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AEC RESEARCH AND DEVELOPMENT REPORT

SAVANNAH RIVER LABORATORY ISOTOPIC POWER AND HEAT SOURCES

QUARTERLY PROGRESS REPORT

APRIL - JUNE 1967

PART I - COBALT-60

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Savannah River Laboratory

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July 1967

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SAVANNAH RIVER LABORATORY
AIKEN, S. C. 29801

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PREFACE

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

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

1. Properties and reactions of isotopes useful or potentially useful as heat sources.

2. Information on the irradiation and postirradiation processing of these materials, when the information is relevant to their use as heat sources and is not in a sensitive area of production technology.

3. Development of design data directed toward manufacturing capability for isotopic heat sources.

The report is issued in two parts: Part I includes only information on cobalt; Part II includes information on the other isotopic heat source materials. Both parts contain principally data from work in April, May, and June 1967. The first report in the series, DP-1088, which was issued as a single volume, presented some background data and reported the data from work in July, August, and September 1966; the second report, DP-1094, which was also a single volume, reported the data from work in October, November, and December 1966; the third report, DP-1105, which was the first to be issued in two parts, reported the data from work in January, February, and March 1967.

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SUMMARY

A total of 8.8 million curies (137 kw) of ^{60}Co was discharged from the reactors during the quarter, including over 5-1/2 million curies as 0.745-inch-diameter by 0.040-inch-thick wafers with specific activities between 300 and 400 Ci/g. (pg 1)

Measured diffusion profiles of ^{60}Co into encapsulating alloys showed that small but significant concentrations of ^{60}Co extend beyond the observable reaction zone. (pg 5)

An "Inconel" 600 capsule containing radioactive cobalt was heated in air at 850°C for 1000 hr without significant changes in dimensions or appearance. (pg 11)

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DISCUSSION

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. Specific objectives are to establish allowable operating limits for:

- Capsules of radioactive cobalt metal contained in an oxidation resistant alloy (by the end of FY-1968), and
- Capsules of a high temperature fuel form of ^{60}Co contained in a refractory alloy (on a low priority basis).

The program is a limited one to meet needs of general applicability to heat source designers and users. Development of specific concepts is not at present included in the scope of the SRL program.

PRODUCTION OF ^{60}Co FOR HEAT SOURCE DEVELOPMENT

A total of 8.8 million curies (137 kw) of high-activity cobalt was discharged from the reactors during the quarter. This ^{60}Co, which includes about 5-1/2 million curies as 0.745-inch-diameter by 0.040-inch-thick wafers with activities between 300 and 400 curies per gram, was produced for experimental programs and possible heat source demonstrations. Table I gives the activities of the various cobalt shapes, as estimated from assays of representative pieces.

MATERIALS TECHNOLOGY AND DEVELOPMENT

Properties of Irradiated Cobalt Metal

Measurements of the hardnesses of five specimens with different levels of specific activity indicated a trend of increasing hardness with increasing nickel content resulting from radioactive decay.[1] No hardness increase is expected from decay per se because the production of point defects by the gamma irradiation is expected to be negligible. The dependency of hardness on nickel content was confirmed using arc-melted and homogenized alloys of unirradiated cobalt and 10 to 80 wt % nickel. The absolute values of the hardnesses may have been affected by the difference in structure (as-cast unirradiated alloys compared with wrought irradiated wafers) and the presence of small gradient of impurities from the surface to the center

TABLE I

^{60}Co for Heat Sources[a]

Cobalt Shape	Average Activity, Ci/g	No. of Pieces	Total Activity, 10^6 Ci	Total Power, kw
0.745-inch Wafers				
Fuel form - cobalt metal (wrought)	400	2280	2.27	35.4
Fuel shape - wafers 0.745 ±0.001-inch diameter	350	912	0.80	12.5
0.040 ±0.003-inch thick	330	1520	1.25	19.5
including 0.0005 to 0.001-inch Ni plate	300	1520	1.14	17.8
Cobalt density - 8.80 ±0.05 g/cm^3	320	152	0.12	1.9[b]
		6384	5.58	87.1
0.800-inch Wafers				
Fuel form - cobalt metal (sintered)	390	255	0.28	4.4[c]
Fuel shape - wafers 0.800 ±0.001-inch diameter	410	136	0.16	2.5[d]
0.040 ±0.003-inch thick	350	136	0.13	2.0
including 0.0005 to 0.001-inch Ni plate		527	0.57	8.9
Cobalt density - 8.60 ±0.10 g/cm^3				
Nickel-Plated Slabs				
Fuel form - cobalt metal (wrought)	390	45	0.29	4.5
Fuel shape - slabs 3.00 ±0.03-inch long	360	48	0.29	4.5
0.640 ±0.002-inch wide		93	0.58	9.0
0.060 ±0.001-inch thick				
including 0.0005 to 0.001-inch Ni plate				
Cobalt density - 8.80 ±0.05 g/cm^3				
Stainless Steel-Canned Slabs				
Fuel form - cobalt metal (wrought)	390	45	0.24	3.7
Fuel shape - slabs 2.96 ±0.03-inch long	330	48	0.22	3.4
0.735 - 0.740-inch wide		93	0.46	7.1
0.092 ±0.001-inch thick				
sheath thickness 0.015-inch min				
cobalt dimensions same as nickel-plated slabs above except 2.44-inch long				
Cobalt density - 8.80 ±0.05 g/cm^3				
Stainless Steel-Coextruded Slabs				
Fuel form - cobalt metal (wrought)	390	60	0.27	4.2
Fuel shape - slabs 3.00 ±0.03-inch long	360	64	0.27	4.2
0.740 ±0.002-inch wide		124	0.54	8.4
0.072 ±0.002-inch thick				
SST thickness 0.015-inch min				
Cobalt dimensions: 2.75-inch long				
0.71-inch wide				
0.042-inch thick				
Cobalt density - 8.80 ±0.05 g/cm^3				
1-cm-dia x 1-mm-thick wafers	540		0.08	1.2
1/2-cm-dia x 1-mm-thick wafers	580		0.15	2.3
1/16-in-dia x 1/16-in-thick pellets	590		0.23	3.6
1-mm-dia x 1-mm-thick pellets	590		0.17	2.7
6-in- and 3-in-long stainless-canned slabs	560		0.46	7.2
			1.09	17.0
Grand Total			8.82 MCi	137.5 kw

(a) Activity and power as of June 30, 1967.
(b) Wafers have central hole of 0.070-inch diameter.
(c) 110 wafers have experimental compositions.
(d) 68 wafers have central hole of 0.070-inch diameter.

of the arc-melted samples. However, the cast alloys also showed an increase in hardness with increasing nickel content up to 20 wt % Ni.[2]

High-Temperature Fuel Candidates

Co-Re Alloys

Alloys and compounds of cobalt that have higher melting temperatures than cobalt metal are being explored for use in high-temperature ^{60}Co heat sources. Cobalt-rhenium alloys have sufficiently high melting temperatures but would have to be fabricated after irradiation because rhenium has a high cross-section for absorption of thermal neutrons and would be converted to osmium with the concurrent formation of a lower-melting alloy. Methods for fabricating Co-Re alloys are being studied that would be suitable for use with irradiated cobalt.

CoO-MgO Solid Solutions

Ceramic forms of CoO-MgO solid solutions are being appraised for heat sources because they have high melting temperatures and could be fabricated before irradiation.

Evaluation of Encapsulating Materials for Cobalt Metal

Selection of suitable encapsulating materials and definition of their limiting operating conditions requires the assessment of their compatibility with cobalt. Data that are required include:

- Rates of diffusion of ^{60}Co through capsule walls,
- Rates of oxidation of the capsule alloys, and
- Effects of these reactions on the mechanical properties of the materials.

Direct measurements of these characteristics are supplemented by tests in which capsules fabricated from the most promising materials and containing unirradiated cobalt are annealed at up to 1000°C for up to 10,000 hr. Effects of the radiation field are being measured in a smaller number of companion tests on similar capsules containing ^{60}Co.

Compatibility

Previous tests of interdiffusion of unirradiated cobalt with encapsulating materials in multilayer couples, for 68 hr at up to 1200°C, showed that the visible diffusion zone -- a region of solid solution terminating in a band of voids -- grew slowest in the nickel-based alloys "Inconel"* 600, TD Nickel**, TD Nickel-Chromium**, and "Hastelloy"† C, and in the cobalt-based alloy "Haynes"† 25.[1,3]

Additional tests showed that the growth of the visible zone was acceptably slow with "Haynes Experimental Alloy"† 8188, but not with "Tophet"‡ A, Figure 1. The calculated width of the zone after five years at 1000°C would be <0.050 inch with "Haynes" 8188, but >0.050 inch in "Tophet" A. These two alloys were tested because they had acceptable oxidation resistance in 1000-hr tests at 1000°C.[4]

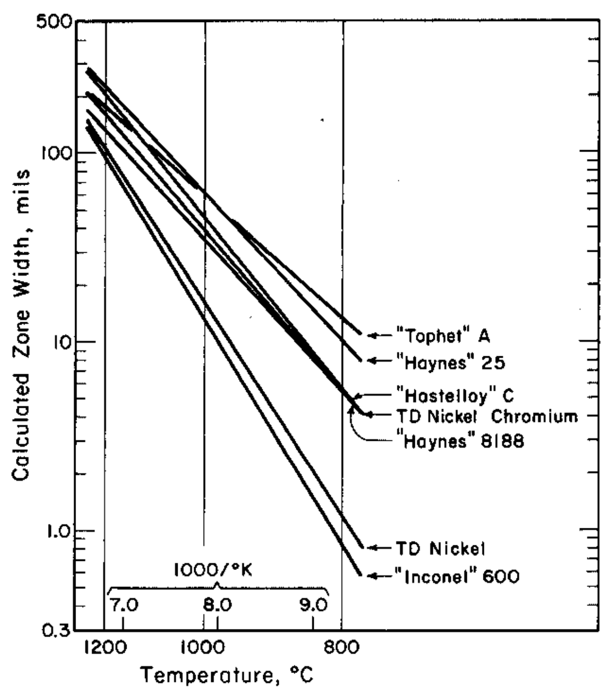


FIG. 1 CALCULATED WIDTH OF DIFFUSION ZONES AFTER ONE HALF-LIFE OF ^{60}Co
Based on 168-hr Heating

Diffusion of ^{60}Co

The rate of diffusion of ^{60}Co through potential capsule alloys is being determined by radiotracer techniques.

Diffusion of ^{60}Co atoms through the wall may be one of the limiting factors in the design of capsules for applications where the capsule is the only barrier to the spread of contamination. Measured diffusion profiles of ^{60}Co into candidate alloys show that small, but significant, ^{60}Co concentrations extend well beyond the metallurgically observable reaction zone between the cobalt and the capsule wall. Figure 2 shows the measured concentration of ^{60}Co in "Inconel" 600 after 100 hours and the calculated concentration profile in a capsule wall after one year at 800°C.

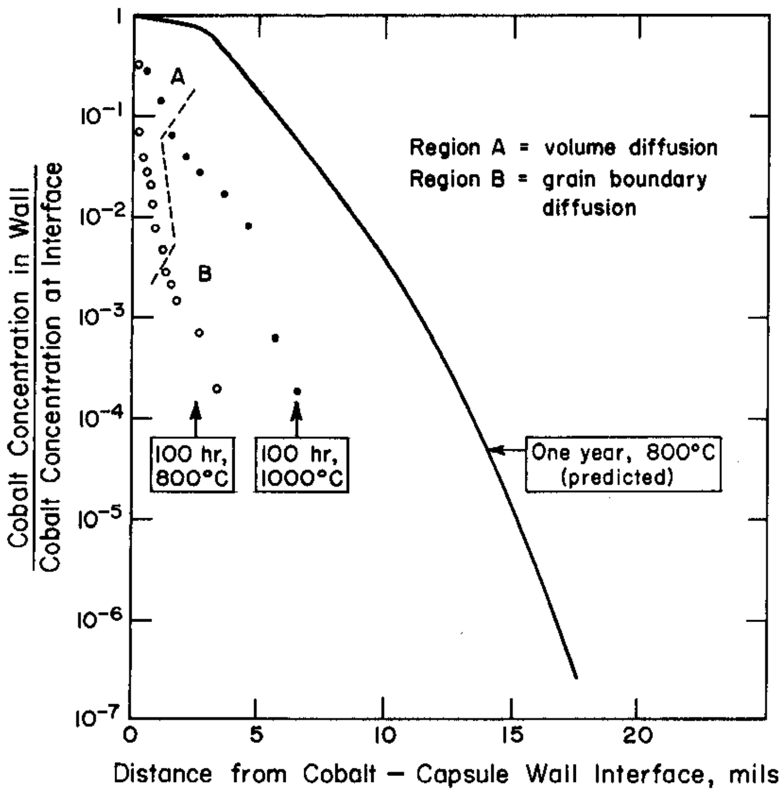


FIG. 2 DIFFUSION OF COBALT THROUGH "INCONEL" 600

* International Nickel Co., Inc., New York, N. Y.
** E. I. du Pont de Nemours and Co., Wilmington, Del.
† Union Carbide Corp., New York, N. Y.
‡ Wilbur B. Driver Co., Newark, N. J.

The allowable concentration of ^{60}Co in that portion of the capsule wall affected by corrosion is dependent on the total amount of corrosion product and the specific application. What constitutes an allowable ^{60}Co concentration can be calculated based on a typical heat source. Assume a 10 kwt heat source using 0.75-inch-diameter capsules, each with an active length of 1.9 inches. If the outer 0.005 inch of each capsule surface were removed by corrosion, this material would weigh about 43 g. If this material contained sufficient ^{60}Co (7.4×10^{-3} Ci) to produce a dose rate of 10 mr/hr at 3 feet without shielding, it would have a specific activity of 1.7×10^{-4} Ci/g.

The inside surface of the capsule is about 50% original metal and 50% cobalt and thus has a specific activity of about 150 Ci/g. The outside surface activity then would be about one millionth of the inside surface activity. This requirement of a 10^6 reduction in activity can be changed by other assumptions, but reference to Figure 2 shows that order of magnitude changes in the concentration requirement will have only a small effect on the wall thickness requirement.

Rate of Oxidation

The oxidation resistance of numerous nickel- and cobalt-based alloys is being measured by exposing test coupons to still air at 1000°C for times up to 10,000 hr. Examination of the first group of alloys after 1000 hr showed that TD Nickel-Chromium had outstanding resistance to oxidation and that "Inconel" 600, "Hastelloy" C, and "Hastelloy" X had satisfactory resistances.[1] Examination of the second group after 1000 hr showed that none of the alloys equaled the resistance of TD Nickel-Chromium, but "Haynes Experimental Alloy" 8188 and "Tophet" A were equivalent to the two "Hastelloys."[4]

Tests of the first group show after 5000 hr that TD Nickel-Chromium is best, followed -- with acceptable quality -- by "Hastelloy" X, "Hastelloy" C, and "Inconel" 600, Tables II and III. The superiority of the TD Nickel-Chromium alloy is due principally to resistance to intergranular penetration. Penetration was essentially zero in this alloy, but as deep as 0.0088 inch in the other alloys.

In addition to scaling and intergranular penetration, air oxidation also affected a zone up to 0.030-inch deep (depending on the alloy) in which as many as three reactions were observed, Figure 3:

- Formation of large "oxide" particles with appearance similar to surface scale

TABLE II

Oxidation of Candidate Encapsulating Alloys

Still air at 1000°C for 5000 hr
Alloy compositions in following table

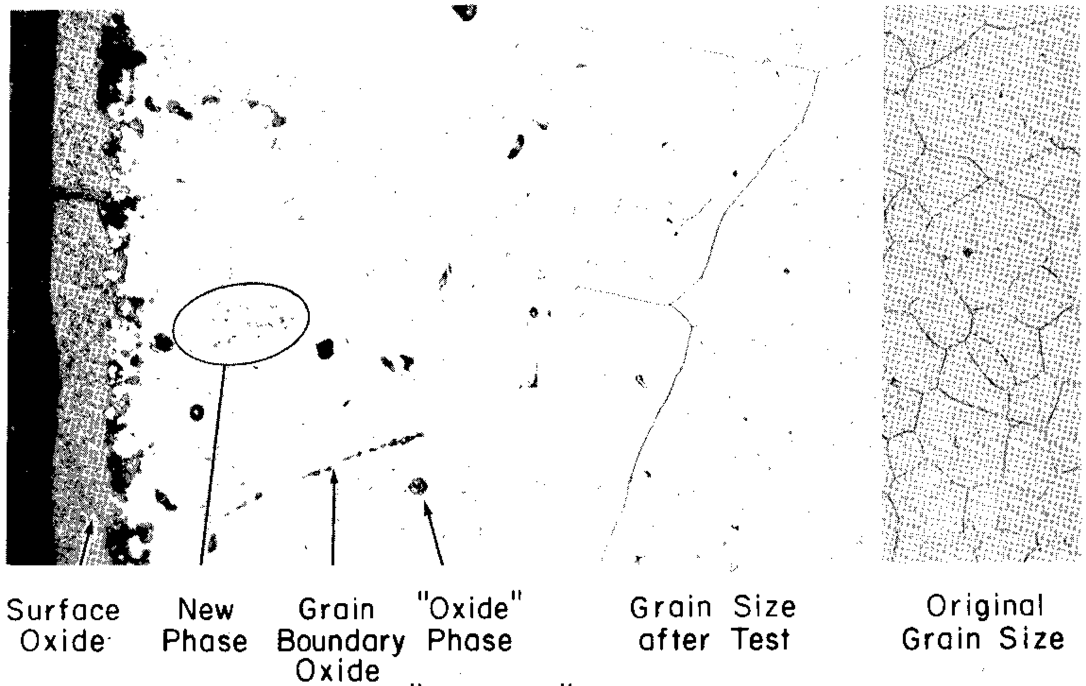
	Thickness affected, mils		
	Surface Scale	Intergranular Oxidation	Zone Affected by Oxidation[a]
TD Nickel Chromium	2.0	None	1.6
"Inconel" 600	1.6	7.1	10.0
"Hastelloy" C	2.8	2.8	6.0
"Hastelloy" X	1.2	3.4	10.0
"Incoloy"* 825	3.6	8.8	8.8
"Inconel" 625	>10.0	5.0	10.0
"Incoloy" 800	>10.0	5.0	Complete[b]
TD Nickel	24.4	None	Complete[b]
Nickel 270	30.4	Complete[b]	Complete[b]

(a) Zone in which either new phases were formed or pre-existing phases disappeared.
(b) Extended throughout the 60-mil thickness of the sample (both surfaces exposed).
* Trademark of International Nickel

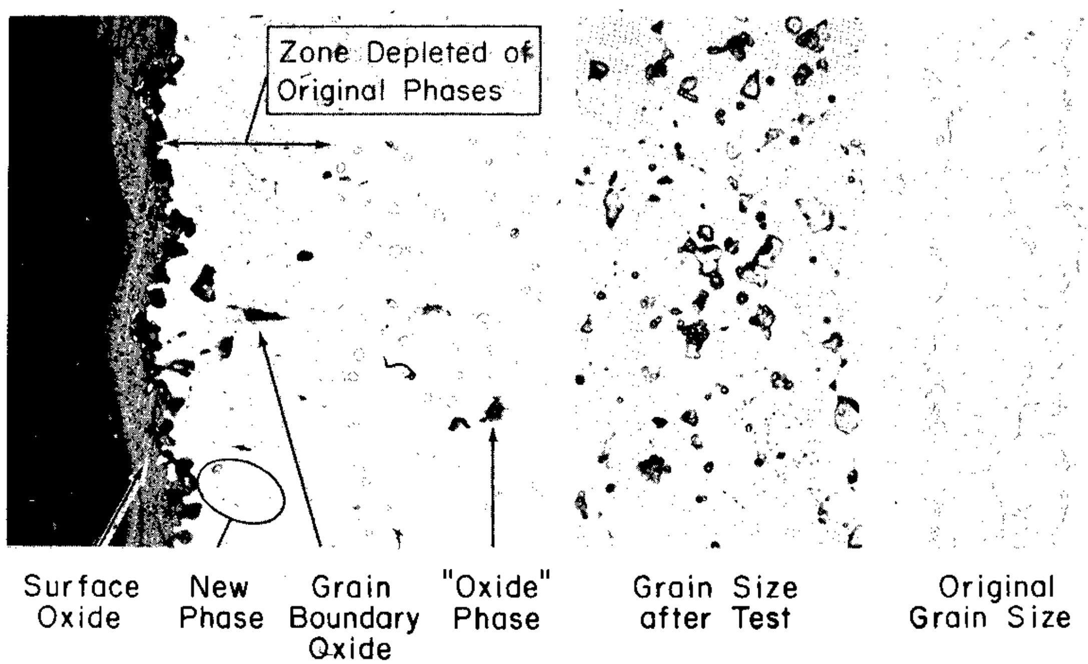
TABLE III

Compositions of Cladding Material in Oxidation Tests

		Nominal Composition, wt %						
	Source	Mo	Cr	Fe	Ni	C	Mn	Other
TD NiC	Du Pont	-	20.0	-	78	0.05	-	ThO_2-2.0
Inconel 600	International Nickel	-	15.8	7.20	76.0	0.04	0.20	
Hastelloy C	Union Carbide	16	15.5	5.0	54	0.08	-	W-4.0, Co-2.5
Hastelloy X	Union Carbide	9	22	18	47	0.10	-	W-0.6, Co-1.5
Incoloy 825	International Nickel	3.0	21.5	30.0	41.8	0.03	0.65	Cu-1.80, Al-0.15, Ti-0.90
Inconel 625	International Nickel	9.0	22.0	3.00	61.0	0.05	0.15	Cb-4.0
Incoloy 800	International Nickel	-	20.5	46.0	32.0	0.04	0.75	Al-0.30, Ti-0.30
TD Ni	Du Pont	-	-	-	98	0.01	-	Co-0.03, ThO_2-2.2
Nickel 270	International Nickel	-	-	-	99.98	0.01	-	



"Inconel" 600



"Hastelloy" X

FIG. 3 OXIDATION OF ENCAPSULATING ALLOYS DURING LONG-TERM HEATING TESTS, 200X
5000 hr in air at 1000°C

- Formation of small particles of unidentified phase

- Disappearance of original phases

The first two reactions may be associated with infusion of oxygen; the third probably occurs by dissolution and diffusion of a constituent from the interior to the surface which acts as a sink. These three reactions may not impair the service of the alloys because their effects on other properties such as strength are expected to be small. Adverse effects on service life may result from the grain growth that results from the prolonged heating, particularly in "Inconel" 600, because less tortuous paths are provided for intergranular oxidation.

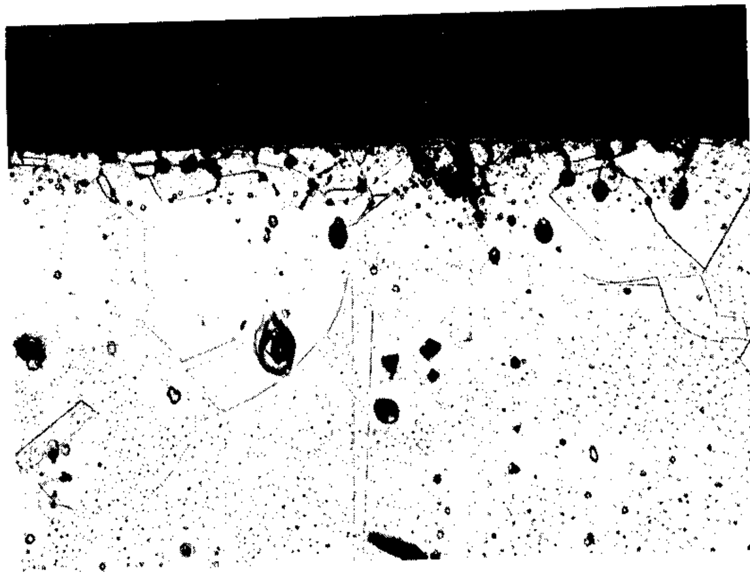
Exposure of companion samples of both groups to 10,000 hr is continuing. The temperature dependency of the oxidation rate will be measured in the coming quarter by heating samples of selected alloys for 500 hr at 850, 1000, and 1150°C.

Capsule Fabrication and Testing

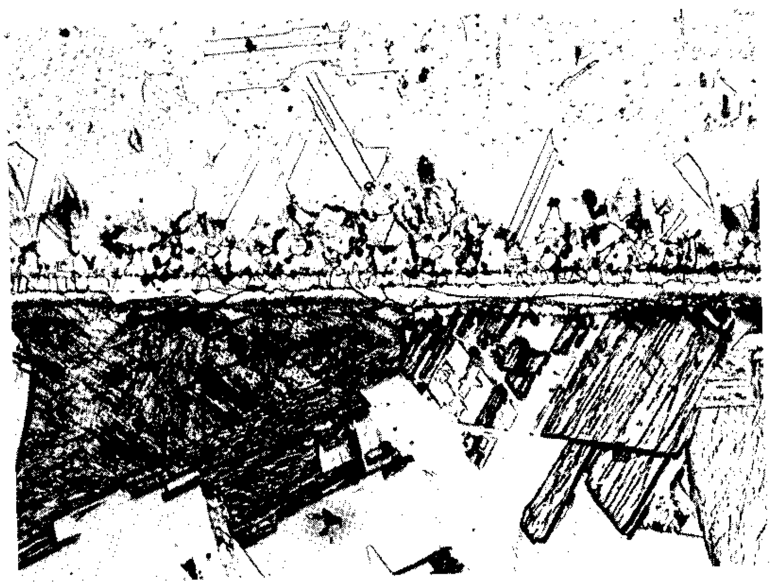
Heating Tests of Inactive Capsules

Reliability and durability of experimental capsules are being evaluated by heating at 850 and 1000°C for up to 10,000 hr in still air. Complete integrity and acceptable oxidation resistance and compatibility have been demonstrated with several "Inconel" 600 and "Hastelloy" C capsules heated for 1000 hr at these temperatures.[1,4] The integrity and compatibility of a TD Nickel capsule was also shown in a 1000-hr test, but oxidation was too severe for TD Nickel to be considered for applications involving oxidizing atmospheres at temperatures above 850°C.

An "Inconel" 600 capsule maintained its integrity and exhibited excellent oxidation resistance and compatibility with the unirradiated cobalt during a 5000-hr test at 850°C. The diameter of the capsule increased about 0.003 inch (0.2%), but no changes were measured in the length or weight of the capsule. The width of the visible diffusion zone between the capsule and the cobalt was 0.004 to 0.005 inch, as compared with 0.001 to 0.002 inch predicted by short-term compatibility tests. The thickness of the oxide scale and the depth of intergranular penetration of the oxide totaled 0.002 inch, as expected from a parabolic extrapolation of the oxidation observed on a similar capsule heated 1000 hr at 850°C. Typical microstructures of the capsule are shown in Figure 4.



a. Oxidation of Exterior Surface (250X)



b. Diffusion Zone (100X)

FIG. 4 OXIDATION AND COMPATIBILITY IN "INCONEL" 600 CAPSULE AFTER 5000 hr AT 850°C

The integrity of a "Hastelloy" X capsule was also maintained during heating for 1000 hr at 1000°C. No significant dimensional changes were observed. Metallographic examinations showed that the degree of oxidation and the width of the solid-solution zone formed by diffusion with the cobalt were in accord with previous short-term measurements of oxidation and compatibility.

Tests of another "Inconel" 600 capsule to 10,000 hr at 850°C and of other "Inconel" 600 and "Hastelloy" X capsules to 5000 hr at 1000°C are in progress. Capsules of other materials are being fabricated for similar testing.

Heating of ^{60}Co Capsules

Experimental capsules containing irradiated cobalt are being heated in air between 850 and 1000°C for up to 10,000 hr to evaluate encapsulation procedures and to measure the effects of radiation on oxidation and compatibility. One capsule each of "Hastelloy" C and "Inconel" 600 was intact and showed no measurable effects of the radiation field during heating for 100 hr at 850°C. The "Inconel" 600 capsule experienced an inadvertent excursion to above 1100°C and swelled a maximum of 0.020 inch.[4]

Two unsuccessful attempts, using inactive capsules, were made to duplicate the swelling that was observed in the radioactive "Inconel" 600 capsule. No swelling occurred; instead, the diameters of the capsules decreased during heating through a temperature cycle that simulated that of the radioactive capsule and also during heating for 2-hr periods at several temperatures up to 1350°C. The diameter decreases resulted from spalling of the oxide at temperatures above 1200°C. These results suggest that the pressure inside the radioactive capsule was higher than normally expected.

An "Inconel" 600 capsule containing alternate layers of radioactive cobalt (totaling about 5000 Ci) and wafers of nickel, tungsten, rhenium, TD Nickel, and TD Nickel-Chromium was intact after heating for 1000 hr at 850°C. Wafers of the latter materials were included to measure the effects of radiation on compatibility. Helium leak tests before and after the heating showed that the capsule was sound. No significant dimensional changes were measured. Metallographic examination is in progress.

Heating tests of two additional "Inconel" 600 capsules for 5000 and 10,000 hr at 850°C is continuing. Another "Inconel" 600 capsule containing about 7000 Ci of ^{60}Co was fabricated and heating at 850°C for 10,000 hr, or more, was begun.

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