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SAVANNAH RIVER LABORATORY COBALT-60 POWER AND HEAT SOURCES

QUARTERLY PROGRESS REPORT

JULY - SEPTEMBER 1970

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COBALT-60 POWER AND HEAT SOURCES

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December 1970

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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

Welding the centers of the caps in rhenium capsules melted small areas of adjacent cobalt oxide wafers and caused vapor transport of rhenium within the capsule. (p 1)

Satisfactory capsule performance was demonstrated through 20,000 hr at 1000°C with "Haynes" 25, "Hastelloy" C, and "Hastelloy" X capsules. (p 7)

The small diameter increases observed in "Haynes" 25 capsules after 10,000 hr at 1000°C can be accounted for semi-quantitatively by the precipitation of cobalt-tungsten intermetallic compounds within the "Haynes" alloy matrix. (p 10)

Nine of the ten superalloy capsules that contained irradiated cobalt and were heated for up to 10,000 hr at 1000°C were intact as measured by helium leak tests; the "Hastelloy" C capsule heated 10,000 hr had an indicated leak rate more than ten times that of the other capsules. (p 13)

Oxidation data obtained after 3000 hr at 1000 and 1125°C on capsule and core materials for the WANL 30 kw(t) demonstration unit indicated that radial gaps between fuel pin and Nickel 201 core should be greater than 0.050 in. to prevent interference during five years of operation at normal conditions. (p 16)

The oxide scale formed on "Haynes" 188 impeded reaction with Nickel 200 at 1125°C for only about 1000 hr. (p 20)

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 in normal and accident environments. Development of specific heat source concepts is not at present included in the scope of the SRL program, but assistance is provided when required for developing and testing demonstration units.

MATERIALS TECHNOLOGY AND DEVELOPMENT

CAPSULE FABRICATION AND TESTING

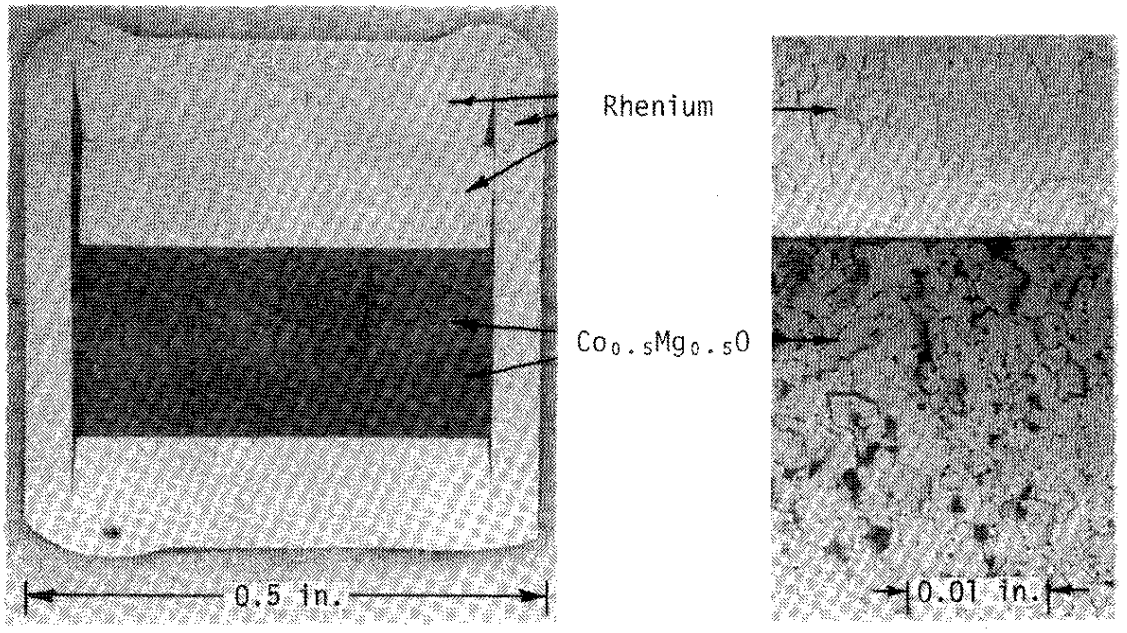
Welding of Refractory Metal Capsules

Techniques for welding test capsules of tungsten, rhenium, and tungsten-25 wt % rhenium alloy (W-25 wt % Re) are being developed. Rhenium and W-25 wt % Re capsules have been welded satisfactorily, but sealing pre-existing cracks in the caps of rhenium capsules by welding led to degradation of the uppermost cobalt oxide wafer and vapor transport of rhenium throughout the capsule. Cracking of welds in tungsten capsules has not been eliminated.

Rhenium

Several as-welded control capsules containing cobalt oxides have been examined for comparison with heat treated rhenium capsules to be examined later. Metallographic techniques have been developed to obtain polished interfaces with good contact and minimal relief between the metal and oxide. Best results were obtained by first grinding through 400 grit paper and then lapping with 500 grit silicon carbide slurry until interfaces are flat. Polishing is then accomplished using 14 micron diamond dust on a rotary wheel for one minute followed by two-weeks on a vibratory polisher using 1/2 micron diamond dust.

Capsules welded with one or two passes, but not heat treated, show no reaction at the oxide-rhenium interfaces, Figure 1. This capsule was welded with one pass at the bottom end and two passes at the top. Some porosity within the weld is visible, particularly on the bottom. One of the oxide wafers is cracked. These cracks



a) Capsule Cross Section

b) Oxide Wafer-Rhenium Spacer Interface

FIG. 1 AS-WELDED COMPATIBILITY CAPSULE

may have existed initially or may have been caused by thermal shock during welding or mechanical stresses during assembly of the capsule.

Welding the centers of the rhenium caps to close cracks in some capsules resulted in melting and severe cracking of the oxide wafers along with vapor transport of rhenium, Figure 2. These cracks were present in the bar stock from which the caps were machined. The high weld-heat input required to seal the cracks melted the oxide in the region adjacent to the center of the cap. In some cases rhenium metal was observed within the resolidified oxide. The extensive cracking of the oxide wafers visible in Figure 2a probably resulted from the severe thermal shock associated with welding.

A high mobility of rhenium was observed in the seal-welded capsules, Figure 3. Rhenium metal that was observed in the region between the spacer and cap of several capsules was apparently deposited from vapor formed during welding of the opposite end of the capsule. A layer of metal, again identified as rhenium, was also observed on the surface of the oxide in several capsules. This layer also may have been vapor deposited during welding.

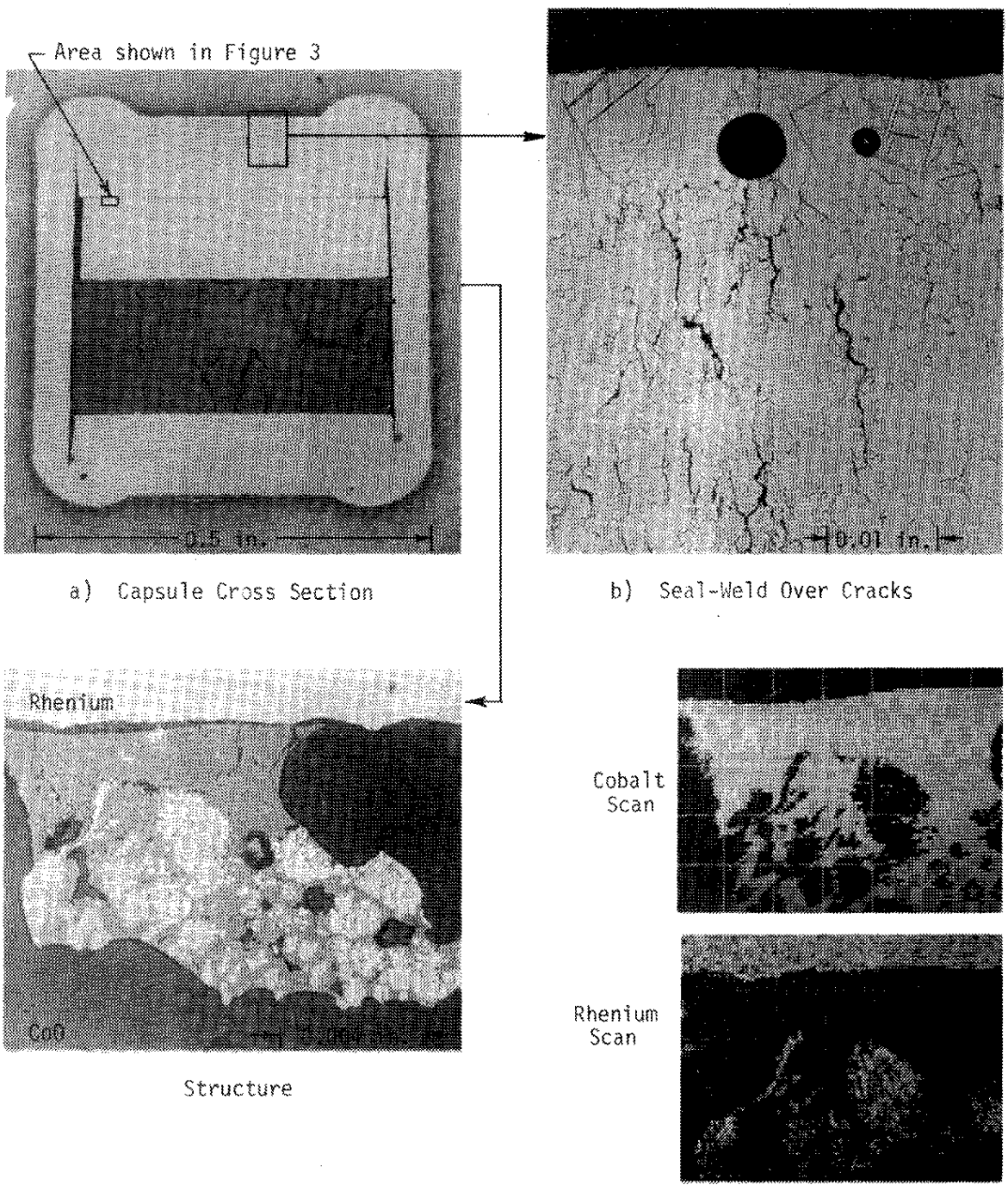
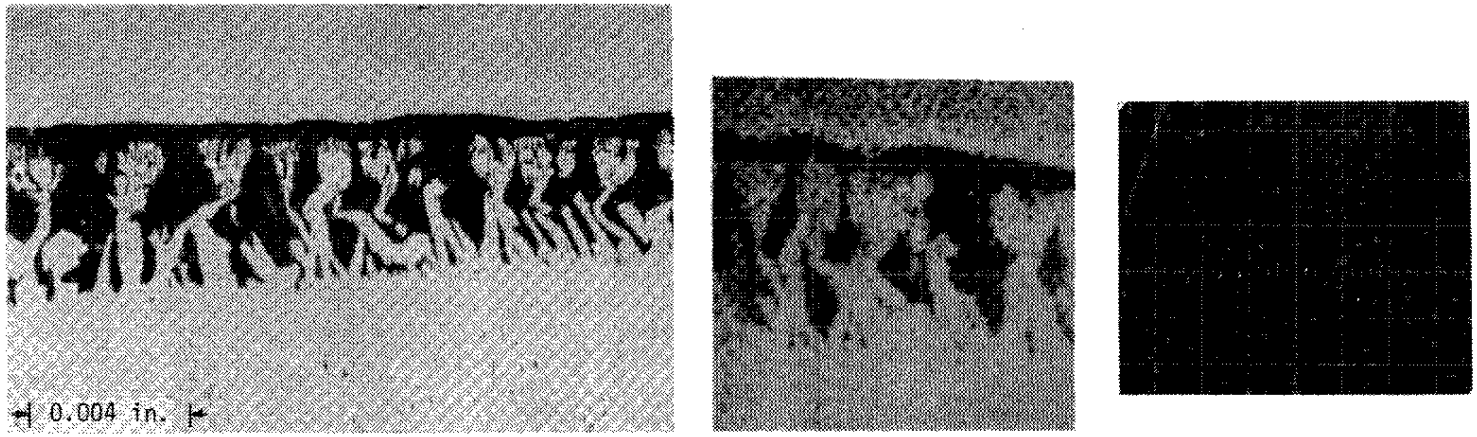
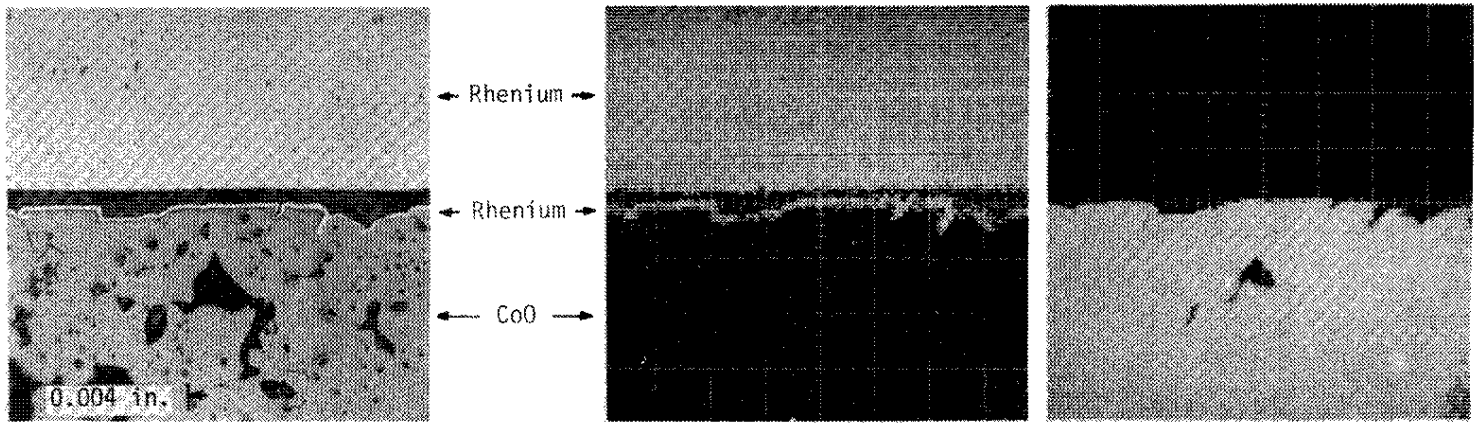


FIG. 2 SEAL-WELDED RHENIUM CAPSULE (Electron microprobe shows that molten rhenium was entrapped during resolidification of CoO wafer melted during seal-welding)



Micrograph | Rhenium Scan | Cobalt Scan

Vapor-deposited metal between rhenium spacer and cap. Accompanying electron microprobe scans show deposit to be rhenium with no cobalt present.



Micrograph | Rhenium Scan | Cobalt Scan

Rhenium, identified by electron microprobe analyses, was vapor deposited on surfaces of oxide wafers contained in seal-welded capsules.

FIG. 3 VAPOR TRANSPORT OF RHENIUM IN SEAL-WELDED CAPSULES

Heating Tests of Capsules with Unirradiated Cobalt Oxides

Superalloy Capsules

Tests completed this quarter show that the oxides of cobalt have no advantage over cobalt metal when encapsulated in "Inconel"* 600 for heat source applications at 1000°C. Three "Inconel" 600 capsules, each containing one of the oxides CoO, $CoAl_2O_4$ or $Co_{0.5}Mg_{0.5}O$, were exposed for 1000 hr at 1000°C. Each of the oxides was partially reduced by the "Inconel" 600, leaving free cobalt metal within the oxide wafers, Figure 4. Reduction of the oxides by other superalloys would be expected.

The internal surfaces of the "Inconel" 600 oxidized. Electron microprobe analysis will be used to determine which components of the "Inconel" 600 exist in the oxidized layer and therefore contributed to the reduction of cobalt oxides. The extent of diffusion between the "Inconel" 600 and the oxides will also be measured.

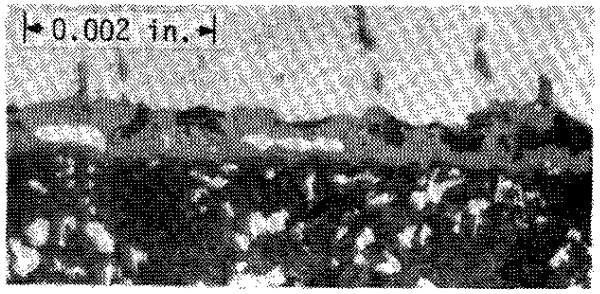


FIG. 4 "INCONEL" 600-$CoAl_2O_4$ INTERFACE AFTER 1000 HR AT 1000°C (Similar structures were seen in "Inconel" 600 capsules containing CoO and $Co_{0.5}Mg_{0.5}O$)

Refractory Metal Capsules

Thirteen rhenium capsules containing either CoO, $CoAl_2O_4$ or $Co_{0.5}Mg_{0.5}O$ are being exposed at 1500, 1700, and 1850°C for 1000 or 5000 hr, Table I. Three of the capsules contain foils of iridium and rhodium to determine compatibility of these metals with the oxides at 1500°C. The remaining ten capsules will be used to investigate the compatibility of rhenium with the oxides at the stated conditions. Six additional small rhenium capsules will be fabricated to complete the programs of 1000- and 5000-hr tests. Larger rhenium capsules to contain standard size (0.700-in.-dia x 0.150-in.-thick) oxide wafers are on order for 10,000- and 50,000-hr tests. Details of exposures of the oxides in iridium capsules, scheduled to begin in June, 1971, will depend on the iridium foil tests presently underway.

*Trademark of International Nickel Co.

TABLE I

^{59}Co HIGH TEMPERATURE FUELS COMPATIBILITY PROGRAM

Capsule Material and Temperature	Fuel Material	Heating Time, hr	Heating Startup	Heating Termination	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 detectable interaction
Rhenium (1525°C)	$Co_{0.5}Mg_{0.5}O$	200	7/69	7/69	No detectable interaction
Rhenium (1525°C)	$CoAl_2O_4$	200	7/69	7/69	No detectable 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	Reaction
TZM (1620°C)	$Co_{0.5}Mg_{0.5}O$	260	12/69	12/69	Reaction
TZM (1620°C)	$CoAl_2O_4$	260	12/69	12/69	Reaction
"Inconel" 600 (1000°C)	CoO	1,000	7/70	9/70	Oxide reduced
"Inconel" 600 (1000°C)	$Co_{0.5}Mg_{0.5}O$	1,000	7/70	9/70	Oxide reduced
"Inconel" 600 (1000°C)	$CoAl_2O_4$	1,000	7/70	9/70	Oxide reduced
		1500°C Tests			
Rhenium; foils of Ir, Rh	CoO	1,000	9/70	11/70	At temperature
Rhenium; foils of Ir, Rh	$Co_{0.5}Mg_{0.5}O$	1,000	9/70	11/70	At temperature
Rhenium; foils of Ir, Rh	$CoAl_2O_4$	1,000	9/70	11/70	At temperature
Rhenium	CoO	1,000	12/70	2/71	
Rhenium	$Co_{0.5}Mg_{0.5}O$	1,000	9/70	11/70	At temperature
Rhenium	$CoAl_2O_4$	1,000	9/70	11/70	At temperature
Rhenium	CoO	5,000	12/70	7/71	
Rhenium	$Co_{0.5}Mg_{0.5}O$	5,000	12/70	7/71	
Rhenium	$CoAl_2O_4$	5,000	9/70	4/71	At temperature
Rhenium	Best Oxide	10,000	2/71	4/72	Capsule ordered
Rhenium	Best Oxide	50,000	2/71	10/76	Capsule ordered
Iridium	CoO	1,000	6/71	8/71	
Iridium	$Co_{0.5}Mg_{0.5}O$	1,000	6/71	8/71	
Iridium	$CoAl_2O_4$	1,000	6/71	8/71	
		1700°C Tests			
Rhenium	CoO	1,000	12/70	2/71	
Rhenium	$Co_{0.5}Mg_{0.5}O$	1,000	9/70	11/70	At temperature
Rhenium	$CoAl_2O_4$	1,000	9/70	11/70	At temperature
Rhenium	CoO	5,000	12/70	7/71	
Rhenium	$Co_{0.5}Mg_{0.5}O$	5,000	9/70	4/71	At temperature
Rhenium	$CoAl_2O_4$	5,000	12/70	7/71	Capsule fabricated
Rhenium	Best Oxide	10,000	2/71	4/72	Capsule ordered
Rhenium	Best Oxide	50,000	2/71	10/76	Capsule ordered
Iridium	CoO	1,000	6/71	8/71	
Iridium	$Co_{0.5}Mg_{0.5}O$	1,000	6/71	8/71	
Iridium	$CoAl_2O_4$	1,000	6/71	8/71	
		1850°C Tests			
Rhenium	$Co_{0.5}Mg_{0.5}O$	1,000	9/70	11/70	At temperature
Rhenium	$CoAl_2O_4$	1,000	9/70	11/70	At temperature
Rhenium	$Co_{0.5}Mg_{0.5}O$	5,000	9/70	4/71	At temperature
Rhenium	$CoAl_2O_4$	5,000	9/70	4/71	At temperature
Rhenium	Best Oxide	10,000	2/71	4/72	Capsule ordered
Rhenium	Best Oxide	50,000	2/71	10/76	Capsule ordered
Iridium	$Co_{0.5}Mg_{0.5}O$	1,000	6/71	8/71	
Iridium	$CoAl_2O_4$	1,000	6/71	8/71	

Heating Tests of Capsules with Unirradiated Cobalt Metal

Superalloy Capsules

Long-Term Performance Tests

Two "Hastelloy"* X capsules and one capsule each of "Hastelloy" C and "Haynes"* 25 achieved their goal exposures of 20,000 hr at 1000°C and were removed from test. The capsules were intact as indicated by helium leak tests and bubble tests. The diameters of the "Hastelloy" capsules increased 0.002 in.; the diameter of the "Haynes" 25 capsule increased 0.006 in., as observed in previous tests for up to 10,000 hr. One of the "Hastelloy" X capsules was returned to test for an additional 10,000-hr exposure. Metallographic examination of the companion "Hastelloy" X capsule, as well as the "Hastelloy" C and "Haynes" 25 capsules, showed the width of the cobalt-capsule reaction zone and the depth into the exterior surface affected by oxidation to be in agreement with values predicted from earlier tests, Figure 5.

Heating tests with other capsules are continuing for up to 50,000 hr at 900 and 1000°C, Table II.

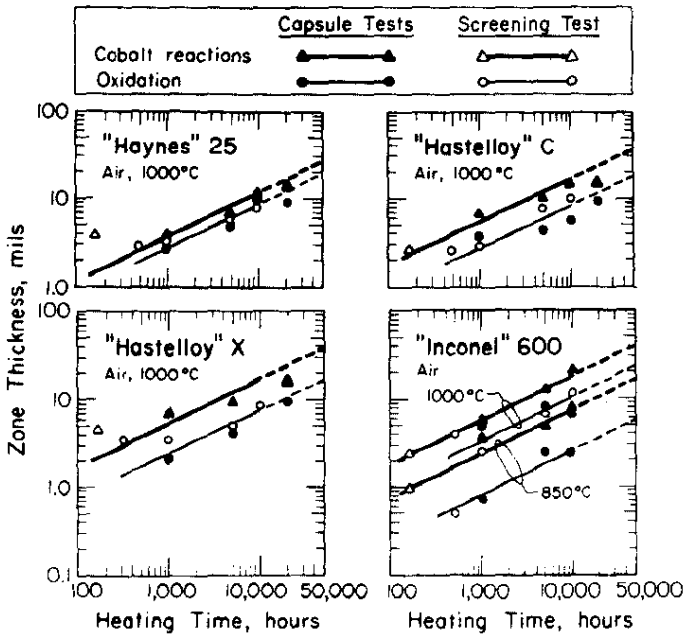


FIG. 5 GROWTH OF COBALT-CAPSULE REACTION AND CAPSULE OXIDATION ZONES (UNIRRADIATED COBALT)

*Trademark of Cabot Corp.

TABLE II

Summary of ^{59}Co Capsule Heating Tests

(All Cobalt Metal Wafers 0.745-in. dia)

Capsule Material	Heating Time, hr	Heating Temp, °C	Wall, mils	No. of Capsules	Approx. Starting Date	Approx. Completion Date	Remarks
"Inconel" 600	1,000	850	50	1	12-66	2-67	Capsule intact
(m.p. 1370°C)	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	Examination in progress
	20,000	900	95	1	11-68	3-71	
	→ 10,000[e]	900	95	1	3-69	5-70	Increased Co/capsule reaction
	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	Examination in progress
"Hastelloy" C	1,000	1,000	50	4	8-66	10-66	3 capsules intact; 1 capsule oxidized[b]
(m.p. 1270°C)	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	Capsule intact
TD Nickel	1,000	850[c]	95	1	10-67	12-67	Capsule intact
(m.p. 1450°C)	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	1,000[a]	1,000	95	2	10-67	12-67	Co near pinhole oxidized
(m.p. 1430°C)	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	Examination in progress
(m.p. 1330°C)	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	Capsule intact
"Hastelloy" X	1,000	1,000	50	1	4-67	6-67	Capsule intact
(m.p. 1260°C)	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	1	5-68	9-70	Capsule intact
	→ 30,000	1,000	95	1	5-68	11-71	

[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.

Effect of Caustic Residues

Metallographic examination was completed on an "Inconel" 600 capsule removed from test last quarter after 10,000 hr at 900°C. This capsule contained wafers intentionally contaminated with a caustic residue and was similar to one heated for 5000 hr (see DP-1226). The cobalt-capsule reaction zones were wider in the capsules containing contaminated wafers than in those containing clean wafers. Microprobe analyses showed that aluminum was present in the grain boundaries of the "Inconel"-contaminated wafer reaction zone. The widths and general appearance of the reaction zones in capsules containing contaminated wafers were similar to those in capsules containing ^{60}Co capsules heated under the same conditions, Figure 6. These results confirm the hypothesis that the increased reaction in the active capsules was caused by insufficient cleaning after dissolution of the aluminum target slug in NaOH rather than an effect of the radiation field on compatibility.

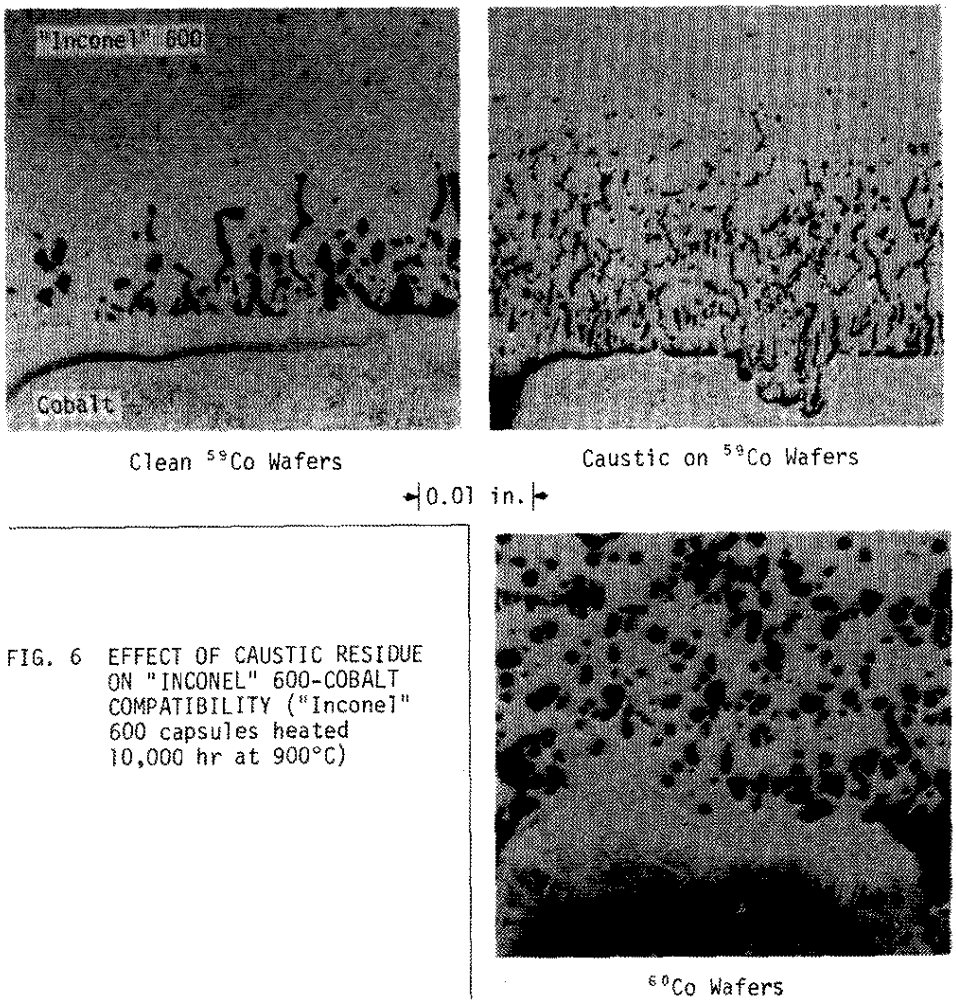


FIG. 6 EFFECT OF CAUSTIC RESIDUE ON "INCONEL" 600-COBALT COMPATIBILITY ("Inconel" 600 capsules heated 10,000 hr at 900°C)

Diameter Increases in "Haynes" 25 Capsules

The diameters of all unirradiated "Haynes" 25 capsules tested to date have increased more than those of capsules fabricated from other materials. Moreover these increases were the same at the ends of the capsules as at the centers. In contrast, the diameters of capsules of other materials increased at the center, as would be expected if internal pressure had caused creep of the capsule wall. These observations indicated that the "Haynes" 25 itself was becoming less dense, probably because of some change in the phases present in the alloy.

The diameter increases can be accounted for semi-quantitatively by the precipitation of an A_2B-type phase where A is cobalt, nickel, or chromium and B is tungsten or molybdenum. The densities of samples cut from the caps of heated "Haynes" 25 capsules were measured and their structures were characterized by X-ray diffraction and both optical and electron metallographic techniques. Table III shows the general agreement between the observed density decreases and those calculated from changes in the lattice parameters of the phases present. A change in the structure of cobalt matrix from the fcc to the hcp form could also contribute to the density decrease, but the hcp form was not detected by X-ray diffraction. Similar changes may occur in "Haynes" 188 and other cobalt-based superalloys containing tungsten.

TABLE III

Analysis of Density Changes in "Haynes" 25

	As-Received	5000 hr at 1000°C	10,000 hr at 1000°C
Maximum increase in capsule diameter, mils (original diameter 0.94 in.)	-	2.0	8.3
Calc ΔD^2, %[a]	-	+0.43	+1.8
Measured $\Delta_\rho/_\rho$, %	-	-0.23	-0.15
Phases present - lattice parameter, Å	Matrix-3.5858 Co_3W_3C-11.030	Matrix-3.5808 Co_3W_3C-11.048 Co_2W-4.74[b] -7.68	Matrix-3.5812 Co_3W_3C-11.050 Co_2W-4.65 -7.55
Volume fraction of phase present[c]	Matrix-0.929 Co_3W_3C-0.071	Matrix-0.882 Co_3W_3C-0.099 Co_2W-0.019	Matrix-0.941 Co_3W_3C-0.049 Co_2W-0.010
Calc $\Delta_{\bar{\rho}}/_{\bar{\rho}}$, %[d]	-	-0.27	-0.52

[a] Change in (diameter)^2 is inversely proportional to change in density for solid cylinder when length and weight of capsule are assumed to be constant.

[b] Two values shown are for a_o and c_o respectively for hcp Co_2W; only a_o values are given for fcc matrix and Co_3W_3C.

[c] Volume fraction was based on relative intensities of the strongest diffraction peaks.

[d] Density of each phase was calculated from lattice parameters, and a weighted average density of each sample was calculated from these values and the volume fraction.

Refractory Metal Capsules

Heating tests for 1000 hr were completed on two nonradioactive W-25 wt % Re alloy capsules, one at 1200°C and one at 1400°C. The capsules were intact and their dimensions did not change. The cobalt-capsule reaction zones, Figure 7, were wider than predicted from earlier 168-hr screening tests because of more reaction and formation of voids within the cobalt. Heating is continuing on seven other capsules, Table IV.

Five nonradioactive tungsten capsules were heated 4 hr at 1200°C prior to the start of long-term heating tests. Three of the capsules developed leaks in their welds, as detected by bubble tests, and will be rewelded. Tests for 1000 and 5000 hr were started on the two intact capsules, Table IV.

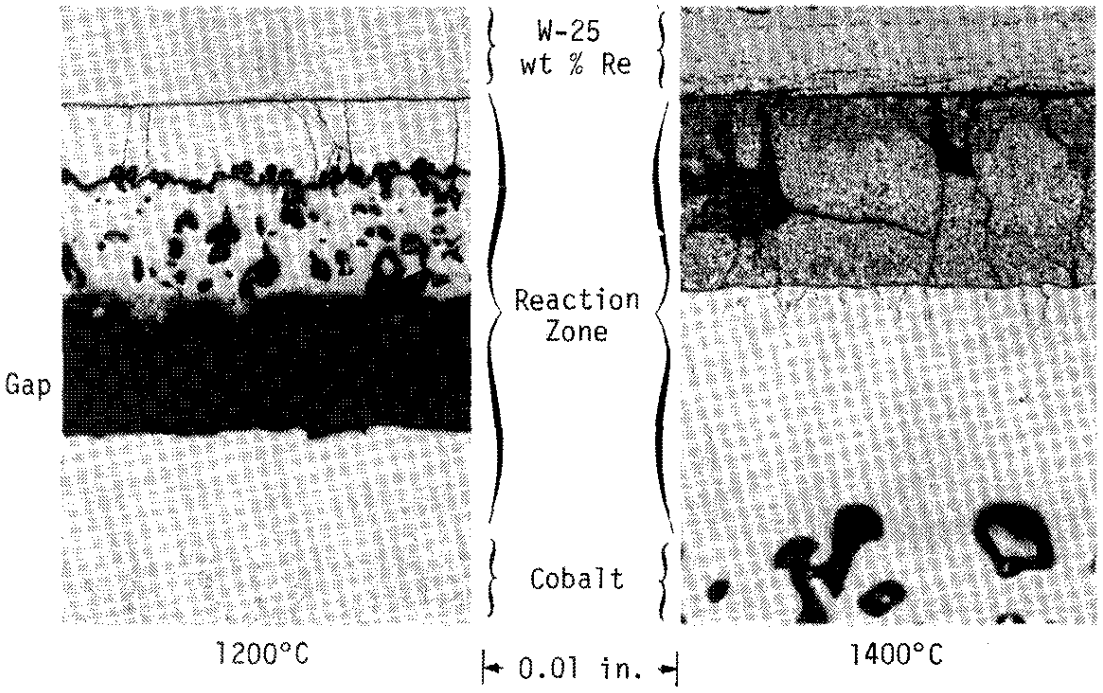


FIG. 7 COMPATIBILITY REACTIONS IN W-25 WT % Re CAPSULES AFTER 1000 HR AT 1200 and 1400°C

TABLE IV

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	Remarks
Tungsten	→ 1,000	1200	9-70	11-70	0.745	TIG	
	→ 1,000	1200	10-70	11-70	0.745	TIG	
	→ 5,000	1200	9-70	4-71	0.745	TIG	
	→ 5,000	1200	10-70	4-71	0.745	TIG	
	5,000	1200	1-71	8-71	1.490	TIG	
	5,000	1200	10-70	5-71	0.745	EB	
	→10,000	1200	10-70	11-71	0.745	TIG	
	10,000[b]	1200	10-70	12-71[b]	0.745	TIG	
	10,000	1200	1-71	3-72	1.490	TIG	
	10,000[b]	1200	1-71	3-72[b]	1.490	TIG	
	10,000	1200	10-70	12-71	0.745	EB	
	50,000	1200	10-70	7-76	0.745	TIG	
Rhenium	1,000	1200	11-70	1-71	0.745	TIG	
	5,000	1200	11-70	6-71	0.745	TIG	
	5,000	1200	11-70	6-71	0.745	EB	
	10,000	1200	11-70	1-72	0.745	TIG	
	10,000[b]	1200	11-70	1-72[b]	0.745	TIG	
	10,000	1200	11-70	1-72	0.745	EB	
	50,000	1200	11-70	8-76	0.745	TIG	
	1,000	1400	11-70	1-71	0.745	TIG	
	5,000	1400	11-70	6-71	0.745	TIG	
	10,000	1400	11-70	1-72	0.745	TIG	
W-25 wt % Re	→ 1,000	1200	8-70	10-70	0.745	TIG	Capsule intact
	5,000	1200	8-70	3-71	0.745	TIG	
	5,000	1200	8-70	3-71	0.745	EB	
	10,000	1200	8-70	10-71	0.745	TIG	
	10,000[b]	1200	8-70	10-71[b]	0.745	TIG	
	10,000	1200	10-70	12-71	0.745	TIG	
	50,000	1200	8-70	5-76	0.745	TIG	
	→ 1,000	1400	8-70	10-70	0.745	TIG	Capsule intact
	5,000	1400	8-70	3-71	0.745	TIG	
	10,000	1400	8-70	10-71	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 with Irradiated Cobalt Metal

Superalloy Capsules

Helium leak tests were completed on ten capsules of the four candidate superalloys that were heated for 5000 or 10,000 hr at 900 or 1000°C, Table V. One "Inconel" 600 capsule and one "Haynes" 25 capsule contained unirradiated cobalt, but were heated along with the radioactive capsules to assess the effects of the radiation field on capsule performance. These capsules have been stored at ∿500°C since the completion of the heating tests last year. The additional cobalt-capsule reactions and oxidation of the capsule surfaces during storage is expected to be negligible (<0.001 in.) in comparison to the reactions that occurred during the heating tests.

All the capsules, except the one "Hastelloy" C capsule heated 10,000 hr at 1000°C, were intact as measured by helium leak tests. The "Hastelloy" C capsule had an indicated leak rate more than 10 times that of the other capsules and maintained this rate for the duration of the test. The smaller leak rates of the other capsules decreased with test time indicating that helium adsorbed on the rough oxidized surface was being driven off. Measurement of dimensions and metallographic examination of all capsules is in progress.

Heating tests are continuing on five other capsules for 20,000 hr at either 900 or 1000°C, Table V. The large furnace that contained all capsules being heated at 1000°C failed in July. The three capsules with goal exposures of 20,000 hr at 1000°C were transferred to another furnace and the heating resumed. Further heating of the four capsules with goal exposures of 50,000 hr at 1000°C will be postponed until the completion of the 20,000-hr tests due to lack of additional furnace space. The approximate completion dates in Table V for all tests have been revised to reflect these and other delays in the test schedule.

TABLE V

Summary of ^{60}Co Capsule Heating Tests

(All Cobalt Metal Wafers 0.745-in. dia)

Capsule Material	Heating		Wall, mils	No. of Capsules	Activity		Approx. Starting Date	Approx. Completion Date	Remarks
	Time, hr	Temp, °C			Spec, Ci/g	Total, Ci			
"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	Examination in progress
	→20,000	900	95	1	288[c]	13,700	7-68	11-70	
	→50,000	900	95	1	282[c]	13,400	7-68	4-74	
	→ 5,000	1,000	95	1	295[c]	14,000	9-68	4-69	Examination in progress
	→10,000	1,000	95	1	288[c]	13,700	9-68	11-69	Examination in progress
	→20,000	1,000	95	1	263[c]	12,500	9-68	3-71	
	→50,000	1,000	95	1	255[c]	12,100	9-68	1-75	
	→10,000	850	95	1	(d)	-	9-68	11-69	Examination in progress
"Hastelloy" C	100	850	50	1	120	9,000	1-67	1-67	Examination in progress
(m.p. 1270°C)	→10,000	900	95	1	276[c]	13,100	7-68	8-69	Examination in progress
	→10,000	1,000	95	1	282[c]	13,400	9-68	11-69	Examination in progress
	→50,000	1,000	95	1	270[c]	12,800	9-68	1-75	
"Haynes" 25	→ 5,000	1,000	95	1	263[c]	12,500	9-68	4-69	Examination in progress
(m.p. 1330°C)	→10,000	1,000	95	1	288[c]	13,700	9-68	11-69	Examination in progress
	→20,000	1,000	95	1	282[c]	13,400	9-68	3-71	
	→50,000	1,000	95	1	295[c]	14,000	9-68	1-75	
	→10,000	850	95	1	(d)	-	9-68	11-69	Examination in progress
"Hastelloy" X	→ 5,000	1,000	95	1	250[c]	11,900	9-68	4-69	Examination in progress
(m.p. 1260°C)	→10,000	1,000	95	1	263[c]	12,500	9-68	11-69	Examination in progress
	→20,000	1,000	95	1	263[c]	12,500	9-68	3-71	
	→50,000	1,000	95	1	301[c]	14,300	9-68	1-75	

[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.

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 (WANL) is the contractor for this project.

Heat Source Design

WANL has completed Phase II, Detailed Design, of the heat source, and are proceeding with plans for Phase III, Fabrication. The designs for heat source core assembly, fuel pin, and capsule are reproduced in Figures 8 through 10. The fuel pins and capsules are based partly on technology previously developed at SRL and on the tests described below.

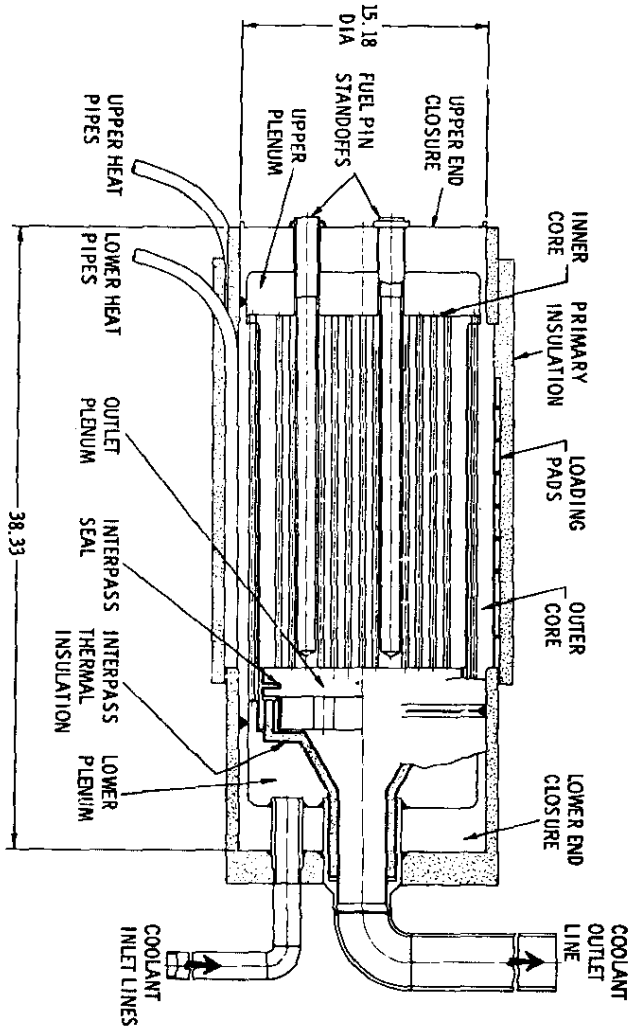


FIG. 8 ^{60}Co CORE ASSEMBLY FOR WANL UNIT

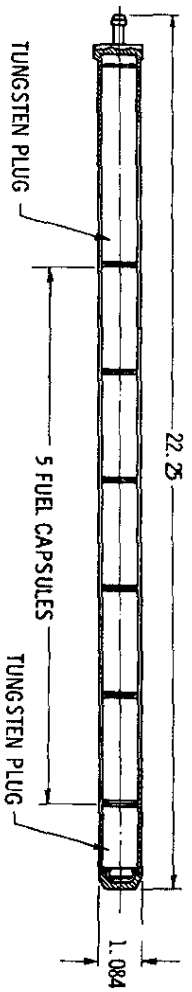


FIG. 9 ^{60}Co FUEL PIN ASSEMBLY FOR WANL UNIT

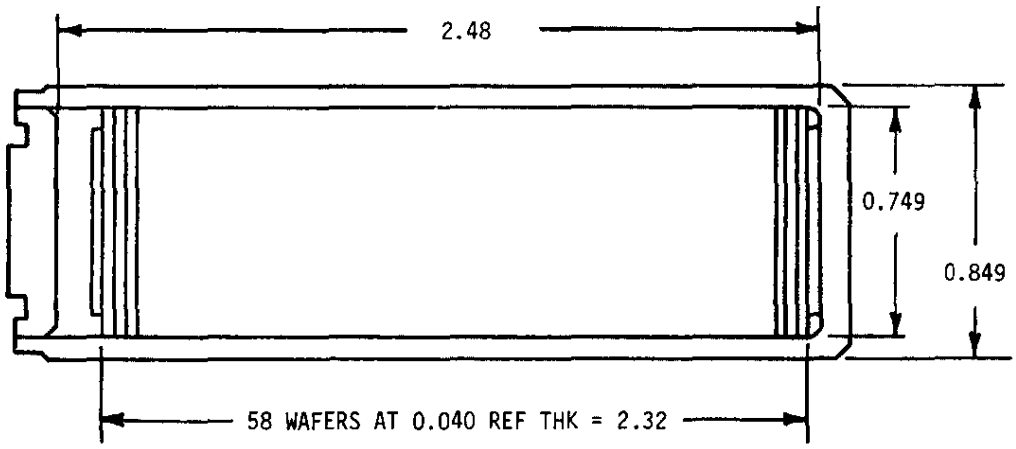


FIG. 10 FUEL CAPSULE FOR WANL UNIT

Capsule Material Tests

"Haynes" 188 is the proposed material for the outer fuel capsule because of (1) its resistance to oxidation, (2) its high strength, and (3) its compatiblity with both "Haynes" 25, the preferred inner capsule material, and Nickel 201, the inner core material. The oxidation and compatibility of these materials are being characterized to ensure that neither reaction would interfere with discharging fuel capsules or compromise capsule integrity.

Oxidation in Still Air

Samples of candidate core and capsule materials are being heated in box furnaces at 1000°C (normal heat source operating temperature) for up to 10,000 hr and at 1125°C (emergency cooling operating temperature) for up to 3000 hr. Oxidation rates determined in these tests are being interpreted as maximum values; actual rates in the heat source should be smaller because of the limited access of air through seams in the shielding.

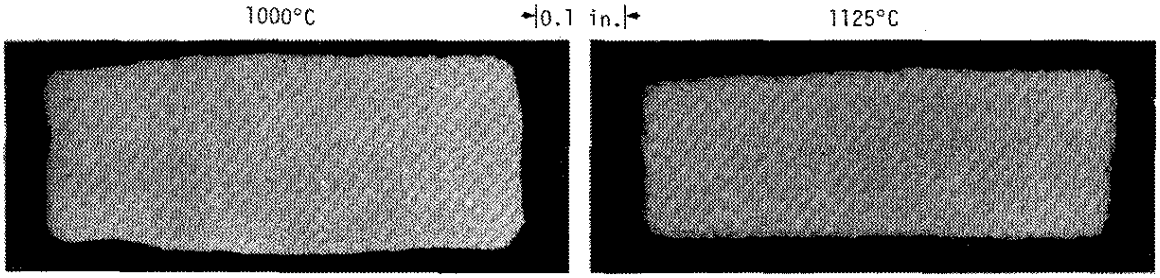
Tests through 3000 hr at 1000 and 1125°C have been completed. Pertinent dimensional data on the oxide scale are summarized in Table VI; sections of the samples after 3000 hr are shown in Figure 11. Essentially all the oxide spalled off the "Haynes" 188, while no spalling occurred on the Nickel 200. The very rapid oxidation of "Haynes" 25 during 3000 hr at 1000°C and 500 hr at 1125°C, was not expected; severe oxidation was observed in previous tests (DP-1177-I) only after 3000 hr at 1150°C. "Hastelloy" X, an alternative outer capsule material, was more resistant to oxidation than any of the other materials.

TABLE VI

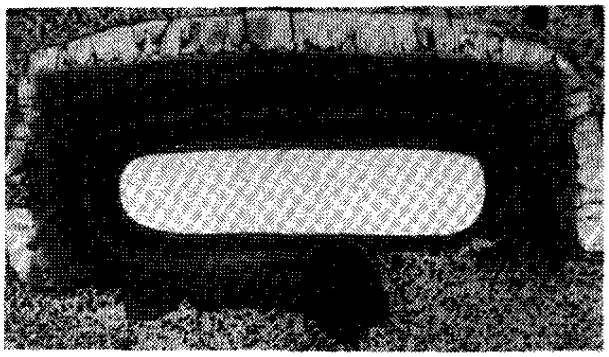
Oxidation Data for Candidate Heat Source Materials

	1000°C				1125°C			
	Hours				Hours			
	500	1000	2000	3000	500	1000	2000	3000
"Haynes" 188								
Specimen thickness[a]	0.250	0.252	0.255	0.230	0.254	0.250	0.225	0.200
Adherent Oxide thickness[b]	<0.002	<0.002	<0.002	0.005	<0.002	<0.002	<0.002	<0.002
Growth[c]	-	-	-	0.006	-	-	0.006	0.011
"Haynes" 25								
Specimen thickness	0.250	0.250	0.220	0.430	0.500			
Adherent Oxide thickness	<0.002	<0.002	<0.002	0.180	0.160	—— Discontinued ——		
Growth	-	-	0.007	0.072	0.064			
"Hastelloy" X								
Specimen thickness	0.253	0.250	0.255	0.245	0.250	0.250	0.248	0.245
Adherent Oxide thickness	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002
Growth	-	-	-	-	-	-	-	-
Nickel 200								
Specimen thickness	0.270	0.270	0.275	0.275	0.270	0.282	0.290	0.290
Adherent Oxide thickness	0.015	0.020	0.032	0.035	0.020	0.030	0.050	0.055
Growth	+0.006	+0.008	+0.012	+0.014	+0.008	+0.014	+0.020	+0.022

[a] All specimens were 0.250- to 0.255-in.-thick originally.
[b] Thickness of unspalled oxide on one side of sample.
[c] "Growth" is 40% of the oxide thickness assuming no spalling had occurred.

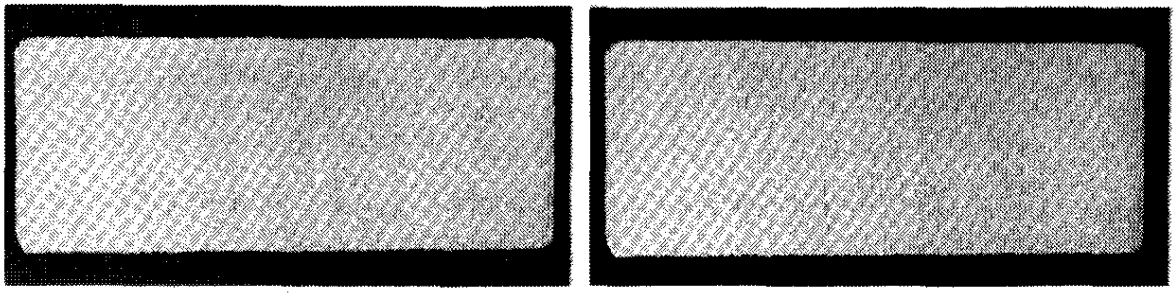


"Haynes" 188

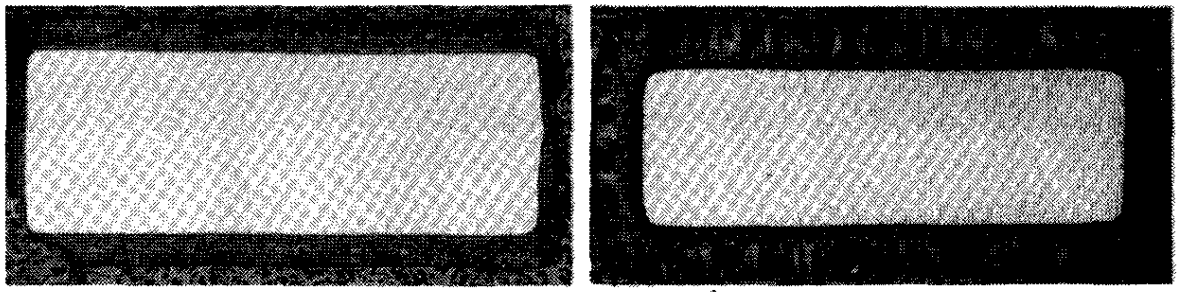


Tests of "Haynes" 25 at 1125°C discontinued after 500 hr.

"Haynes" 25



"Hastelloy" X



Nickel 200

FIG. 11 OXIDATION SAMPLES AFTER 3000 HR AT 1000 AND 1125°C

Although the preferred core material has been changed from Nickel 200 to Nickel 201, Nickel 200 is being used in these tests to provide conservative results. Previous tests at 1000°C for up to 10,000 hr showed that Nickel 200 was less resistant to oxidation than Nickel 201, Figure 12. Data from the current tests are in good agreement with those from previous tests, except for one "Haynes" 188 sample (3000 hr at 1000°C).

The values listed in Table VI for growth represent the advance of a plane surface as it is oxidized, and is equivalent to about 40% of the total thickness of the oxide assuming that no spalling of the oxide had occurred. These values can be used to assess potential interferences between components of the heat source. For example, a radial gap of 0.020 in. between a "Haynes" 188 clad fuel pin and a Nickel 200 core could be completely filled with oxide after 3000 hr at 1000°C. Similarly, previous data in Figure 12 for "Haynes" 188 and Nickel 201 indicate that the fuel pin-core gap must be greater than 0.050 in. to prevent an interferrence after 5 yr at 1000°C, the goal conditions for normal operation of the heat source.

In addition to the oxide scale, microstructural changes occurred to varying depths in the residual metal, depending on the heating conditions. The significance of these changes is being evaluated by hardness measurements and additional metallographic examinations.

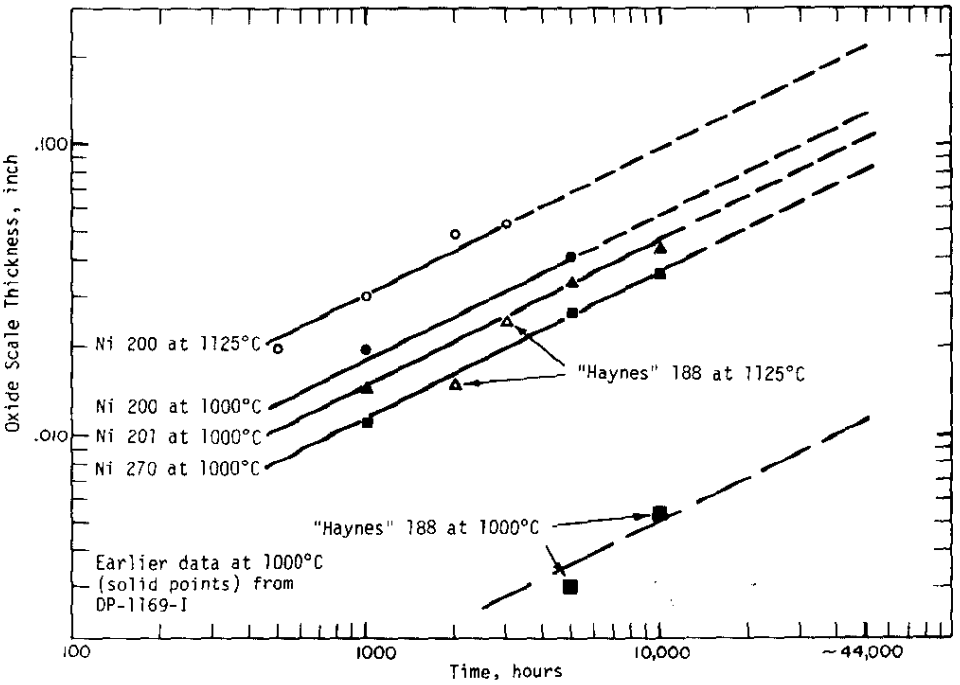


FIG. 12 OXIDATION OF NICKEL ALLOYS AND "HAYNES" 188 AT 1000 AND 1125°C

Oxidation Tests in Limited Air

The oxidation rates of Nickel 200 and "Haynes" 188 are being measured in a limited supply of air such as expected in the actual heat source. In initial tests, samples that were heated 500 hr at 1125°C reacted with the sealed ceramic chamber into which air was admitted through a hole 1/8-in. dia x 8 in. long. Further testing with this equipment was discontinued.

New tests will be started with a "Haynes" 188 cylinder inserted in a piece of Nickel 200 and sealed in a "Hastelloy" C capsule that has a pinhole in one end to admit a limited amount of air. The amount of oxidation and the force necessary to push the "Haynes" 188 cylinder out of the hole in the nickel will be measured after suitable exposures.

Compatibility Tests

Tests are in progress that will measure the reaction between nickel and both clean and pre-oxidized (1000°C for one week in air) "Hastelloy" X, "Haynes" 25, and "Haynes" 188, and between "Haynes" 25 (primary cladding) and "Haynes" 188 or "Hastelloy" X (secondary cladding) at 1000°C for 1000, 5000 and 10,000 hr and, at 1125°C for 500, 1000, and 3000 hr. Pre-oxidation simulates the expected surface conditions of as-fabricated capsules.

Couples examined after 3600 hr at 1000°C showed essentially no reaction between pre-oxidized "Haynes" 188 or "Haynes" 25 and the Nickel 200. The specimens separated easily.

Couples exposed up to 3000 hr at 1125°C have also been examined; the results are summarized in Table VII. At this temperature the oxide layer formed by pre-oxidizing "Haynes" 188 and "Haynes" 25 provided only a temporary barrier to reaction with Nickel 200. After 3000 hr the reaction zone widths were the same for both pre-oxidized and clean samples, Figures 13 and 14. In contrast the oxide layer on "Hastelloy" X did not inhibit the reaction at all, Figure 15.

Voids were formed on the superalloy side of each of the couples. The majority of these voids are probably the result of the Kirkendall effect (unequal diffusion rates across the interface). However, a high concentration of voids occurred on the grain boundaries intersecting the surfaces of the pre-oxidized "Haynes" alloys but not on the boundaries of the clean "Haynes" alloys. These grain boundary voids may have been formed by volitalization of the tungsten oxide constituents in the pre-existing oxide film.

There was little reaction apparent (<0.002 in.) between "Haynes" 25 and "Haynes" 188 after 3000 hr at 1125°C, Figure 16.

TABLE VII

Width of Affected Zone After Exposure at 1125°C

	Material Tested	500 Hours	1000 Hours	3000 Hours	Remarks
Nickel 200	"Haynes" 188	0.008 in. in Nickel 200 0.004 in. in "Haynes" 188	Inadequate contact between wafers	0.010 in. in Nickel 200 0.030 in. in "Haynes" 188	Voids form on "Haynes" 188 side
	Pre-oxidized "Haynes" 188	No Reaction	No Reaction	0.010 in. in Nickel 200 0.030 in. in "Haynes" 188	
	"Haynes" 25	0.008 in. in Nickel 200	Inadequate contact between wafers	0.010 in. in Nickel 200 0.035 in. in "Haynes" 25	Voids form on "Haynes" 25 side
	Pre-oxidized "Haynes" 25	No Reaction	No Reaction	0.010 in. in Nickel 200 0.035 in. in "Haynes" 25	
	"Hastelloy" X	0.012 in. in Nickel 200 0.020 in. in "Hastelloy" X	0.020 in. in Nickel 200 0.030 in. in "Hastelloy" X	0.020 in Nickel 200 0.040 in. in "Hastelley" X	Voids form on "Hastelloy" X side
	Pre-oxidized "Hastelloy" X	0.012 in. in Nickel 200 0.020 in. in "Hastelloy" X	0.020 in. in Nickel 200 0.030 in. in "Hastelloy" X	Not tested	
"Haynes" 25	"Haynes" 188	Spotty bonding but no zone detectible		Small amount of reaction product <0.010 in. thick formed in some places	
	"Hastelloy" X	0.015 in. Total	0.025 in. Total	Not tested	

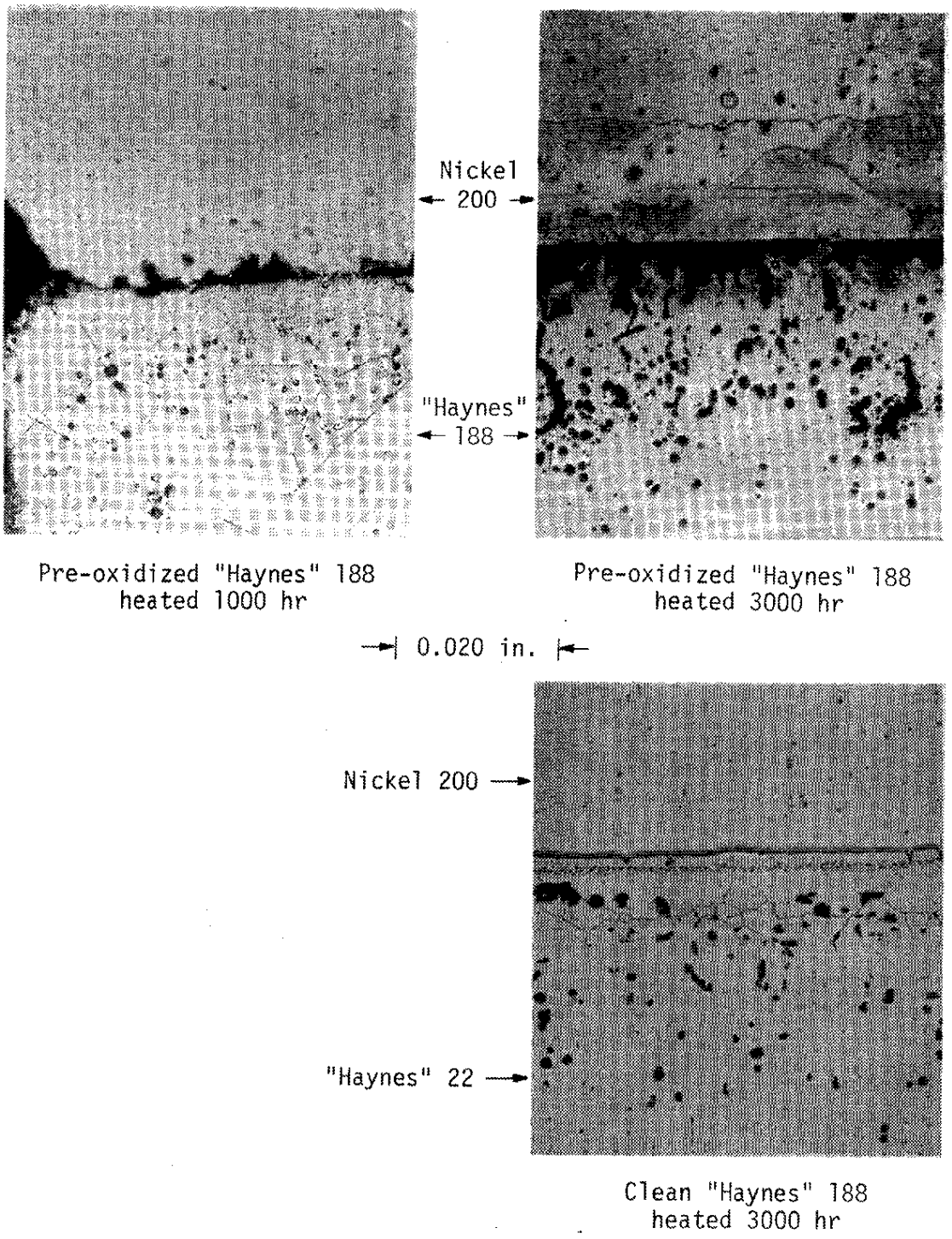
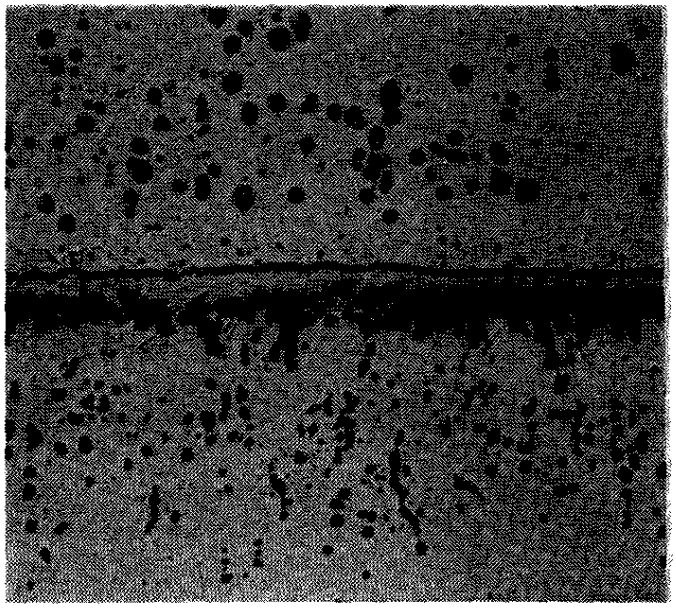
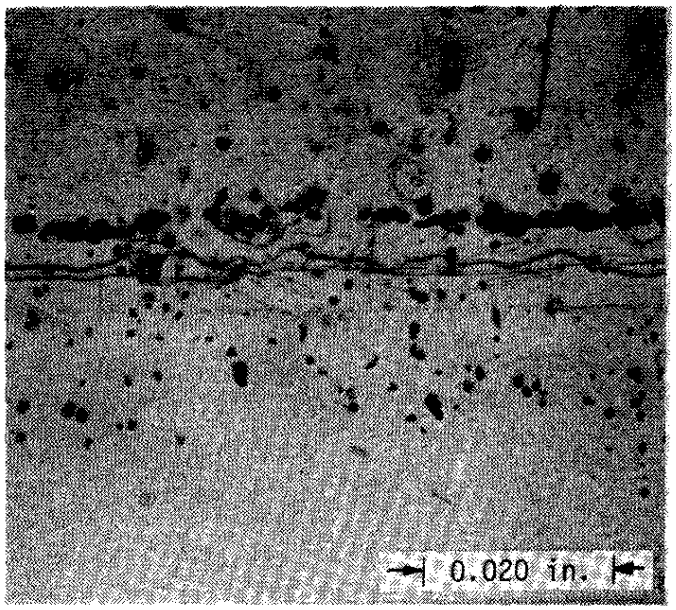


FIG. 13 REACTION BETWEEN NICKEL 200 AND "HAYNES" 188 AT 1125°C



Pre-oxidized "Haynes" 25



Clean "Haynes" 25

FIG. 14 REACTION BETWEEN NICKEL 200 AND "HAYNES" 25 AFTER 3000 HR AT 1125°C

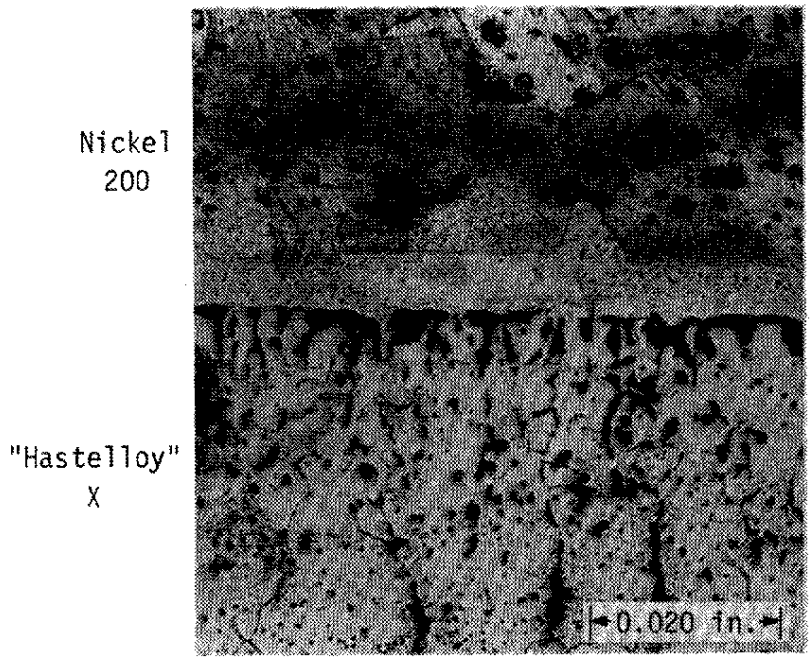


FIG. 15 REACTION BETWEEN NICKEL 200 AND PRE-OXIDIZED "HASTELLOY" X AFTER 3000 HR AT 1125°C

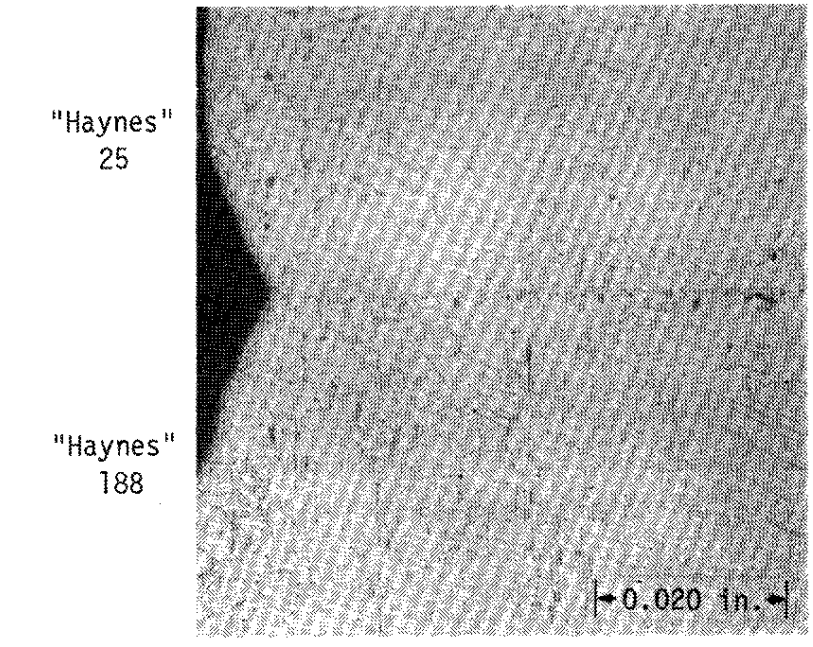


FIG. 16 REACTION BETWEEN "HAYNES" 25 AND "HAYNES" 188 AFTER 3000 HR AT 1125°C

^{60}Co LOAN PROGRAM

Because of the potential application of high activity cobalt metal in heat sources, the AEC has established a loan program for this material. About 10 MCi (150 kw) of ^{60}Co at over 350 Ci per gram of cobalt will be available for this program. Most of this material is in the form of 0.745-in.-dia wafers, plated with nickel. Individual companies or groups of companies are invited to participate by contacting the Savannah River Operations Office of the AEC. Moderate activities and amounts of ^{60}Co can be obtained commercially.

Radioactive cobalt metal shapes available under this loan program are listed in Table VIII.

TABLE VIII

^{60}Co Metal for Heat Source Development

(Activity as of 12/31/70)

	No. of Pieces	Wt of Co, g/piece	Avg Sp Activity, Ci/g Co	Total Activity, MCi	Total Power, kw(t)
Wafers, 0.040-in. thick					
0.745-in. dia	2090	2.5	250	1.31	20.4
	3800	2.5	200	1.90	29.6
0.800-in. dia	391	2.8	230	0.25	3.9
Wafers, 0.073-in. thick					
0.745-in. dia	3080	4.5	470	6.51	101.5
	2660	4.5	350	4.18	65.1
	1860	4.5	230	1.92	30.0
	4560	4.5	200	4.10	63.8
Half-wafers, 0.073-in. thick					
1.00-in. dia	682	4.1	300	0.84	13.1
1.25-in. dia	434	6.4	300	0.83	12.9
1.49-in. dia	620	9.1	300	1.69	26.3
Slabs,					
3.00 x 0.64 x 0.060-in. Ni-plated	93	16.6	230	0.35	5.5
2.96 x 0.735 x 0.092-in. SST-canned	93	13.5	230	0.29	4.5
3.00 x 0.740 x 0.072-in. SST-bonded	124	11.8	230	0.34	5.3
Total				24.51	381.9

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
DP-1226	October - December 1969
DP-1237	January - March 1970
DP-1247	April - June 1970

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).

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