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AEC RESEARCH AND DEVELOPMENT REPORT

SAVANNAH RIVER LABORATORY
COBALT-60 POWER AND HEAT SOURCES

QUARTERLY PROGRESS REPORT

OCTOBER - DECEMBER 1970

DU PONT
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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.

HIGHLIGHTS

Wafers of CoO, $Co_{0.5}Mg_{0.5}O$, and $CoAl_2O_4$ were irradiated successfully to specific activities of up to 700 Ci/g. (p 1)

The strength of 90% dense $CoAl_2O_4$ is 12,000 psi, or twice the strength of the other two oxides at the same density. Fracture of $Co_{0.5}Mg_{0.5}O$ wafers by a diametrical compression test yielded the least fines; fracture of CoO wafers resulted in the most fines. (p 2)

CoO was more resistant to corrosion than the other two compounds during 9-week tests in fresh water at 125°C flowing at 10 ft/sec. (p 3)

The thermal conductivities of the three oxide compounds, which were calculated from measured thermal diffusivities, are relatively low, typical of most ceramic materials. (p 7)

The walls of rhenium capsules containing unirradiated oxides were blistered and thinned after up to 1000 hr at 1500 to 1850°C due to vapor transport of rhenium within the capsules. (p 10)

"Inconel" 600, "Hastelloy" C, and "Hastelloy" X capsules containing unirradiated cobalt metal were intact after heating for up to 30,000 hr at 1000°C; a similar "Haynes" 25 capsule failed after 30,000 hr at 1000°C due to oxidation. (p 14)

"Hastelloy" X has been selected as the capsule and pin material for the WANL 30 kw(t) Demonstration Unit. (p 21)

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PROGRAM

The purpose of the Savannah River Laboratory (SRL) program on ^{60}Co is to provide data that will be required for designing, fabricating, and operating ^{60}Co heat sources. Primary emphasis is on selecting materials for encapsulating cobalt fuel forms and establishing temperature limits for 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 to other contractors when required for the development and testing of demonstration units.

MATERIALS TECHNOLOGY AND DEVELOPMENT

CERAMIC COBALT COMPOUNDS

Ceramic compounds may provide higher temperature capability and a larger margin of safety than ^{60}Co metal in some isotopic heat sources. Fabrication techniques have been developed for CoO, $Co_{0.5}Mg_{0.5}O$, and $CoAl_2O_4$.[1] Properties of irradiated and unirradiated oxides that are of interest to heat source applications are being measured in a continuing program sponsored by the Division of Production.

Irradiation

Successful production of ^{60}Co as wafers of the three ceramic compounds has been demonstrated by irradiation of 360 wafers in high-flux reactor charges to specific activities of up to 700 Ci/g or a total of 7.5 kw. Disassembly of two target slugs containing 17 kCi as 12 wafers showed that the wafers were intact and no volatile or particulate cobalt was released.

Properties of Unirradiated Oxides

Tensile Strength

The tensile strength of the ceramic materials was measured by a diametrical compression technique,[2] on wafers 0.700-in. dia x 0.150-in. thick. The strength of 90% dense $CoAl_2O_4$ was approximately 12,000 psi, or twice the strength of the other two oxides at the same density. The tensile strengths of the cobalt oxides are lower than analogous ceramic materials; for example, the strengths of Al_2O_3, $MgAl_2O_4$, and MgO at approximately 90% of theoretical density are 20,000 psi, 18,000 psi, and 14,000 psi,[3] respectively.

The relatively low tensile strengths of most ceramics, including the cobalt oxides, result in poor resistance to thermal shock. A tensile strength in excess of 100,000 psi or a very low thermal expansion is required to withstand thermal stresses arising from rapid temperature cycles greater than about 200°C.[4] Since the thermal expansions of the cobalt oxides are high, their resistance to thermal shock is relatively poor. However, all three cobalt oxides did survive repeated thermal cycling between 700°C and room temperature and steady-state thermal gradients of about 200°C during irradiation.

The variation of strength with density for both the $CoAl_2O_4$ and $Co_{0.5}Mg_{0.5}O$ followed the standard exponential form, $S = S_O e^{-bP}$, found for most ceramic materials,[5] where

P = fractional porosity

b = empirical exponent (usually between 3 and 9)

S_O = strength at zero porosity, psi

S = strength at porosity, P, psi

This relationship for these two oxides is shown graphically in Figures 1 and 2. The small variation in density (88.5 to 90.2%) of the CoO tested did not allow a similar plot to be obtained for this oxide. The average strength of CoO wafers is 6010 ±555 psi.

Tensile strength is dependent on grain size, as well as porosity. The CoO and solid solution material ($Co_{0.5}Mg_{0.5}O$) have fairly uniform grain sizes of ∿40 μ and ∿35 μ, while the aluminate ($CoAl_2O_4$) has a variable grain size. Most of the aluminate grains are small, ∿5 μ; however, some grains are as large as 100 μ.

Particle Size Distribution of Fragments

The particle size distribution of fragments resulting from the diametrical compression test was measured. Fragmented $Co_{0.5}Mg_{0.5}O$ wafers yielded the least fines (particles less than 44 μ), while fragmented CoO wafers yielded the most fines, as shown in Table I. Most of the fragments are greater than 2 mm with some being approximately one-half the size of the original disk (0.700-in. dia x 0.150-in. thick). As is typical of ceramics of higher strength, the $CoAl_2O_4$ wafers tended to break into larger particles.

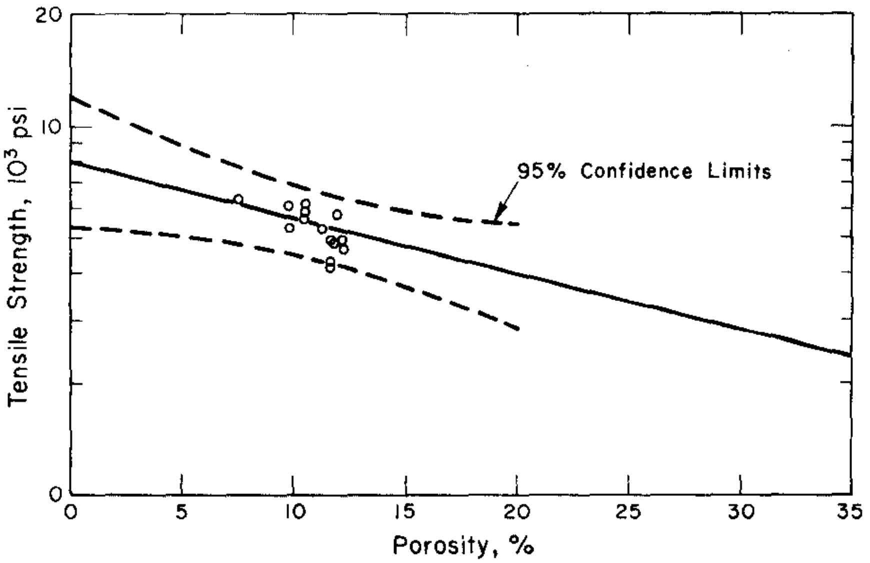


FIG. 1 STRENGTH OF $Co_{0.5}Mg_{0.5}O$

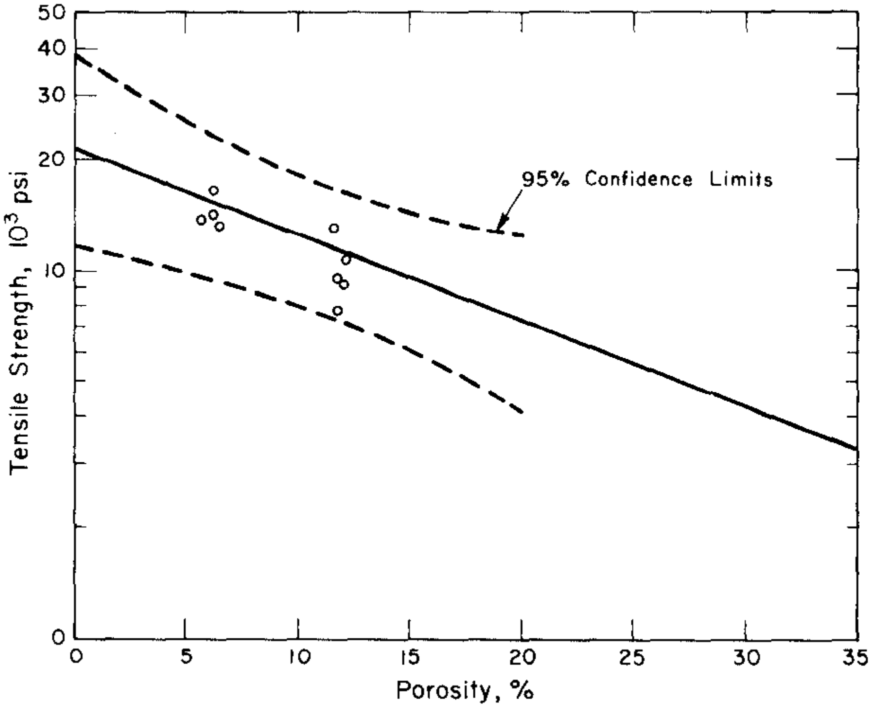


FIG. 2 STRENGTH OF $CoAl_2O_4$

TABLE I

Particle Size After Fracture in Compression

	Weight % Less Than			
	2 mm	0.42 mm	149 μ	44 μ
CoO	1.80	0.45	0.35	0.083
$Co_{0.5}Mg_{0.5}O$	1.68	0.20	0.09	0.003
$CoAl_2O_4$	6.61	0.73	0.19	0.013

The fraction of fines is important in radiological safety analyses because of the danger of inhalation if fines become airborne. These wafers were broken in tension by applying a compressive load across the diameter of the wafers. In this fracture mode, approximately 830 μCi/Ci of fines less than 44 μ (-325 mesh) were produced from CoO compared to 30 μCi/Ci for $Co_{0.5}Mg_{0.5}O$ and 130 μCi/Ci for $CoAl_2O_4$. Failure in other modes may result in different fine particle distributions.

Corrosion in Fresh Water

Preliminary corrosion measurements were made in a fresh water loop on all three cobalt oxides at 125°C. The CoO wafers had the lowest average weight loss and suffered the least surface damage. Two of four $CoAl_2O_4$ wafers had less weight loss and looked as undamaged as the CoO wafers; however, the other two wafers had started to powder and crack, particularly along laminations formed during fabrication. The solid solution, $Co_{0.5}Mg_{0.5}O$, wafers deteriorated severely, and were removed early in the test. The corrosion behavior described below is severe compared to the expected corrosion rates by surface water and streams because under exposure conditions the water will not be pressurized and the surface temperatures should be lower than 125°C.

The tests were made in a pressurized corrosion loop operating at a water temperature of 125°C. The conductivity of the water was nominally 0.5 micro-mho/cm at a pH of 6.1 to 6.2; the flow rate was 10 ft/sec. These water conditions were maintained for approximately 9 weeks of testing. The specimens were nominally 0.700-in. dia x 0.150-in. thick and 90% dense, and were examined periodically during the test.

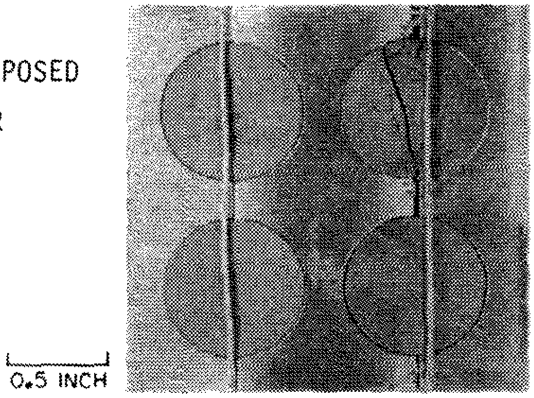
One of the four solid solution wafers swelled and broke up after only two days. This wafer is shown in Figure 3, along with three other solid solution wafers, in the mount used to hold the wafers during testing. When a second solid solution wafer deteriorated after a total of 5 days, testing of solid solution specimens was discontinued. X-ray diffraction analyses showed that magnesium was leached from the solid solution leaving oxidized Co_3O_4 with a corresponding color change from dark red to black as shown in Figure 4. The ratio of cobalt to magnesium in the center region was unchanged from the original value.

The CoO and $CoAl_2O_4$ wafers were also oxidized on the surface during testing. Since the weight gain from oxidation offsets the weight loss due to leaching, interpretation of the net weight loss is uncertain.

The weight losses for CoO and $CoAl_2O_4$ are shown in Figures 5 and 6, respectively. If the wafers were uniformly leached, the rate of loss of radioactivity for CoO ranges from 25 to 175 µCi/day, assuming that the specific activity was 500 Ci/g Co. The rate appears to decrease rapidly with increasing time because of weight gain from oxidation. Similarly for $CoAl_2O_4$ the leach rate ranges from 1.5 to 25 µCi/day over the 9-week period.

Future work will include tests at lower temperatures, particularly with the solid solution specimens, and sea water tests.

FIG. 3 $Co_{0.5}Mg_{0.5}O$ WAFERS EXPOSED 2 DAYS TO 125°C WATER FLOWING AT 10 ft/sec



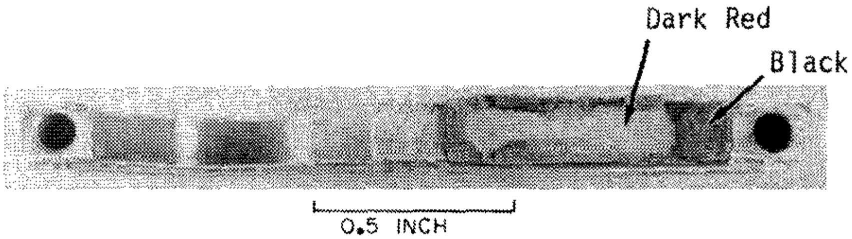


FIG. 4 CROSS SECTION OF $Mg_{0.5}Co_{0.5}O$ WAFER EXPOSED 2 DAYS to 125°C WATER FLOWING AT 10 ft/sec

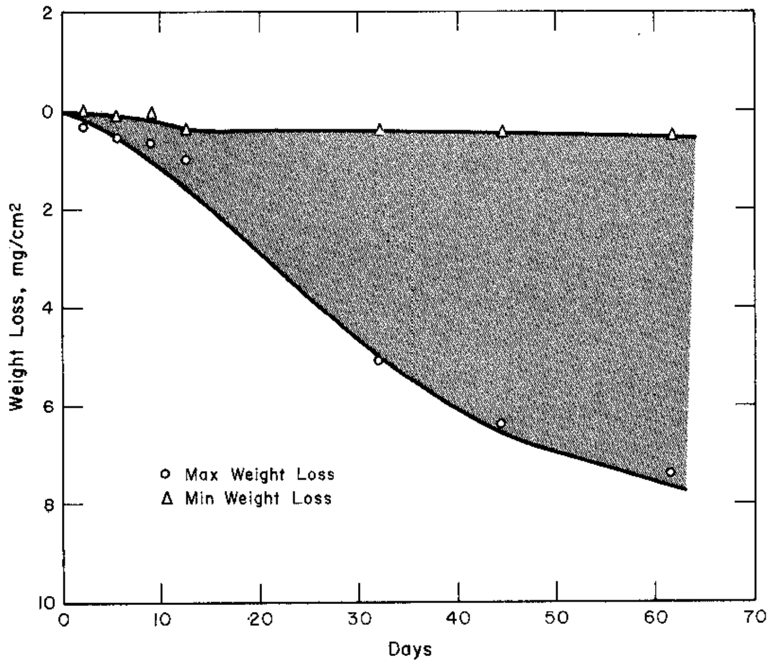


FIG. 5 LEACHING OF CoO (at 125°C in flowing water at pH ~6)

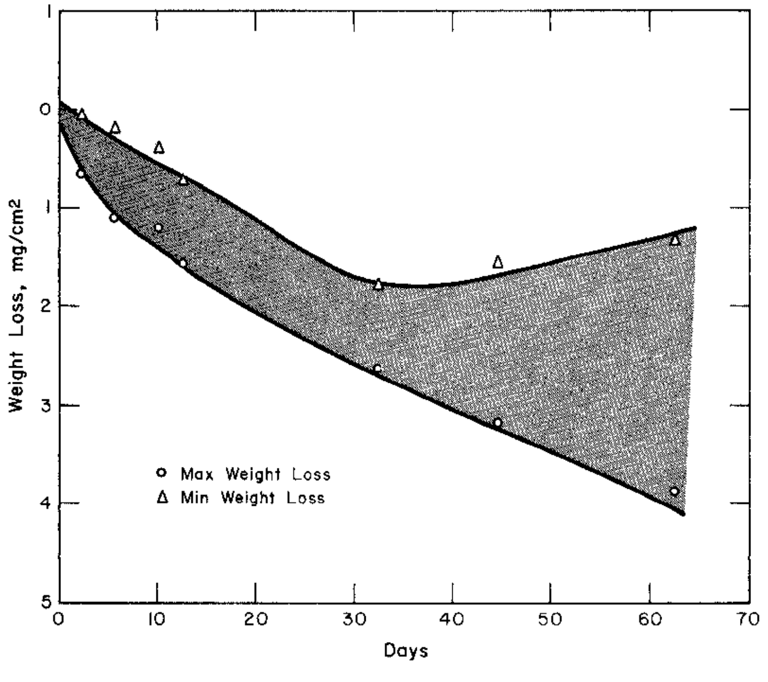


FIG. 6 LEACHING OF $CoAl_2O_4$ (at 125°C in flowing water at pH ~6)

Thermal Conductivity

Thermal conductivities were calculated from thermal diffusivities that were measured at the Columbus Laboratories of Battelle Memorial Institute on wafers supplied by SRL. The thermal conductivities of CoO and $Co_{0.5}Mg_{0.5}O$ are nearly the same, while that of $CoAl_2O_4$ is a factor of 1.5 higher. The conductivities for CoO and $Co_{0.5}Mg_{0.5}O$ are similar to that of Tm_2O_3, but significantly lower than for MgO. The conductivity of $CoAl_2O_4$ is approximately half that of $MgAl_2O_4$ for the temperature range measured.

The thermal diffusivities were measured by the "flash laser" technique. Thin disk-shaped specimens are placed in an isothermal zone of a furnace. The front face of the specimen is irradiated by a laser pulse of short duration. As the heat pulse travels through the specimen, the temperature rise of the back face is recorded as a function of time. The time required for the back face of the specimen to reach one-half of its maximum temperature rise is directly related to the thermal diffusivity of the specimen.

Calculation of the thermal conductivity was made using the relationship,

$$k = \alpha \rho C_p \text{ (cal/sec·cm·°C)},$$

where α is the thermal diffusivity (cm^2/sec), ρ is the density (g/cm^3) and C_p is the heat capacity (cal/g °C). The density of each sample was measured at room temperature. At higher temperatures, the density was corrected for thermal expansion assuming a linear expansion coefficient of $13.5 \times 10^{-6}/°C$, $14.3 \times 10^{-6}/°C$,[6] and $8 \times 10^{-6}/°C$[3] for CoO, $Co_{0.5}Mg_{0.5}O$, and $CoAl_2O_4$, respectively. Heat capacity data reported for CoO and MgO[7] were assumed to be valid over the temperature range of interest. The molar heat capacity of $Co_{0.5}Mg_{0.5}O$ was assumed to be a numerical average of the molar heat capacities of CoO and MgO. Although there are no data available for $CoAl_2O_4$, the value of 0.947 reported for $FeAl_2O_4$[7] should afford a reasonable approximation for these calculations because the heat capacities of CoO and FeO are very similar.

The measured diffusivities along with calculated conductivities are shown in Figures 7-9. Since two specimens of different densities were used for diffusivity measurements, correction for porosity was made using the Loeb equation,[8]

$$k_o = \frac{k_p}{1-P}$$

where k_o is the conductivity of fully dense material, P is the fractional porosity, and k_p is the conductivity at porosity, P.

The thermal conductivities will be used to calculate temperature profiles and cobalt transport in heat sources as well as to reassess thermal stresses occurring during irradiation.

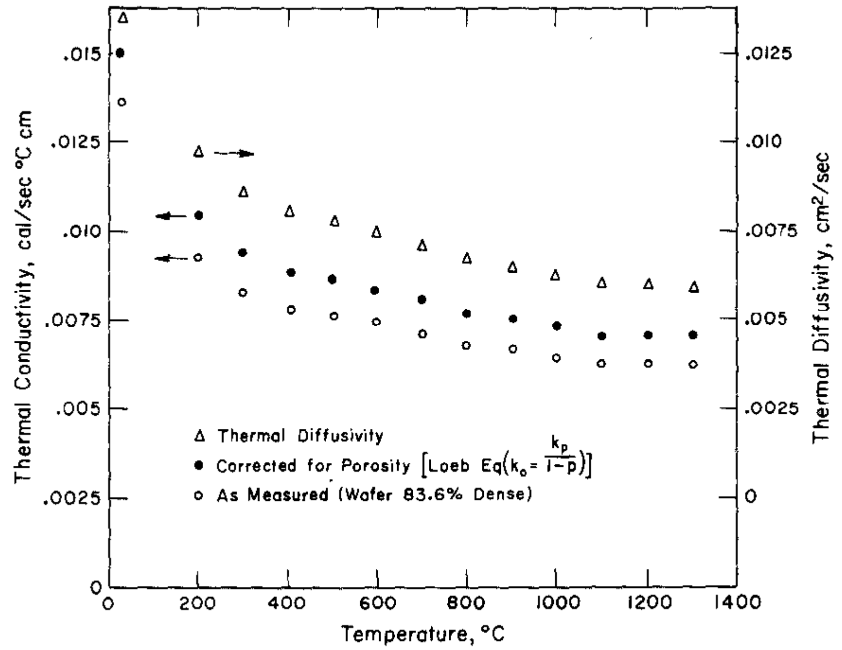


FIG. 7 CoO THERMAL CONDUCTIVITY AND DIFFUSIVITY

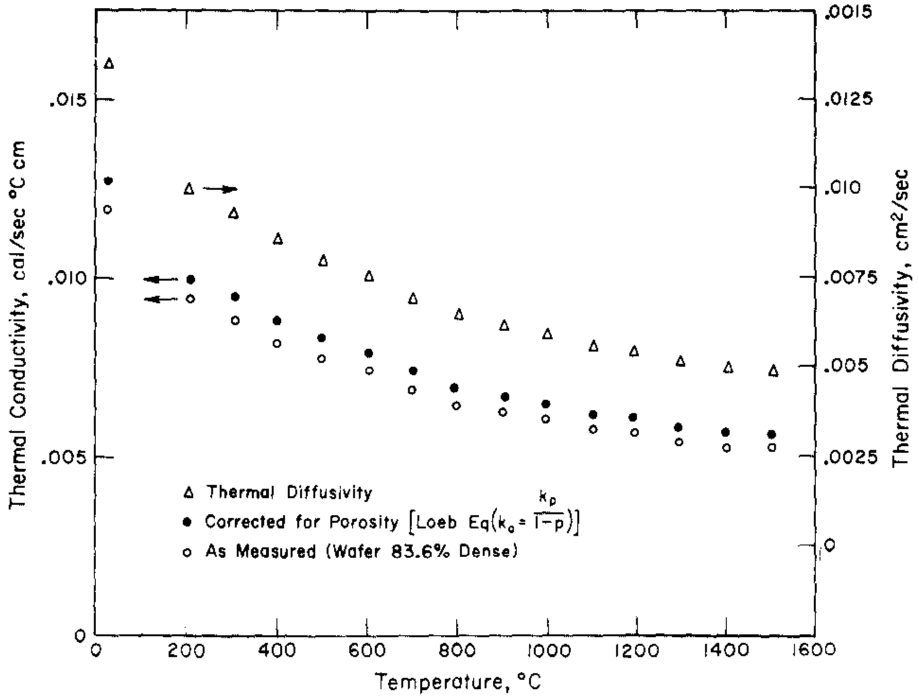


FIG. 8 $Co_{0.5}Mg_{0.5}O$ THERMAL CONDUCTIVITY AND DIFFUSIVITY

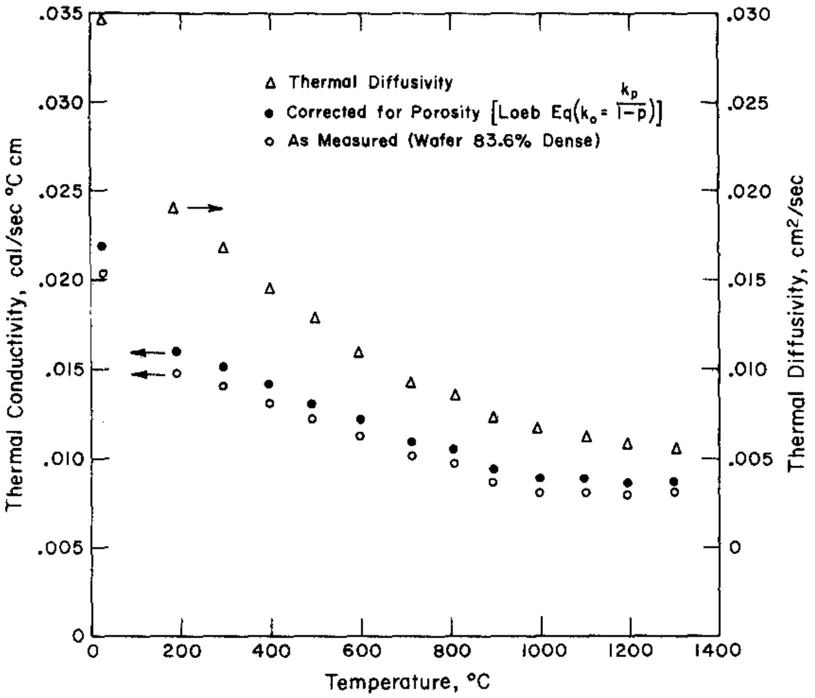


FIG. 9 $CoAl_2O_4$ THERMAL CONDUCTIVITY AND DIFFUSIVITY

CAPSULE FABRICATION AND TESTING

Heating Tests of Capsules with Unirradiated Cobalt Oxides

Refractory Metal Capsules

All planned 1000-hr exposures of the oxides in rhenium capsules have been completed with the exception of the capsules containing CoO, Table II. Vapor transport of rhenium within sealed rhenium capsules caused blistering, bulging, and thinning of the capsule walls after 1000 hr at 1500 and 1700°C, and after 700 hr at 1850°C. Initial metallographic examination revealed that in general the rhenium vaporized from the sides of the capsule and deposited on the oxide wafers and rhenium spacer. There was no evidence of interaction between oxide and rhenium in the condensed phase. Metallographic examination is continuing on these capsules, as well as those containing iridium and rhodium foils.

1500°C Tests

Of five capsules exposed 1000 hr at 1500°C, one containing only $CoAl_2O_4$ wafers developed a leak in the weld. The diameters of all the capsules increased 0.4%. Another capsule containing $CoAl_2O_4$ being heated to a goal exposure of 5000 hr was intact after 1000 hr.

1700°C Tests

One of three capsules exposed for 1000 hr at 1700°C developed dull gray blisters across the wall from the oxide wafers. The capsule, which contained $CoAl_2O_4$, leaked in the region of the blisters. Another capsule containing $Co_{0.5}Mg_{0.5}O$ also leaked and had one blister in the region above the oxide wafers. The third capsule contained $Co_{0.5}Mg_{0.5}O$, did not leak, and had no outside indication of any defects.

1850°C Tests

Three of four capsules exposed for 700 hr at 1850°C, (two containing $Co_{0.5}Mg_{0.5}O$ and one containing $CoAl_2O_4$) were bulged by 0.010 to 0.018 in. (2.0 to 3.6%) in the region of the oxide wafers. Blisters were apparent on the fourth capsule in the region of the $CoAl_2O_4$ wafers, but bulging of only 0.6% was observed. None of the capsules leaked as determined by a helium leak check and a bubble test. One capsule containing $Co_{0.5}Mg_{0.5}O$ is being saved for possible further exposure.

TABLE II

^{59}Co HIGH TEMPERATURE FUELS COMPATIBILITY PROGRAM

		Heating			
Capsule Material and Temperature	Fuel Material	Time, hr	Startup	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	$Co_{0.5}Mg_{0.5}O$	125	9/70	10/70	Weld failed; Re vapor transport
→ Rhenium; foils of Ir, Rh	CoO	1,000	9/70	11/70	Examination in progress
→ Rhenium; foils of Ir, Rh	$Co_{0.5}Mg_{0.5}O$	1,000	9/70	11/70	Examination in progress
→ Rhenium; foils of Ir, Rh	$CoAl_2O_4$	1,000	9/70	11/70	Examination in progress
Rhenium	CoO	1,000	4/71	6/71	
→ Rhenium	$Co_{0.5}Mg_{0.5}O$	1,000	9/70	11/70	Examination in progress
→ Rhenium	$CoAl_2O_4$	1,000	9/70	11/70	Examination in progress
Rhenium	CoO	5,000	4/71	11/71	
Rhenium	$Co_{0.5}Mg_{0.5}O$	5,000	4/71	11/71	
→ Rhenium	$CoAl_2O_4$	5,000	9/70	4/71	Intact after 1000 hr
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	$CoAl_2O_4$	60	9/70	10/70	Weld failed; Re vapor transport
Rhenium	CoO	1,000	4/71	6/71	
→ Rhenium	$Co_{0.5}Mg_{0.5}O$	1,000	9/70	11/70	Examination in progress
→ Rhenium	$CoAl_2O_4$	1,000	9/70	11/70	Examination in progress
Rhenium	CoO	5,000	4/71	11/71	
→ Rhenium	$Co_{0.5}Mg_{0.5}O$	1,000	9/70	11/70	Examination in progress
Rhenium	$CoAl_2O_4$	5,000	4/71	11/71	
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	$Co_{0.5}Mg_{0.5}O$	700	9/70	11/70	Examination in progress
→ Rhenium	$CoAl_2O_4$	700	9/70	11/70	Examination in progress
→ Rhenium	$Co_{0.5}Mg_{0.5}O$	700	9/70	11/70	Reserved for further testing
→ Rhenium	$CoAl_2O_4$	700	9/70	11/70	Reserved for further testing
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	

→ New information reported

Vapor Transport of Rhenium

Metallographic examination has been completed on two rhenium capsules removed from the furnace because their welds leaked. One capsule contained $CoAl_2O_4$ and was exposed 60 hr at 1700°C; the other contained $Co_{0.5}Mg_{0.5}O$ and was exposed for 125 hr at 1500°C. Vapor transport of rhenium was observed in both capsules, Figure 10, but there was no reaction between the oxide and the rhenium.

Vapor transport of rhenium is caused by temperature gradients and is possible because of the relatively high oxygen pressure within the capsule. At 1500°C the pressure of oxygen above CoO is $10^{-5.7}$ atm. Comparing this pressure with the thermodynamic data available for the oxides of rhenium shows that ReO_3 has a higher vapor pressure ($10^{-2.5}$ atm) than the oxides, ReO, ReO_2, or Re_2O_7. ReO_3 is likely to be the carrier for rhenium by the reaction:

$$Re(s) + 3/2O_2(g) = ReO_3(g).$$

The reaction moves to the right at the surface where rhenium is vaporized and to the left where rhenium is deposited.

The kinetics of rhenium transport can be approximated by differentiating the equation for escape of material from a surface;

$$P = \frac{W}{44.33\ At} \sqrt{T/M},$$

where P is the pressure (atm), W the weight of material removed (g), A the area of vaporization (cm^2), t the time (sec), and T the absolute temperature (°K), and M the molecular weight of the transport species. For a rhenium capsule at 1500°C, the result is $\Delta h/t\Delta T = 7.1 \times 10^{-7}$ cm sec^{-1} $°K^{-1}$ where Δh is the change in wall thickness and ΔT is the temperature gradient.* A 0.100-in.-wall capsule with a 0.01°C temperature gradient would be penetrated in 10,000 hr (14 months) at 1500°C.

Similar calculations for cobalt oxide encapsulated in iridium indicate that vapor transport of the capsule material will not be a problem. The transport species can be shown to be IrO_2 and the factor $\Delta h/t\Delta T$ is 5×10^{-15} at 1500°C, eight orders of magnitude less than for rhenium.

An estimate of the temperature gradient driving the rhenium vapor from the walls of the capsules which failed after 1000 hr at 1700°C and 700 hr at 1850°C can be made by using the wall thickness, 0.040 in., as Δh in the relationship for $\Delta h/t\Delta T$. At 1700°C the temperature gradient is calculated to have been 0.005°C and at 1850°C it was 0.003°C. Measurement of Δh from the micrographs at the location of maximum removal of rhenium from the capsules shown in Figure 10 results in a calculated ΔT of 0.06°C for the capsule exposed to 1500°C and 0.02°C for the capsule exposed at 1700°C.

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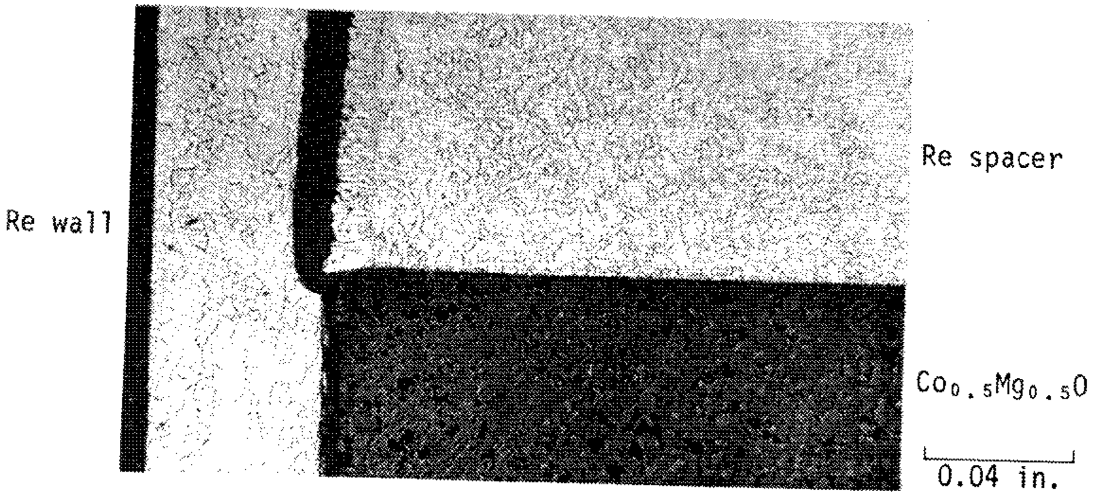
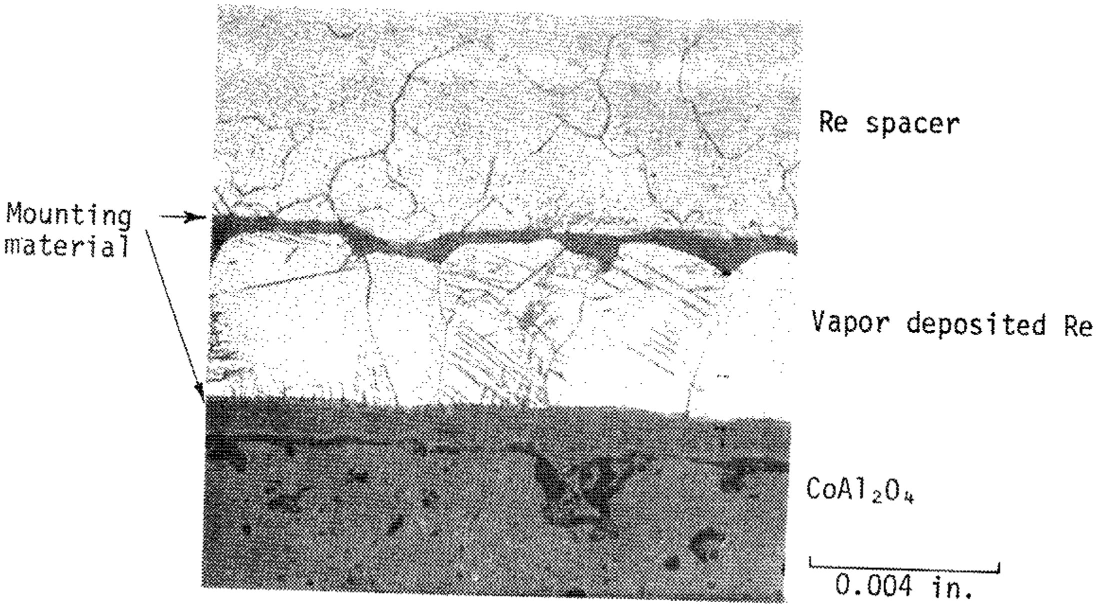
* $\Delta h/t\Delta T = 5.6 \times 10^{-6}$ cm sec^{-1} $°K^{-1}$ at 1700°C and 1.4×10^{-5} cm sec^{-1} $°K^{-1}$ at 1850°C.



A. Rhenium vaporized from wall and deposited on spacer in capsule exposed 125 hours at 1500°C



B. Rhenium deposited between spacer and oxide wafer in capsule exposed for 60 hours at 1700°C

FIG. 10 RHENIUM TRANSPORT IN SHORT-TERM TESTS

Heating Tests of Capsules with Unirradiated Cobalt Metal

Superalloy Capsules

Seven capsules being heated at 850 to 1000°C toward goal exposures of 50,000 hr, Table III, were examined nondestructively after 17,000 to 30,000 hr to determine their preformance to date. All but the "Haynes"* 25 capsule that had been heated at 1000°C were intact and diameter increases were small, <0.002 in. Heating of the six intact capsules was resumed.

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	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	Intact at 26,800 hr
	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	Capsule intact
	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	Intact at 17,000 hr
	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	Intact at 29,300 hr
	→ 10,000[d]	1,000	95	1	11-68	1-70	No oxidation of Co or capsule
"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	Intact at 29,300 hr
	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	Intact at 29,300 hr
"Haynes" 25	→ 10,000	850	95	1	11-68	1-70	Capsule intact
(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
	→ 29,300	1,000	95	1	10-67	11-70	Failed at 29,300 hr from oxidation
	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	Intact at 22,500 hr
	20,000	1,000	95	1	5-68	9-70	Capsule intact
	→ 22,500	1,000	95	1	5-68	12-70	Capsule intact

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[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.

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* Trademark of Cabot Corp.

"Haynes" 25 Oxidation

The "Haynes" 25 capsule oxidized severely, resulting in loss of weld integrity. Nondestructive examination of this same capsule after 11,500 hr had shown that the capsule was intact. The oxidation reduced the wall thickness from 0.095 in. to <0.050 in., as shown in Figure 11, by comparison with a companion "Haynes" capsule that was intact after 20,000 hr at 1000°C.[9] This amount of oxidation is essentially equivalent to the initial weld penetration and, therefore, caused the loss of weld integrity.

Rapid oxidation of "Haynes" 25 has been observed in previous tests for 3000 hr at 1125°C[10] and has been reported in the literature to be related to the silicon and manganese contents of the particular heat of the alloy.[11] The composition of the particular heat from which the capsule was fabricated falls in the transition region between stable (parabolic) and unstable oxidation behavior reported in Reference 11. This same work indicated that the threshold for unstable behavior could be time and temperature dependent.

There were two distinguishing features in the microstructure of the oxidized capsule that were not evident in any of the previous capsules or oxidation-test coupons heated at less severe conditions. First, the zone depleted in alloying elements by the oxidation process was narrower than expected, Figure 12. This observation might be explained by the mechanism for the rapid oxidation, proposed by the NASA workers,[11] in which small areas of underlying metal fracture to relieve stresses in the oxide and subsequently oxidize or spall off with the oxide. The second characteristic was the formation of a phase, believed to be sigma (Ni-Cr compound) from its metallographic appearance, that decreased in concentration with increasing distance from the surface, Figure 13. The alloying elements (manganese, tungsten, etc.) initially present in this alloy normally inhibit the formation of sigma.

Additional work is being planned to understand the oxidation of "Haynes" 25. Except for this oxidation behavior, "Haynes" 25 is a more attractive encapsulating material for many applications than either "Hastelloy"* C, "Hastelloy" X, or "Inconel"** 600 because of its higher strength and lower ^{60}Co diffusivity.

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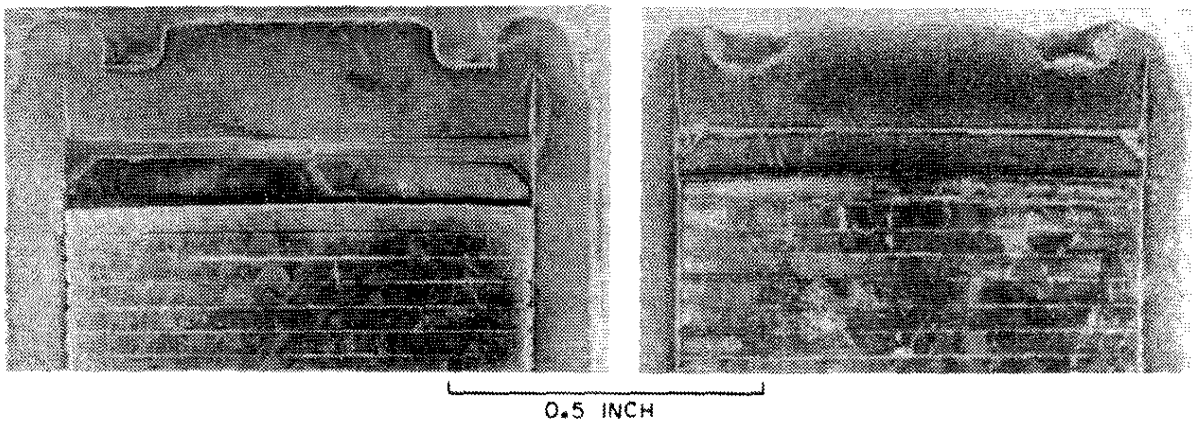
* Trademark of Cabot Corp.

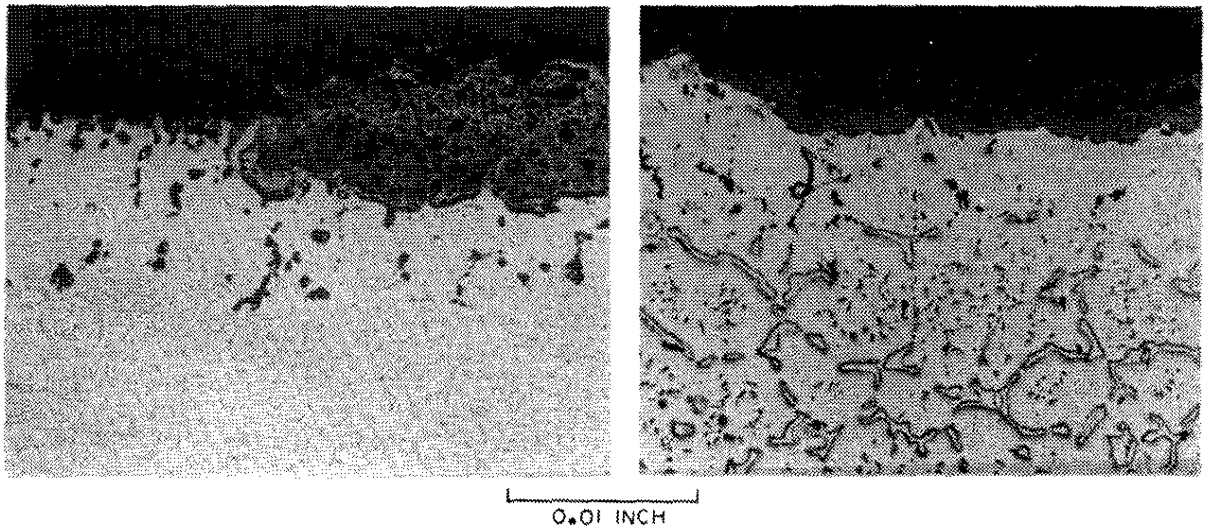
** Trademark of International Nickel Co.


20,000 hr

29,300 hr

Note thinning of capsule wall and rounding of cap corners in capsule heated 29,300 hr

FIG. 11 "HAYNES" 25 CAPSULES HEATED AT 1000°C


20,000 hr

29,300 hr

The alloy depleted zone in capsule heated 29,300 hr is ~0.008 in. thick, the same as in capsule heated 20,000 hr.

FIG. 12 COMPARISON OF OXIDATION ZONES IN "HAYNES" 25 CAPSULES HEATED AT 1000°C

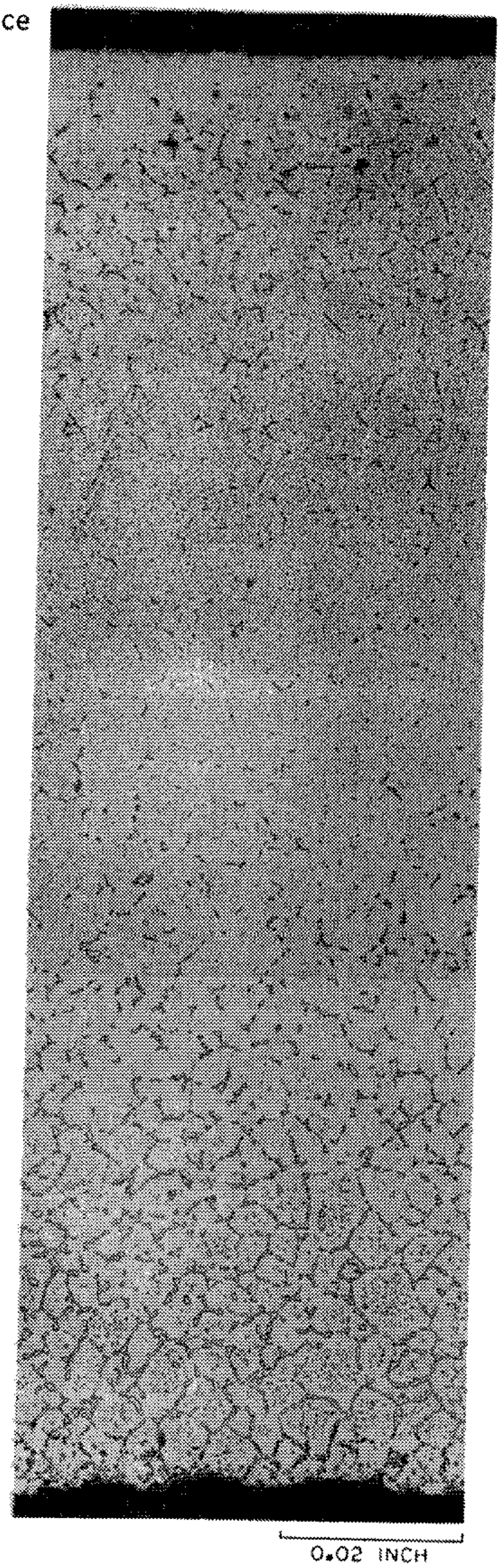


FIG. 13 VARIATION OF STRUCTURE IN CAP OF "HAYNES" 25 CAPSULE

10,000-hr Capsules

Examination was completed on three capsules removed from test previously[12]; two "Inconel" 600 capsules that had been heated for 10,000 hr at 900 and 1000°C, respectively, and a "Haynes" 25 capsule that had been heated for 10,000 hr at 850°C. All three capsules were intact and the increases in diameter were small, <0.002 in. The total depths affected by oxidation of the external surfaces were in agreement with previous tests. In the majority of the areas examined there was little bonding between the cobalt and the walls of the capsules. Where the contact was intimate the widths of the resultant reaction zones were in general agreement with previous tests. Review of initial dimensions indicated that the cobalt-capsule clearances may have been up to 0.004 in. larger in these capsules than in those used in previous tests.

The "Inconel" 600 capsule heated at 1000°C had an internal atmosphere of air instead of helium as used in the majority of other capsules tested. No oxidation of the cobalt or the interior capsule surfaces was evident.

A "Hastelloy" X capsule being heated at 1000°C was removed from test after 22,500 hr instead of the intended 30,000 hr to provide additional data on this material for the WANL 30 kw(t) Demonstration Unit (See section below). The capsule was intact. The depth affected by oxidation (0.008 in.) and the width of the compatibility zone (0.017 in.) were as predicted by extrapolation of previous tests.[9]

Refractory Metal Capsules

The cobalt metal melted and penetrated the walls of five tungsten-25 wt % rhenium alloy capsules while they were being heated at an indicated temperature of 1200°C. The melting was detected when a routine check of the control thermocouples with an optical pyrometer indicated a furnace temperature of 1550°C. The furnace had been at an indicated 1200°C for only 20 min following a shutdown to repair a leak in one of the cooling-water lines. No overtemperature alarm (set at 1350°C) was experienced and the continuous record of furnace temperature showed no excursions. The current-limit control on furnace power was at a setting corresponding to 1400°C. Recalibration of the thermocouples after the incident showed that their output was normal. Examination of the furnace control circuits showed no abnormalities and the furnace has been operated for ∿300 hr since the incident with no additional temperature excursions as indicated by repeated calibrations of the control thermocouples with an optical pyrometer.

Experiments currently in progress will test the hypothesis that the thermocouples were in contact, or near contact, with the molybdenum heat shields. A gap ∿1/8-in. wide existed between the pieces of ceramic insulation at the time of the temperature excursion and could have allowed the postulated contact. Preliminary results indicate that a difference of over 150°C between indicated and actual temperature is produced by this thermocouple configuration.

Heating tests are continuing with two tungsten capsules at 1200°C and two W-Re capsules at 1400°C, Table IV. These tests have been interrupted several times because of malfunctions in the furnace cooling-water systems; the expected completion dates for these tests, shown in Table IV, have been revised to reflect these delays.

TABLE IV

^{59}Co — Refractory Metal Capsule Tests[a]

Capsule Material	Heating		Approx. Starting Date	Approx. Completion Date	Welding Diameter, inch	Welding Technique	Remarks
	Time, hr	Temp, °C					
Tungsten	1,000	1,200	9/70	3/71	0.745	TIG	
	5,000	1,200	9/70	5/71	0.745	TIG	
W-25 wt % Re	1,000	1,200	8/70	10/70	0.745	TIG	Capsule intact
	1,000	1,400	8/70	10/70	0.745	TIG	Capsule intact
	5,000	1,400	8/70	5/71	0.745	TIG	
	10,000	1,400	8/70	12/71	0.745	TIG	

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[a] One capsule containing ∿10 Co wafers 0.073 in. thick at each listed condition.

Heating Tests of Capsules with Irradiated Cobalt Metal

Superalloy Capsules

An "Inconel" 600 capsule containing initially 13,700 Ci of ^{60}Co was removed from test after 20,000 hr at 900°C, Table V. Examination of this capsule and those removed from test earlier after 5000 to 10,000 hr at 900 and 1000°C has been delayed until the coming quarter due to scheduling difficulties in the High Level Caves. Heating of four other capsules to 20,000 hr is continuing; capsules with goal exposures of 50,000 hr are in temporary storage.[9]

TABLE V

Summary of ^{60}Co Capsule Heating Tests

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 (m.p. 1370°C)	130	850[a]	50	1	120	16,000	2-67	2-67	Swelled due to overheating
	1,000	∿900	50	1	100	5,000	4-67	6-67	Capsule intact
	5,000	∿900	50	1	150[b]	15,000	4-67	10-67	Capsule intact
	10,000	∿900	50	1	150[b]	15,000	4-67	6-68	Increased Co/capsule reaction
	10,000	∿900	50	1	150[b]	9,000	5-67	10-68	Increased Co/capsule reaction
	10,000	900	95	1	255[c]	36,500	2-68	8-69	Examination in progress
	→ 20,000	900	95	1	288[c]	13,700	7-68	11-70	Examination in progress
	50,000	900	95	1	282[c]	13,400	7-68	4-74	Examination in progress
	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 (m.p. 1270°C)	100	850	50	1	120	9,000	1-67	1-67	Examination in progress
	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	1-75		
"Haynes" 25 (m.p. 1330°C)	5,000	1,000	95	1	263[c]	12,500	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	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 (m.p. 1260°C)	5,000	1,000	95	1	250[c]	11,900	9-68	4-69	Examination in progress
	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	

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[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

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 2 megacuries (30 kw) of ^{60}Co. Westinghouse Astronuclear Laboratory (WANL) is the contractor for this project.

A final review of design and material selection for the rechargeable fuel pins and their relationship to the Nickel-201 heat source core resulted in the following decisions.

- Retain original design in which no liner is provided to protect the Ni-201 core from oxidation. Minimize oxidation by the addition of a replaceable metallic seal above the fuel pin.
- Provide as much clearance as possible ($\sim$0.080-in. radial) between the fuel pin and the bore to minimize the possibility of bonding. Provide a ceramic shoulder to support the fuel pin at the top and a ceramic guide to center the fuel pin at the bottom.
- Specify "Hastelloy" X for fuel pin and capsules, primarily because of its excellent and predictable oxidation resistance. Outgas the components and control the internal pressure during fabrication to minimize creep. 0.070-in.-wall capsules inside a 0.100-in.-wall pin should provide adequate protection against distortion and ^{60}Co diffusion.
- Provide the capability to backfill the space between fuel pins and core with helium.
- Investigate incentives for reducing the primary coolant temperature and evaluate the effect of such a change on the heat pipe design.

^{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/g 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 VI.

TABLE VI

^{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
DP-1254	July - September 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).

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

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

C. L. Angerman and J. P. Faraci. "Heating Tests of Encapsulated Cobalt Heat Sources", *Nuclear Metallurgy, Vol. 14, Symposium on Materials for Radio-Isotope Heat Sources*, D. E. Thomas, W. O. Harms, and R. T. Huntoon (Editors), American Institute of Mining, Metallurgical, and Petroleum Engineers, Inc., New York, New York, pages 309-22 (1969).

J. A. Donovan and W. R. McDonell. "Cobalt-Rhenium Alloys for High Temperature ^{60}Co Heat Sources", *Trans. Amer. Nucl. Soc. 12(2)*,480-1 (1969).

P. K. Smith, P. E. McBeath, J. R. Keski, W. R. Kanne, Jr., and W. C. Mosley, "Cobalt-60 Oxides as Thermonic Fuels," *IEEE Conference Record of 1969 Thermonic Conversion Specialists Conference, October 21-23, 1969, Carmel, California.* 69 C 51-ED, pp 417-24, The Institute of Electrical and Electronics Engineer, Inc., New York (1969).

S. Mirshak, C. P. Ross, and V. Whatley. "Cobalt-60 for Heat and Power," *Chemical Engineering Progress Symposium Series No. 106 Application of Radioisotopes, Nuclear Engineering Part XXII, Vol. 66*, L. H. Meyer (ed.). American Institute of Chemical Engineers, New York, 7-15 (1970).

C. P. Ross. "AEC ^{60}Co Heat Source Programs." *Trans. Amer. Nucl. Soc. 13(2)*, 492-3 (1970).

REFERENCES

1. C. L. Angerman and C. P. Ross. "Savannah River Laboratory Cobalt-60 Power and Heat Source, Quarterly Progress Report, October-December 1969." USAEC Report DP-1226, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, S. C. 29801 (1970).

2. A. Rudnick, A. R. Hunter, and F. C. Holden. "An Analysis of the Diametrial-Compression Test." *Materials Research Standards, 3(4)*, 283-89 (1963).

3. J. R. Hague, J. F. Lynch, A. Rudnick, F. C. Holden, and W. H. Duckworth. "Refractory Ceramics for Aerospace." The American Ceramic Society, Inc., Columbus (1964).

4. W. D. Kingery. "Factors Affecting Thermal Stress Resistance of Ceramic Materials." *J. Amer. Ceram. Soc. 38(1)*, 3-15 (1955).

5. W. H. Duckworth. "Discussion of Raphkeivetch Paper." *J. Amer. Ceram. Soc. 36(2)*, 68 (1953).

6. P. K. Smith, J. R. Keski, P. E. McBeath, W. R. Kanne, and W. C. Mosley. "Cobalt-60 Oxides in Thermoinic Fuels." *IEEE Conference Record of 1969 Thermoinic Conversion Specialist Conference*, 417-24 (1969).

7. K. K. Kelly. "Contributions to the Data on Theoretical Metallurgy XIII. High Temperature Heat-Content, Heat Capacity and Entropy Data for the Elements and Inorganic Compound." Bulletin 584, Bureau of Mines (1960).

8. A. L. Lock. "Thermal Conductivity (VIII). A Theory of Thermal Conductivity of Porous Materials." *J. Amer. Ceram. Soc. 37*, 96 (1954).

9. C. L. Angerman and C. P. Ross. "Savannah River Laboratory Cobalt-60 Power and Heat Source, Quarterly Progress Report, July-September 1970. USAEC Report DP-1254, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, S. C. 29801 (1971).

10. H. S. Hilborn. "Savannah River Laboratory Isotopic Power and Heat Sources, Quarterly Progress Report, July-September 1968, Part I - Cobalt." USAEC Report DP-1177-I, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, S. C. 29801 (1968).

11. J. S. Wolf and G. D. Sandrock. "Some Observations Concerning the Oxidation of the Cobalt-Base Superalloy L-605 (HS-25)." NASA TN D-4715, National Aeronautics and Space Administration, Lewis Research Center, Cleveland, Ohio.

12. C. L. Angerman and C. P. Ross. "Savannah River Laboratory Cobalt-60 Power and Heat Sources, Quarterly Progress Report, April-June 1970. USAEC Report DP-1247, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, S. C. 29801 (1970).