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AEC RESEARCH AND DEVELOPMENT REPORT

SAVANNAH RIVER LABORATORY ISOTOPIC POWER AND HEAT SOURCES

QUARTERLY PROGRESS REPORT

JULY - SEPTEMBER 1967

PART I - COBALT-60

DU PONT
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H. S. Hilborn, Compiler

October 1967

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AIKEN, S. C. 29801

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PREFACE

This report is one 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 July, August, and September performed during the report period. Previous reports are listed on the last page.

SUMMARY

Estimated production costs for large quantities of ^{60}Co range from about 10¢/Ci at 100 Ci/g to 20¢/Ci at 400 Ci/g. (pg 1)

Diffusion of ^{60}Co along grain boundaries may result in concentrations greater than 10^{-6} of the interface concentration at a depth of 0.060 inch in TD Nickel but <0.020 inch in several other alloys after one year at 800°C. (pg 4)

Preliminary examination of the "Inconel" 600 capsule containing radioactive cobalt that was heated in air at ~850°C for 1000 hr indicates that this capsule will be suitable for long-term service. (pg 7)

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. The initial objective is to establish allowable operating limits for capsules of radioactive cobalt metal contained in oxidation resistant alloys. Development of specific heat source concepts is not at present included in the scope of the SRL program.

COSTS FOR LARGE-SCALE PRODUCTION OF ^{60}Co

Estimated production costs for large quantities of ^{60}Co are shown in Figure 1 as a function of the time available for irradiation. Minimum costs range from about 10¢/Ci at 100 Ci/g to about 20¢/Ci at 400 Ci/g, and do not change rapidly with irradiation time if at least 1 to 1½ years can be allowed. They increase sharply for specific activities above about 450 Ci/g due to burnup and decay of ^{60}Co in the reactor.

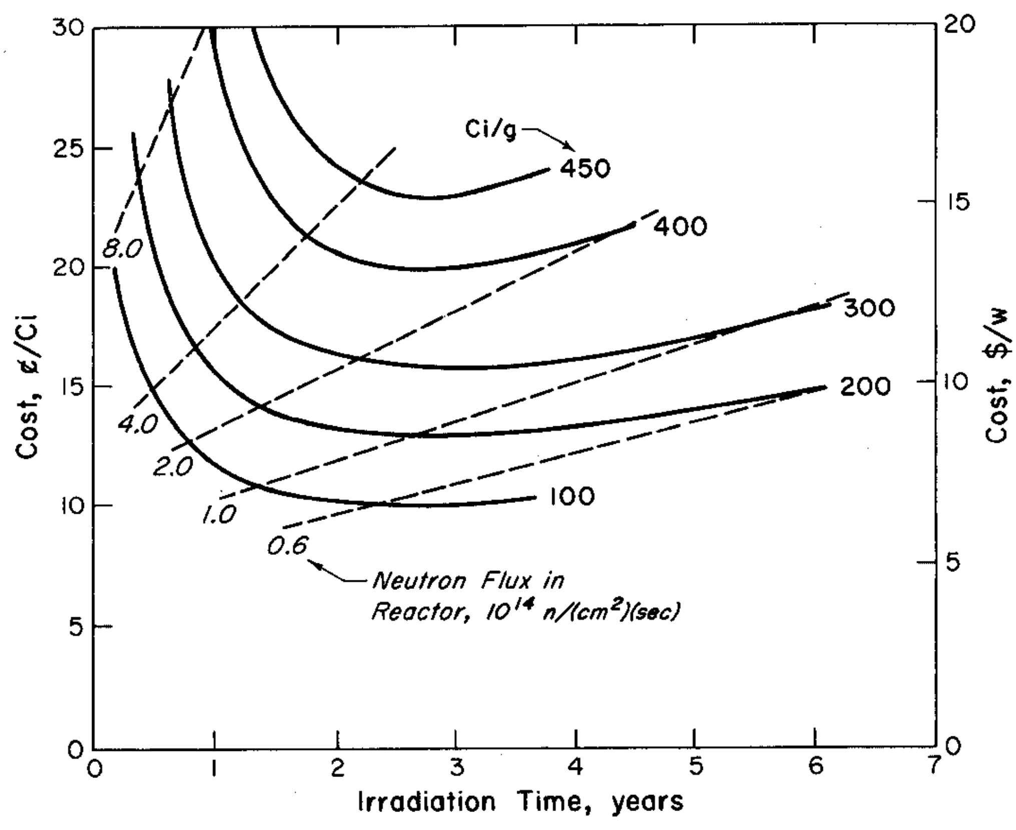


FIG. 1 ^{60}Co PRODUCTION COST ESTIMATES
(For Large Amounts)

These costs include estimates of all direct expenditures and related costs of plant operation, including appropriate allocation of fixed operating costs and costs of components and fuel burnup. These estimated costs do not include factors related to capital, such as depreciation and amortization charges, nor miscellaneous AEC factors. Encapsulation also is not included, and is expected to add about 2¢/Ci.

The curves are based on one reactor producing ^{60}Co as its major product. Quantities of ^{60}Co less than about 10 megacuries per year would be considered incidental production, and would be made in reactor charges designed for other products. Irradiation time would depend on the neutron flux in those charges, but ^{60}Co costs would not be substantially higher than shown on the curves. For quantities much larger than 10 MCi/yr, design of reactor charges would be influenced by the need to minimize irradiation time for the ^{60}Co product.

A listing of the shapes and activities of cobalt metal irradiated to date for experimental programs is given in Reference 1.

MATERIALS TECHNOLOGY AND DEVELOPMENT

High Temperature Fuel Candidates

Development of alloys and compounds of cobalt that have higher melting temperatures than cobalt metal have been deferred.

Evaluation of Encapsulating Materials for Cobalt Metal

The materials evaluation program is designed to select the most promising alloys for encapsulating ^{60}Co, to define the limiting operating conditions of these alloys, and to demonstrate capsule integrity at conditions typical of heat source operation. The kinetics of compatibility reactions,[1,2] diffusion of ^{60}Co,[1] oxidation,[1,3] and creep are being measured using short-term (<500 hr) laboratory tests. Selection of the most promising alloys is based on extrapolation of these results to the expected service life (1 to 5 yr). Limiting operating conditions are defined by the time and temperature dependency of each of the reactions. The predicted behavior of the materials is being verified by long-term (10,000-hr or more) heating tests of experimental capsules, Table I.[1,3] Tests of companion capsules of unirradiated and irradiated cobalt will measure any effects of the radiation field and the increased nickel content (from radioactive decay of the cobalt) on the performance of the capsule materials.

TABLE I

SUMMARY OF COBALT CAPSULE HEATING TESTS

Capsule Material	Heating Time, hr	Heating Temp, °C	Wall, mils	Activity Specific Ci/g	Activity Total, Ci	Approximate Completion Date	Remarks
A. Inactive Capsules							
"Inconel" 600	1,000	850	50	---	---	2-67	Capsule intact
	5,000	850	50	---	---	7-67	Capsule intact
	10,000	850	50	---	---	1-68	
	10,000	850	95	---	---	9-68	
	1,000	1000	50	---	---	10-66	3 capsules intact; 1 capsule oxidized(b)
	5,000(a)	1000	50	---	---	11-67	
	1,000	1000	95	---	---	9-67	No severe oxidation of Co
	5,000	1000	95	---	---	2-68	
	10,000	1000	95	---	---	10-68	
"Hastellov" C	1,000	1000	50	---	---	10-66	3 capsules intact; 1 capsule oxidized(b)
"Incoloy" 825	1,000	1000	50	---	---	8-66	1 capsule intact; 1 capsule oxidized(b)
TD Nickel	1,000	1000	50	---	---	2-67	Capsule intact
"Hastelloy" X	1,000	1000	50	---	---	6-67	Capsule intact
	5,000	1000	50	---	---	11-67	
B. ^{60}Co Capsules							
"Inconel" 600	130	850(c)	50	120	16,000	2-67	Swelled due to over-heating
	1,000	850	50	100(d)	5,000(d)	7-67	Capsule intact
	5,000	850	50	150(d)	15,000(d)	11-67	
	10,000	850	50	150(d)	15,000(d)	6-68	
	10,000	850	50	150(d)	9,000(d)	7-68	
"Hastelloy" C	100	850	50	120	9,000	1-67	Capsule intact

(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) Excursion to >1100°C for 3-6 hr.
(d) Activity as of 2-67.

Diffusion of ^{60}Co

Diffusion of ^{60}Co may be one of the limiting factors in designing capsules for uses in which the capsule wall is exposed to a flowing coolant. Two significant aspects of diffusion have been demonstrated by analyses of earlier tests:[1]

- An acceptable ^{60}Co concentration on the outer capsule surface is about 10^{-6} of that at the cobalt-capsule interface.
- This concentration might be reached by diffusion along grain boundaries but probably not by diffusion through the volume of the grains.

Recent analyses indicate that diffusion will not be limiting in "Inconel"* 600, "Hastelloy"** C, "Hastelloy"** X, or "Haynes"** 25 capsules operating for one year at 800°C. In contrast, diffusion may be limiting in TD Nickel*** capsules because of the greater depth of penetration by grain boundary diffusion.

Coefficients of grain boundary and volume diffusion were calculated from the concentration profiles obtained by standard radiotracer techniques on polycrystalline samples of several candidate capsule materials that were heated for 100 hr at 800°C, as shown in Table II. These calculations are based on two theoretical models developed for bicrystalline samples,[4] but are applied to polycrystalline candidate alloys to provide estimates of the contributions of volume and grain boundary diffusion. Coefficients of volume diffusion obtained with these models on control samples of pure nickel and cobalt were higher than those reported in the literature by factors of 4 and 20, respectively. Differences in initial grain size, depth of penetration measurements, and analysis technique, as well as effects of changes in grain size during heating, are being analyzed to explain these discrepancies.

The diffusion coefficients calculated from the 100-hr tests were used to predict the concentration profiles that would occur in each of the materials after one year at 800°C. These calculations showed that only in TD Nickel would the allowable ^{60}Co concentration be reached at depths greater than 0.020 inch, as shown in Table II and in Figure 2.

* Trademark of International Nickel Co., New York, N. Y.
** Trademark of Union Carbide Corp., New York, N. Y.
*** Product of E. I. du Pont de Nemours and Co., Wilmington, Del.

TABLE II

^{60}Co Diffusion in Capsule Materials at 800°C
(Instantaneous Source Model)[4]

Material	Volume Diffusion Coefficient, 10^{-13} cm²/sec	Grain Boundary Diffusion Coefficient, 10^{-8} cm²/sec	Depth after 1 yr for 10^{-6} Initial ^{60}Co Conc., mils
"Hastelloy" X	0.68	0.40	8
"Hastelloy" C	0.87	1.15	11
"Haynes" 25	0.53	1.14	13
"Inconel" 600	1.26	3.45	20
TD Nickel	4.00	69.2	60

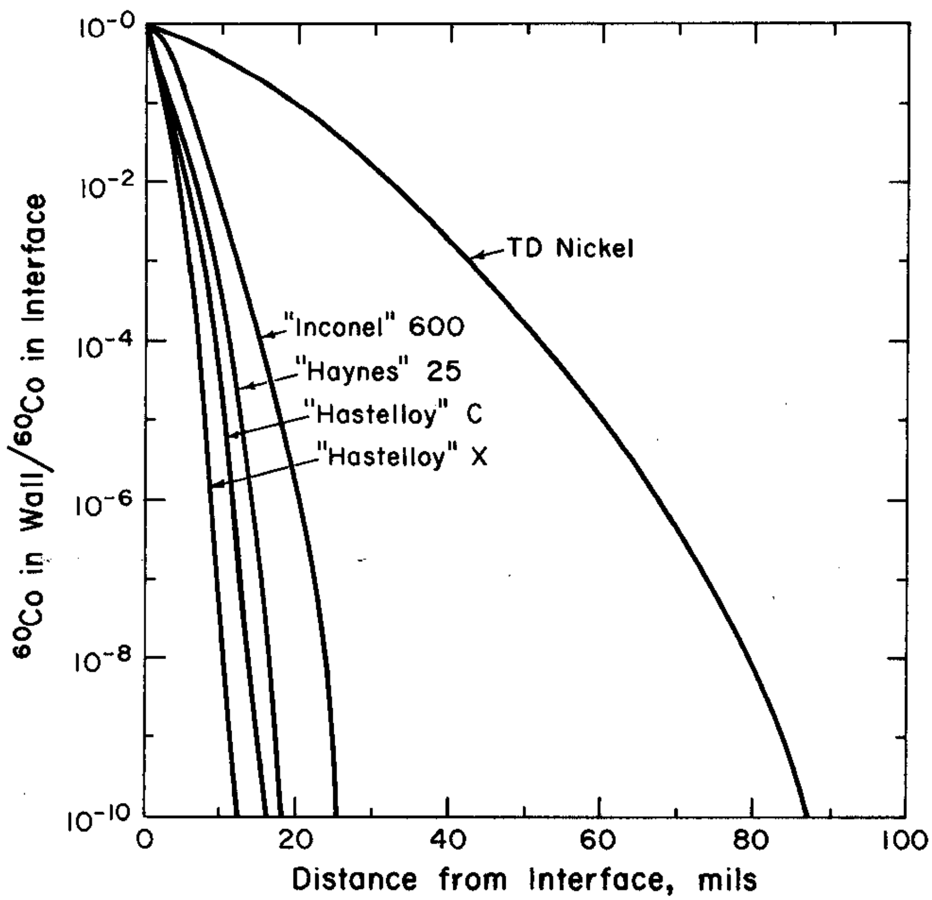


FIG. 2 PREDICTED DIFFUSION OF ^{60}Co INTO CAPSULE MATERIALS AFTER ONE YEAR AT 800°C. Instantaneous Source Model[4].
Based on Measurements After 100 Hours

Three computer codes have been developed for use in diffusion analysis. CODD (Calculation Of Diffusion Data) integrates the theoretical concentration contours due to grain boundary diffusion and is used as a subroutine in the other programs. This code duplicates one reported in the literature,[5] but was needed because the diffusion times and distances that are typical of heat source applications are greater than those covered by calculations in the literature. The values produced by this program have been compared with the published values. ADD (Analysis of Diffusion Data) fits either of the two theoretical diffusion models to measured diffusion concentration profiles to calculate diffusion parameters. PREDICT (Prediction of Diffusion Profiles) accepts known diffusion parameters and calculates the diffusion profile that would occur after a specified time interval based on theoretical models.

These codes will be used to analyze 100-hr diffusion anneals at 1000°C, and to calculate wall thickness requirements for various capsule operating times. The extrapolations will then be compared with profiles measured in long-term capsule tests to determine the validity of the theoretical extrapolation.

Rate of Oxidation

Exposure of nine candidate alloys for 9400 hr at 1000°C has been completed and metallographic examination is in progress. Examinations of duplicate samples after 1000 and 5000 hr are described in References 1 and 3, respectively.

Capsule Fabrication and Testing

Heating Tests of Capsules Containing Unirradiated Cobalt

Several capsules remained sound and demonstrated acceptable oxidation resistance and compatibility during heating for either 1000 or 5000 hr at 850°C and for 1000 hr at 1000°C.[1,3]

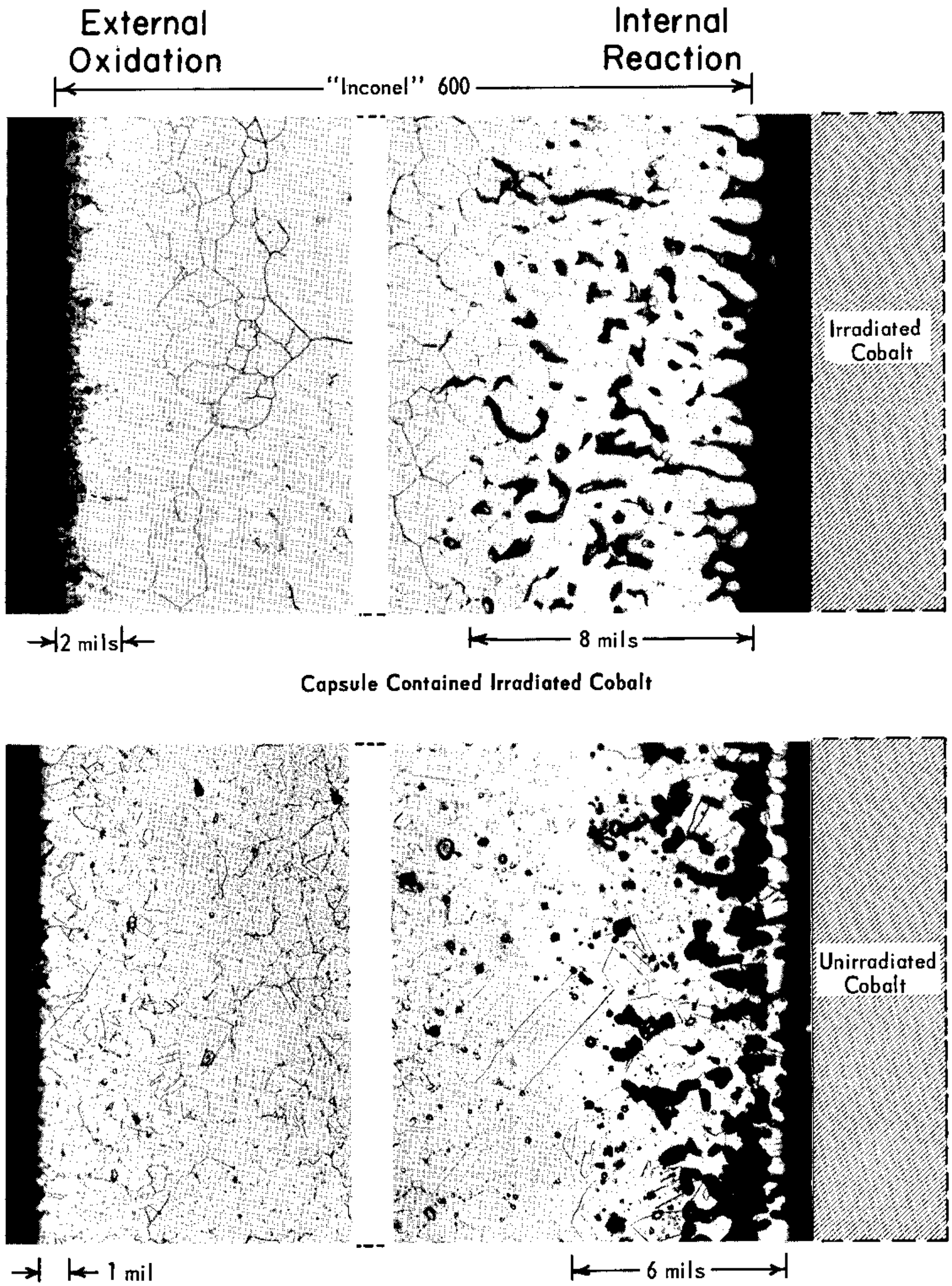
Recent tests showed little effect of simulated capsule defects on the performance of two "Inconel" 600 capsules that were heated for 1000 hr at 1000°C. One capsule had its cap pressed in but not welded; the other was welded but had an 0.008-inch diameter hole drilled through the capsule wall. Examination of both capsules showed essentially no oxidation of the cobalt inside the capsules or of the internal surfaces of the capsules. The drilled hole and the gap between the cap and the capsule were almost completely filled with products of the oxidation of the "Inconel" 600. Microprobe analyses of the oxide will be made to detect any potential release of cobalt.

Heating Tests of Capsules Containing Irradiated Cobalt

Capsules of irradiated cobalt in "Hastelloy" C and "Inconel" 600 were sound after 100 hr at 850°C and showed no apparent effect of the ^{60}Co radiation.[3] Another "Inconel" 600 capsule remained stable after 1000 hr at ~850°C.[1] Recently, metallographic examinations were completed of the surface and contents of the capsule heated 1000 hr at 850°C. This capsule would be satisfactory for long-term service, although slightly more oxidation and interaction reaction were observed in the active capsule than in a companion inactive capsule.

The "Inconel" 600 capsule heated for 1000 hr at 850°C contained about 5000 Ci of cobalt wafers. It also contained wafers of tungsten, rhenium, nickel, TD Nickel and TD Nickel Chromium that were arranged to determine the compatibility of these encapsulating materials with ^{60}Co in an oxygen-free environment. When the capsule was sectioned for metallographic examination, some of the wafers fell out, indicating lack of bonding to the cobalt. In cases where reaction zones occur, they are slightly thicker than those measured in diffusion couples with unirradiated cobalt.

External oxidation of "Inconel" 600 was similar for the active and inactive capsules, Figure 3. The oxide scale and the depth of intergranular oxidation total 0.001 to 0.002 inch on the active capsule and ~0.001 inch on the inactive capsule. The "Inconel" has not formed a complete metallurgical bond with the cobalt, but a porous zone is evident. This porous reaction zone is similar to the normal zone of Kirkendall voids in bonded diffusion couples. It is slightly thicker in the active capsule than in the inactive capsule; 0.008 and 0.006 inch, respectively. These zones are being examined further.



Capsule Contained Unirradiated Cobalt

FIG. 3 SURFACE REACTIONS OF "INCONEL" 600 CAPSULES HEATED 1000 HR AT 850°C

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Previous Quarterly Progress Reports issued include:

DP-1088
DP-1094
DP-1105-I and -1105-II
DP-1120-I and -1120-II