as soon as welding techniques required for the encapsulation have been developed, Table II. Initial long-term heating tests of rhenium capsules will be postponed because of a delay in delivery of the capsules. The delivery date has not yet been established by the vendor.



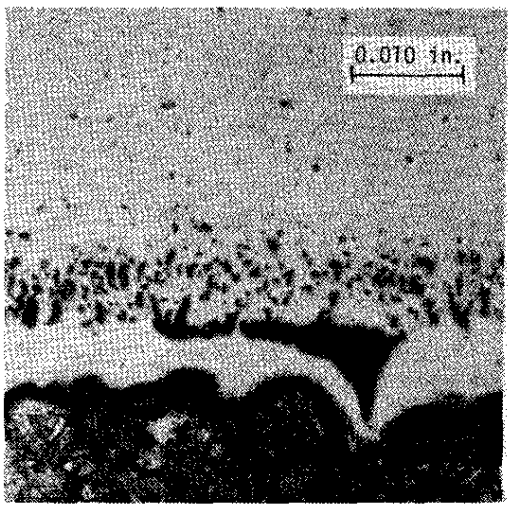
a. Residue on ^{59}Co wafers. Heated 5000 hr at 900°C.



b. Residue on ^{60}Co Wafers. Heated 10,000 hr at 900°C.



c. Clean ^{59}Co wafers. Heated 5000 hr at 900°C.



d. Clean ^{60}Co wafers. Heated 5000 hr at 900°C.

FIG. 2 EFFECT OF CAUSTIC RESIDUE ON Co-"INCONEL" 600 COMPATIBILITY

TABLE I

Summary of ^{59}Co Capsule Heating Tests
All Co wafers 0.745-in. diameter

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	Increased Co/capsule reaction
	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.
→ New information reported.

TABLE II

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

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

→New information reported.

Heating Tests of Capsules Containing Irradiated Cobalt Metal

Superalloy Capsules

Heating tests for 10,000 hr at 1000°C were completed on one capsule each of "Hastelloy" C, "Inconel" 600, "Hastelloy" X, and "Haynes"* 25. Heating of duplicate capsules of the latter three materials will be continued to 20,000 hr, unless examination of the other capsules indicates unsatisfactory performance. Testing was also completed on one capsule each of "Inconel" 600 and "Haynes" 25 containing ^{59}Co, but heated in the same furnaces as the ^{60}Co capsules. The surface temperature of these capsules was between 860°C, the temperature of the furnace atmosphere, and 900°C, the temperature of the ^{60}Co capsule surfaces.

Examination of these capsules and those previously removed from test is expected to begin in March, instead of January as originally scheduled. Refurbishment of the cell and installation of new equipment (funded by the Division of Production) to be used for metallography has begun.

Heating of the nine remaining capsules at 900 and 1000°C to demonstrate capsule performance is continuing, Table III. These tests have been consolidated into two furnaces, one at each temperature, instead of the four originally used. A thermocouple was spot-welded to one of the remaining capsules being heated at 900°C to permit recalibration of the furnace controller. The previous controller setting on the 1000°C furnace was maintained because the change in total activity in the furnace was negligible. Added protection against overtemperatrues in these furnaces is provided by backup controllers operated by independent thermocouples and set ∿20°C above the normal operating temperature. Settings and operability of the backup controllers are verified weekly.

* Trademark of Union Carbide Corp.

TABLE III

Summary of ^{60}Co Capsule Heating Tests

All Co wafers 0.745-in. diameter

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 tc 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 3-70
	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 3-70
	→ 10,000	1,000	95	1	288[c]	13,700	9-68	11-69	Begin destructive examination 3-70
	→ 20,000	1,000	95	1	263[c]	12,500	9-68	1-71	
	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 3-70
	→ 10,000	1,000	95	1	282[c]	13,400	9-68	11-69	Begin destructive examination 3-70
	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 3-70
(m.p. 1330°C)	→ 10,000	1,000	95	1	288[c]	13,700	9-68	11-69	Begin destructive examination 3-70
	→ 20,000	1,000	95	1	282[c]	13,400	9-68	1-71	
	50,000	1,000	95	1	295[c]	14,000	9-68	5-74	
	→ 10,000	850	95	1	d	-	9-68	11-69	Begin destructive examination 3-70
"Hastelloy" X	→ 5,000	1,000	95	1	250[c]	11,900	9-68	4-69	Begin destructive examination 3-70
(m.p. 1260°C)	→ 10,000	1,000	95	1	263[c]	12,500	9-68	11-69	Begin destructive examination 3-70
	→ 20,000	1,000	95	1	263[c]	12,500	9-68	1-71	
	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 was heated along with ^{60}Co capsules.
→New information reported.

Refractory Metal Capsules

Capsule tests with irradiated cobalt in refractory metal capsules are scheduled to start in January 1971, Table IV. Design of a shielded facility in the Isotope Process Development Laboratory (IPDL) for these tests is essentially complete, and construction of the cell has started. Preparation of a Project Authorization Request for operating and process equipment in the cell is continuing.

Surface temperatures were measured on capsules with internal (electric) heat generation in a vacuum furnace that will be used for tests of refractory metal capsules. The measurements indicated that the planned capsules will not overheat, but that capsule surface temperatures may be up to 350°C higher than the indicated furnace temperature. Development is continuing on methods to measure surface temperatures of refractory alloy capsules containing ^{60}Co.

TABLE IV

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

Capsule Material	Heating		Approx. Starting Time	Approx. Completion Date	Wafer Diameter, inch
	Time, hr	Temp, °C			
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.

Heating Tests of Capsules Containing Unirradiated Cobalt Compounds

Wafers of CoO, $Co_{0.5}Mg_{0.5}O$, and $CoAl_2O_4$ and capsules of rhenium are being fabricated for tests up to 50,000 hr at 1500 to 1850°C. Materials for nine capsules containing $CoAl_2O_4$and $Co_{0.5}Mg_{0.5}O$ wafers have been procured. These tests should confirm the predicted compatibility between the oxides and rhenium beyond the previous 200-hr tests at 1525°C.[4] Schedules for long-term heating tests of ^{59}Co and ^{60}Co oxides are presented in Tables V and VI.

TABLE V

^{59}Co High Temperature Fuels Compatibility Program

Capsule Material (and Temp)	Fuel Material	Heating Time, hr	Heating Startup	Heating Terminated	Remarks
		Preliminary Tests			
"Inconel" 600 Foils of Ir, Rh, Re, Pt (1200°C)	$Co_{0.5}Mg_{0.5}O$	200	5/69	6/69	Oxide reduced by "Inconel" 600
"Inconel" 600 Foils of Ir, Rh, Re, Pt (1200°C)	$Co_{0.5}Mg_{0.5}O$	500	6/69	7/69	Oxide reduced by "Inconel" 600
Rhenium (1525°C)	CoO	200	7/69	7/69	No gross interaction
Rhenium (1525°C)	$Co_{0.5}Mg_{0.5}O$	200	7/69	7/69	No gross interaction
Rhenium (1525°C)	$CoAl_2O_4$	200	7/69	7/69	No gross interaction
Tungsten(1620°C)	CoO	260	12/69	12/69	Reaction
Tungsten(1620°C)	$CoAl_2O_4$	260	12/69	12/69	Reaction
TZM (1620°C)	CoO	260	12/69	12/69	Completed
TZM (1620°C)	$Co_{0.5}Mg_{0.5}O$	260	12/69	12/69	Completed
TZM (1620°C)	$CoAl_2O_4$	260	12/69	12/69	Completed
		1500°C Tests			
Molybdenum	CoO	1,000	6/70	8/70	
Molybdenum	$Co_{0.5}Mg_{0.5}O$	1,000	6/70	8/70	
Molybdenum	$CoAl_2O_4$	1,000	6/70	8/70	
Rhenium; foils of Ir, Rh	CoO	1,000	3/70	5/70	
Rhenium; foils of Ir, Rh	$Co_{0.5}Mg_{0.5}O$	1,000	3/70	5/70	
Rhenium; foils of Ir, Rh	$CoAl_2O_4$	1,000	3/70	5/70	
Iridium	CoO	1,000	6/70	8/70	
Iridium	$Co_{0.5}Mg_{0.5}O$	1,000	6/70	8/70	
Iridium	$CoAl_2O_4$	1,000	6/70	8/70	
W-25 Re	CoO	1,000	6/70	8/70	
W-25 Re	$Co_{0.5}Mg_{0.5}O$	1,000	6/70	8/70	
W-25 Re	$CoAl_2O_4$	1,000	6/70	8/70	
Rhenium	CoO	1,000	2/70	4/70	Capsule machined
Rhenium	$Co_{0.5}Mg_{0.5}O$	1,000	2/70	4/70	Capsule machined
Rhenium	$CoAl_2O_4$	1,000	2/70	4/70	Capsule machined
Rhenium	CoO	5,000	3/70	10/70	
Rhenium	$Co_{0.5}Mg_{0.5}O$	5,000	2/70	9/70	Capsule machined
Rhenium	$CoAl_2O_4$	5,000	3/70	10/70	
Rhenium	Best Oxide	10,000	6/70	8/71	
Rhenium	Best Oxide	50,000	6/70	2/76	

TABLE V, Continued

Capsule Material	Fuel Material	Heating Time, hr	Heating Startup	Heating Terminated	Remarks
		1700°C Tests			
Rhenium	CoO	1,000	2/70	4/70	Capsules machined
Rhenium	$Co_{0.5}Mg_{0.5}O$	1,000	2/70	4/70	Capsules machined
Rhenium	$CoAl_2O_4$	1,000	2/70	4/70	Capsules machined
Rhenium	CoO	5,000	3/70	10/70	
Rhenium	$Co_{0.5}Mg_{0.5}O$	5,000	3/70	10/70	
Rhenium	$CoAl_2O_4$	5,000	3/70	10/70	
Rhenium	Best Oxide	10,000	6/70	8/71	
Rhenium	Best Oxide	50,000	6/70	2/76	
Iridium	CoO	1,000	6/70	8/70	
Iridium	$Co_{0.5}Mg_{0.5}O$	1,000	6/70	8/70	
Iridium	$CoAl_2O_4$	1,000	6/70	8/70	
		1850°C Tests			
Rhenium	$Co_{0.5}Mg_{0.5}O$	1,000	2/70	4/70	Capsule machined
Rhenium	$CoAl_2O_4$	1,000	2/70	4/70	Capsule machined
Rhenium	$Co_{0.5}Mg_{0.5}O$	5,000	3/70	10/70	
Rhenium	$CoAl_2O_4$	5,000	3/70	10/70	
Rhenium	Best Oxide	10,000	6/70	8/71	
Rhenium	Best Oxide	50,000	6/70	2/76	
Iridium	$Co_{0.5}Mg_{0.5}O$	1,000	6/70	8/70	
Iridium	$CoAl_2O_4$	1,000	6/70	8/70	

TABLE VI

^{60}Co High Temperature Fuels Compatibility Program

Capsule[a] Material	Fuel Material	Heating Time, hr	Heating Startup	Heating Terminated	Total[b] Activity, Ci	Thermal[b] Energy, watts
		1500°C Tests				
Rhenium	CoO	5,000	1/71	8/71	10,700	160
Rhenium	$Co_{0.5}Mg_{0.5}O$	5,000	1/71	8/71	5,500	82
Rhenium	Best Oxide	10,000	1/71	3/72	5,500	82
Rhenium	Best Oxide	10,000[c]	1/71	3/72	5,500	82
Rhenium	Best Oxide	50,000	1/71	10/76	5,500	82
		1850°C Tests				
Rhenium	Best Oxide	1,000	1/71	3/71	5,500	82
Rhenium	Best Oxide	5,000	1/71	8/71	5,500	82
Rhenium	Best Oxide	10,000	1/71	3/72	5,500	82
Rhenium	Best Oxide	10,000[c]	1/71	3/72	5,500	82
Rhenium	Best Oxide	50,000	1/71	10/76	5,500	82

a. Assumes rhenium is satisfactory capsule after 1000-hr tests on ^{59}Co, otherwise capsule material will be iridium liner in rhenium.

b. Based on ^{60}Co specific activity of 400 Ci/g; assumes $Co_{0.5}Mg_{0.5}O$ for "Best Oxide."

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

SAFETY TESTS

Cobalt Release

The release of ^{60}Co through a defect in the capsule wall was measured while an "Inconel" 600 capsule was heated in flowing air at 1000°C for 5000 hr. The capsule had a 0.008-in.-diameter hole drilled through the wall and contained a solid rod of unirradiated cobalt that had been plated with ^{60}Co. An amount of ^{60}Co corresponding to 7 c/m above background was deposited each day on filter paper in the effluent air stream. The cumulative amount of ^{60}Co released followed a parabolic relationship with time during the first 50 days of the test, but became linear with time after 80 days, Figure 3. This linear rate of release was 6.5×10^{-2} µg/hr. If a heat source capsule containing 350 Ci/g of ^{60}Co had a similar defect, 180 µCi would be released in 8 hr; the maximum permissible intake during 8 hr is 370 to 470 µCi for a dose of 0.3 rem in one week.[5]

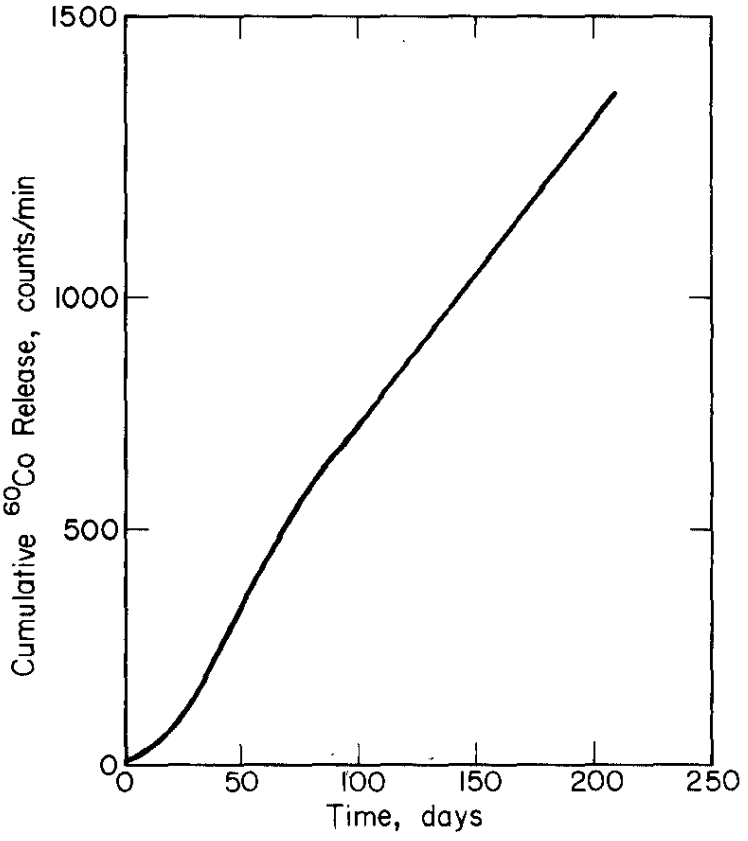


FIG. 3 ^{60}Co RELEASE THROUGH PINHOLE IN CAPSULE WALL

Metallographic examination of the capsule is in progress to explain the cause of the change in release rate. This change may be related to the oxidation of the cobalt and the "Inconel" in the vicinity of the defect, as previously observed.[6] Particles of ^{60}Co released from the capsule and deposited on the filter paper will be identified and their sizes measured by electron diffraction and microscopy. These data will be needed for further evaluation of the biological hazards associated with cobalt.

Compatibility Above the Cobalt Melting Temperature

Tests of compatibility of molten cobalt and superalloys with tungsten and rhenium are continuing to evaluate the consequences of temperatue excursions, producing melting of capsule components, that might occur under abnormal conditions, such as loss of coolant, re-entry from space, or burial in the earth.

Heating tests were run with three tungsten capsules to measure the compatibility of molten cobalt with tungsten. These capsules have different internal void volumes; initial tests showed appreciable dissolution of the tungsten only at the void space at the top of the capsules.[3] The capsules were sealed by TIG welding, checked with a helium leak detector, and heated for 4 hr at 1550°C. Examination of the capsules is in progress. Additional capsules will be tested to establish the effect of superalloys on compatibility and the effectiveness of rhenium as a barrier layer, as soon as the effect of the void space is explained. Tests at higher temperatures will also be started.

Parametric Study

A parametric study was begun to identify temperatures and other conditions that are possible under a wide variety of accident conditions in simple ^{60}Co heat sources. The ANISN code is being used for shielding and energy deposition calculations and the TOSS code for temperature profile calculations. The safety test program will be developed, based on consultation with personnel from Sandia Laboratories and others, to complement the parametric study. The test program will identify limiting conditions that ^{60}Co heat sources can withstand and the mechanisms of the reactions between ^{60}Co fuel forms, capsule materials, and the environment. For example, tests will consider reactions of cobalt fuel forms and capsule materials with air, soil, water, and other capsule and shield materials, up to and beyond the melting points of the materials involved.

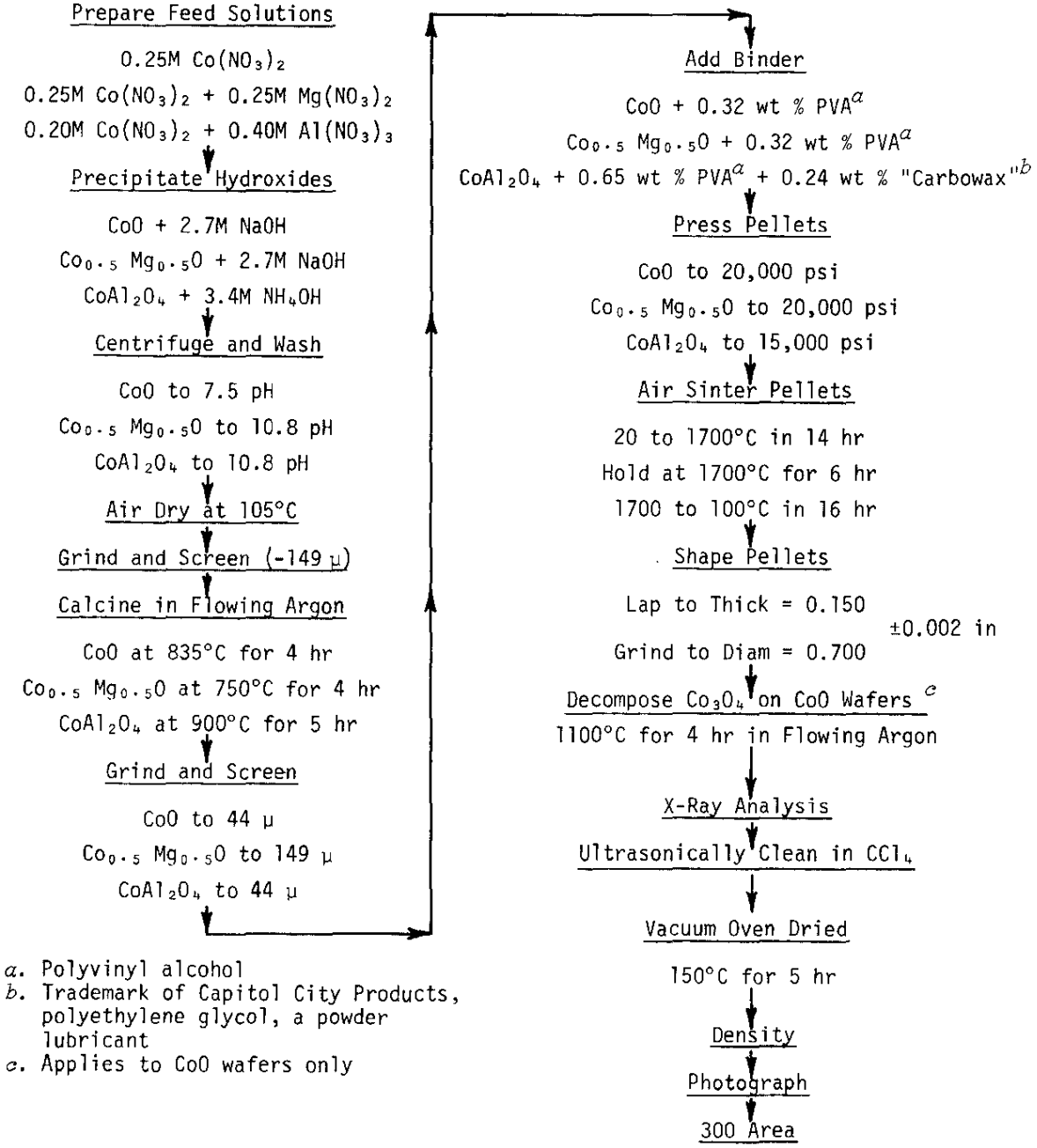
HIGH-TEMPERATURE FUEL FORMS

High-melting forms of radioactive cobalt are being considered for possible high-temperature applications, including thermionic systems (>1500°C), and for lower temperature applications for which they might provide increased safety over cobalt metal. The fuel forms include CoO, CoO-MgO solid solutions, $CoAl_2O_4$, and Co-Ru and Co-Re alloys. The metallic forms are less attractive than the oxidized forms; Co-Ru has a low power density and Co-Re must be fabricated after irradiation.[4]

Fabrication of Oxide Wafers

One hundred twenty wafers each of CoO, $Co_{0.5}Mg_{0.5}O$, and $CoAl_2O_4$ were fabricated by cold-pressing and sintering. Irradiation of these wafers in high-flux reactor changes was started to demonstrate production and to provide ceramic ^{60}Co fuel for characterization and compatibility tests. A total activity of >400,000 Ci will be produced in these wafers. Specific activities are expected to be over 450 Ci/g of Co. The densities of the $CoAl_2O_4$ and $Co_{0.5}Mg_{0.5}O$ wafers vary between 88 and 96% of theoretical; densities of the CoO wafers are between 80 and 90% of theoretical.

Processes, shown in the flow chart in Figure 4, for fabricating wafers of $Co_{0.5}Mg_{0.5}O$, $CoAl_2O_4$ and CoO were established. Yields of nearly 100% were achieved for $CoAl_2O_4$ and $Co_{0.5}Mg_{0.5}O$ wafers but only ∿50% of the CoO wafers were acceptable because of cracking during sintering.



a. Polyvinyl alcohol
b. Trademark of Capitol City Products, polyethylene glycol, a powder lubricant
c. Applies to CoO wafers only

FIG. 4 OXIDE WAFER FABRICATION PROCEDURE

Hydroxide Precipitation

The oxide powders were prepared by calcining the desired hydroxide precipitate. Cobalt and aluminum hydroxide were coprecipitated by adding 3.4M NH_4OH to a solution of 0.2M $Co(NO_3)_2$ and 0.4M $Al(NO_3)_3$. Cobalt or cobalt-magnesium hydroxides were precipitated by adding 2.7M NaOH to 0.25M $Co(NO_3)_2$ or to a solution of 0.25M $Co(NO_3)_2$ and 0.25M $Mg(NO_3)_2$. Although ammonia is preferred to NaOH to avoid differential grain growth during sintering caused by residual sodium, NaOH was used for the precipitations from cobalt and cobalt-magnesium solutions. Apparently, a sufficient OH^- concentration could not be reached with NH_4OH to cause precipitation of $Co(OH)_2$.

The precipitate was centrifuged and the supernate was decanted, then the precipitate was washed and recentrifuged until a pH of 7.5 for CoO and 10.8 for $Co_{0.5}Mg_{0.5}O$ and $CoAl_2O_4$ was achieved. The precipitates could not be satisfactorily separated by the centrifuge if washing was carried to lower values of pH.

Calcining to Oxide

The precipitates were dried ~16 hr in air at 105°C, ground, and screened through a 100 mesh (-149 μ) sieve. The dried precipitate was calcined under flowing argon in Al_2O_3 dishes at 835°C for CoO, 750°C for $Co_{0.5}Mg_{0.5}O$, and 900°C for $CoAl_2O_4$. These calcining temperatures are the lowest temperatures that will decompose the hydroxide and ensure compound formation, and yet minimize Co_3O_4 formation and preserve small grain size and powder texture for subsequent sintering. The calcined powder was furnace cooled in flowing argon to prevent oxidation to Co_3O_4, and then ground, and screened to break up agglomerates and produce the desired particle size for sintering to high density. The binders, as indicated in Figure 4, were mixed with the calcined powders by kneading in sealed plastic bags for about 20 minutes.

Pressing and Sintering

The powder was pressed in a 0.950-in.-diameter die. The diameter of the die and amount of oxide were selected to accommodate shrinkage during sintering and to produce a sintered wafer 10% oversize for final lapping and grinding to close tolerances. The die was lubricated prior to loading with a 3 wt % solution of stearic acid in carbon tetrachloride.

Pressing was performed in three steps at 6,500 psi, 13,000 psi, and finally 15,000 or 20,000 psi, with the load released between steps to allow occluded gas to escape from the wafers and minimize laminations. The pressure was released slowly after each step to prevent breaking the wafers as the die contracts. The wafers were also ejected slowly to prevent edge breakage.

The cold-pressed wafers were stacked in alumina dishes for sintering. They were slowly heated in air to 1650-1700°C over a period of 14 hr, held at temperature for 6 hr, and furnace cooled in 16 hr to ∿100°C. Powder of the same composition as the wafers was sprinkled on the bottom of the alumina dishes to prevent reaction with the Al_2O_3, and on top of and between the stacked wafers to reduce vaporization of cobalt oxide from the wafer surface. The stacked wafers were enclosed in Al_2O_3 by inverting a second, smaller dish over the stacked wafers and pouring Al_2O_3 powder into the gap between the dishes. While this system prevented preferential loss of cobalt oxide, the dishes were severely impregnated with the loose cobalt powder and could be used only once or twice.

Finishing

The wafers were ground and lapped to the required dimensions (0.150 ±.002 in. thickness x 0.700 ±0.002 in. diameter). Any Co_3O_4 formed during sintering of the CoO wafers was decomposed to CoO by heating in flowing argon at 1100°C for 4 hr. Finally, the wafers were cleaned ultrasonically in CCl_4 and dried at 150°C for 8 hr.

Evaluation of Wafers for Irradiation

Each wafer was measured, weighed, and photographed to characterize it and to provide a comparison with postirradiation appearance. Selected wafers were analyzed by X-ray diffraction for unreacted components, Co_3O_4, impurity phases, and compositional variations. Other wafers were examined metallographically for grain size gradients, microcracks, and impurity phases to provide a comparison with postirradiation microstructures.

The $Co_{0.5}Mg_{0.5}O$ wafers being irradiated have high densities and are free of defects; 64 of the $CoAl_2O_4$ wafers are laminated and 20 of the CoO wafers have minor surface cracks, Table VII. Typical examples of these defects are illustrated in Figure 5.

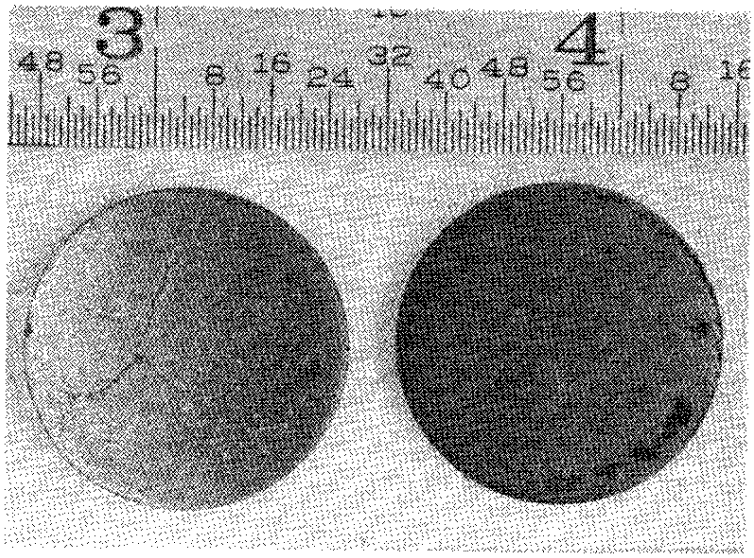
The laminations and the resulting variations in density occurred in $CoAl_2O_4$ wafers made early in the development of the process. Changes in the binder and in pressing techniques during the production run produced more uniform and higher (90 to 95%) density wafers free of laminations and cracks.

Cracking and low density were persistent problems with CoO wafers. Cracking was apparently caused by thermal shock, coupled with a grain size gradient. Low densities may be caused by large particles in the green wafers which resulted in lower sintering rates and larger grains in the sintered wafer. Work is continuing to reduce cracking and increase density.

TABLE VII

Density of Cobalt Oxide Wafers for Irradiation to High Specific Activity

% of Theoretical Density	Numbers of Wafers		
	CoO	$Co_{0.5}Mg_{0.5}O$	$CoAl_2O_4$
90 - 95	-	115	90
85 - 90	40	5	30
80 - 85	80	-	-
Total	120	120	120
Theoretical Density, g/cm^3	6.437	5.024	4.416



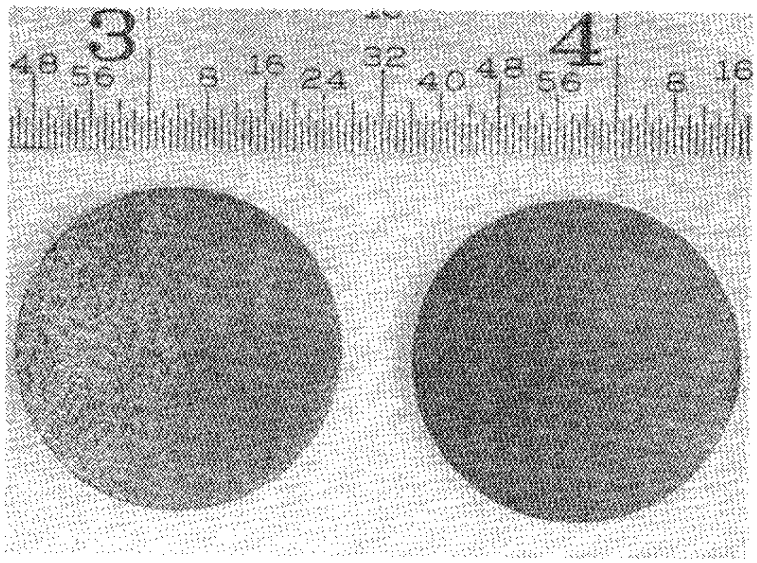


FIG. 5 TYPICAL DEFECTS IN CoO AND $CoAl_2O_4$ WAFERS

HEAT SOURCE DEMONSTRATION TESTS

WANL 30 kw(t) UNIT

SRL is providing technical assistance in the program to design, fabricate, and test an experimental heat source containing 30 kw(t) of ^{60}Co. Westinghouse Astronuclear Laboratory completed the preliminary design phase of this program in December and is proceeding with detailed design.

A steel shield block was machined at SRL for tests to verify calculations of radiation streaming through penetrations in the biological shield. These penetrations simulate inlet and outlet ports for the primary nitrogen gas coolant, openings for the heat pipes provided for emergency cooling, and refueling ports for the ^{60}Co fuel pins. Tests with a 0.115 curie source of ^{60}Co and CaF_2 and LiF thermoluminescent detectors will be made during the next quarter.

SRL 3 kw(t) UNIT

Authorization was received from the Division of Production for an early demonstration of the feasibility of using available ^{60}Co as a heat source for thermoelectric power generation using a tubular thermoelectric module provided by WANL. This type of module is being developed by WANL under AEC contract.

Design will be completed and the test program will be written in the next quarter. Objectives of the test are to confirm that

- ^{60}Co sources can be assembled easily in a hot cell and than transported and used safely in inhabited areas,
- Operating temperatures are low enough for practical long-term use, and
- Performance is predictable as the ^{60}Co decays.

HIGH ACTIVITY ^{60}Co FOR HEAT SOURCE DEVELOPMENT

High activity cobalt metal that is being stored or produced at Savannah River for experimental programs and heat source development is listed in Table VIII. This material will be available under the AEC's new ^{60}Co loan program.

TABLE VIII

Savannah River High Activity ^{60}Co for Heat Source Development

Cobalt Shape							
Diameter, in.	Thickness, in.	No. of Pieces	Wt. of Cobalt, g/piece	Average Specific Activity, Ci/gCo	Total Activity, MCi	Total Power, kw(t)	Date Available (Activity calculated as of this date)
Wafers, Ni-plated							
0.745	0.040	2090	2.5	270	1.41	22.0	Before 6-70
		3800[a]	2.5	220	2.09	32.5	Before 6-70
0.745	0.057	112	3.5	450	0.18	2.8	6-70
0.745	0.073	3240	4.5	450	6.56	102.1	6-70
		2660	4.5	450	5.38	83.8	3-71[c]
		1700	4.5	250	1.91	29.7	6-70
		4560	4.5	200	4.10	63.8	3-71
0.800	0.040	391[b]	2.8	250	0.27	4.3	Before 6-70
Half wafers, Ni-plated							
1.00	0.073	682	4.1	450	1.26	19.6	3-71[c]
1.25	0.073	434	6.4	450	1.26	19.6	3-71[c]
1.49	0.073	620	9.1	450	2.52	39.2	3-71[c]
Slabs							
3.00 x 0.64 x 0.060" Ni-plated		93	16.6	250	0.39	6.0	Before 6-70
2.96 x 0.735 x 0.092" SST-canned		93	13.5	240	0.30	4.7	Before 6-70
3.00 x 0.740 x 0.072" SST-bonded		124	11.8	250	0.37	5.7	Before 6-70

a. 152 wafers have central hole 0.070-in. diameter.
b. 68 wafers have central hole 0.070-in. diameter.
c. <450 Ci/g can be provided if required, by early removal from reactor.

SAVANNAH RIVER LABORATORY ^{60}Co PUBLICATIONS

QUARTERLY PROGRESS REPORTS

"Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report

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
DP-1196-I	January - March 1969, Part I - Cobalt
DP-1206-I	April - June 1969, Part I - Cobalt
DP-1216	July - September 1969

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).

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2. 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).

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4. C. L. Angerman and C. P. Ross. *Savannah River Laboratory Cobalt-60 Power and Heat Sources Quarterly Progress Report, July-September 1969.* USAEC Report DP-1216, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1969).

5. K. Z. Morgan, D. L. I. Evans, and M. R. Ford. "Maximum Permissible Concentration of Radioisotopes in Air and Water for Short Period Exposure." *Proc. U. N. Intern. Conf. Peaceful Uses of Atomic Energy, 1st Geneva* **13**, *139-159 (1955).*

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