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RADIOACTIVE COBALT FOR HEAT SOURCES

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October 1965

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ABSTRACT

Large quantities of radioactive cobalt with a high specific activity (curies of cobalt-60 per gram of product) can be produced at reasonable cost in the high neutron fluxes attained in a Savannah River reactor. This high-activity cobalt is suitable for use in multi-kilowatt heat sources, and a program is in progress to develop the required data for this application. The penetrating radiation of cobalt-60 is converted to usable heat with good efficiency in compact, high-strength sources, and in such sources the ratio of shielding weight to contained activity is kept small. Pertinent data on natural and radioactive cobalt are presented as background information.

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RADIOACTIVE COBALT FOR HEAT SOURCES

INTRODUCTION

Recent operation of a Savannah River Plant reactor at high neutron fluxes has established the feasibility of producing large amounts of cobalt-60 at high specific activity (up to 700 curies of cobalt-60 per gram of initial cobalt). This material is well suited for use in relatively large heat sources to generate electricity, useful heat, or propulsion force for aerospace, terrestrial, or marine applications. Cobalt-60 can be provided in larger quantities than other radioisotopes, and at relatively low cost.

Because of the advantages of cobalt-60 for such applications, the Savannah River Laboratory has undertaken a program to develop the technology required to design, fabricate, and operate cobalt-60 heat sources capable of producing up to 1000 thermal kilowatts.

More than four million curies (4 megacuries) of cobalt-60 have been produced in Savannah River Plant reactors during the past ten years for use in gamma radiation sources for food preservation, teletherapy, radiography, and other radiation applications. Most of this material has been produced with a specific activity of less than 30 curies per gram.

This report summarizes the available information on radioactive cobalt and on natural cobalt at the start of the development program. Most of the data on cobalt properties were obtained from the literature, but production information is from the Savannah River Plant. Important properties in the design of heat sources are emphasized and areas of incomplete knowledge are identified.

The more significant areas that require further information include:

- High temperature properties, particularly of the radioactive cobalt itself, and also of the essential materials of construction such as encapsulating material, heat removal systems, and shielding.
- Effects of radioactive decay and of irradiation upon the required system.
- Requirements and means of achievement for complete, total containment of the radioactive cobalt during all stages of use and subsequent handling (encapsulating materials and techniques, for example).

- Over-all safety to provide adequate assurance against accidents, including radiation accidents or activity releases, in the transportation and use of large heat sources fueled with cobalt-60. This factor has many aspects, and will require wide and continuing attention in all phases of the program to develop and utilize cobalt-60 heat sources.

SUMMARY

Cobalt-60 can be produced in sufficient quantity to satisfy most foreseeable needs for relatively large-scale utilization of heat sources. The Savannah River Plant can produce hundreds of megacuries of cobalt-60 per year if reactor space is available. Expressed in terms of initial thermal output of the cobalt, one megacurie is equivalent to 15.6 kw. Deliveries of 30 curies/gram cobalt can usually be made within a year after an order is placed. Production of high-activity cobalt may take longer unless suitable high-flux reactor charges are available.

At the present time, the Atomic Energy Commission has established the minimum price for cobalt of less than 30 curies/gram at 50¢/curie ($32/watt) for sales to customers outside the Federal Government and less than 33¢/curie for transfers to other Government agencies. These transfer prices have ranged between 25¢/curie and less than 10¢/curie for cobalt with an activity up to 120 curies/gram, depending on the amount of cobalt ordered, the desired specific activity, the required delivery dates, and the current operating schedule for the reactors. At 10¢/curie the cost of cobalt-60 is $6.40/watt or $1700/kilowatt-year assuming a useful life of one isotopic half-life. The average power over one half-life is 72% of the initial power. The cost per kilowatt-year can be reduced if the cobalt-60 can be resold for irradiation sources after it becomes too weak for heat sources.

Radioactive cobalt is a mixture of cobalt-59, cobalt-60, nickel-60, and nickel-61. Cobalt-59, the naturally occurring isotope, is irradiated in a nuclear reactor to produce cobalt-60, the radioactive isotope with a half-life of 5.27 years. The cobalt-60 decays by emitting beta particles and gamma rays to form nickel-60. Nickel-61 is formed from the neutron bombardment of nickel-60 and cobalt-60 (with a subsequent beta decay).

The activity and heat generation of radioactive cobalt is proportional to the concentration of cobalt-60 in the product. This concentration can be matched to the requirements of a specific application by selecting the desired neutron flux and exposure time for irradiation. The current practical limit is about 60% cobalt-60, which corresponds to a specific activity of 700 curies/gram and a thermal power of 10.9 watts/gram.

Natural cobalt has physical and chemical properties intermediate between those of iron and nickel, but very little information is available for radioactive cobalt. Available information and analogies with the effects of irradiation on iron and nickel indicate that values for the physical properties of unirradiated cobalt can be used in the preliminary design of heat sources. The major uncertainty is the influence of irradiation on corrosion behavior.

Relatively thick shielding is required to reduce the penetrating gamma radiation from radioactive cobalt sources. The weight of such shielding can be reduced by using cobalt with a high specific activity, to reduce the size of the source. Because of its relatively low metabolic retention, the biological hazard for ingestion of cobalt-60 is appreciably less than that for most of the isotopes that are considered for heat source applications.

DISCUSSION

PRODUCTION OF RADIOACTIVE COBALT

Production and Decay Reactions

Cobalt-60 is produced by irradiating cobalt-59 in a nuclear reactor. The production process is shown schematically in Figure 1[1].

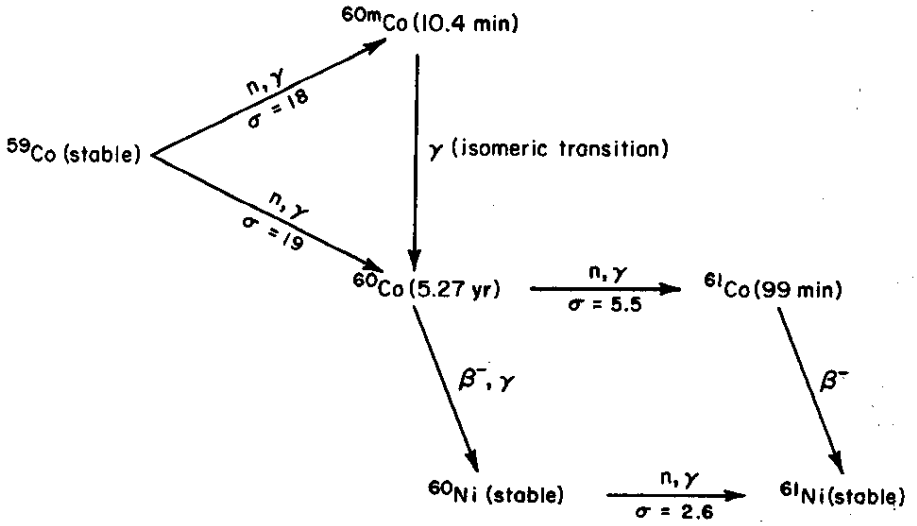
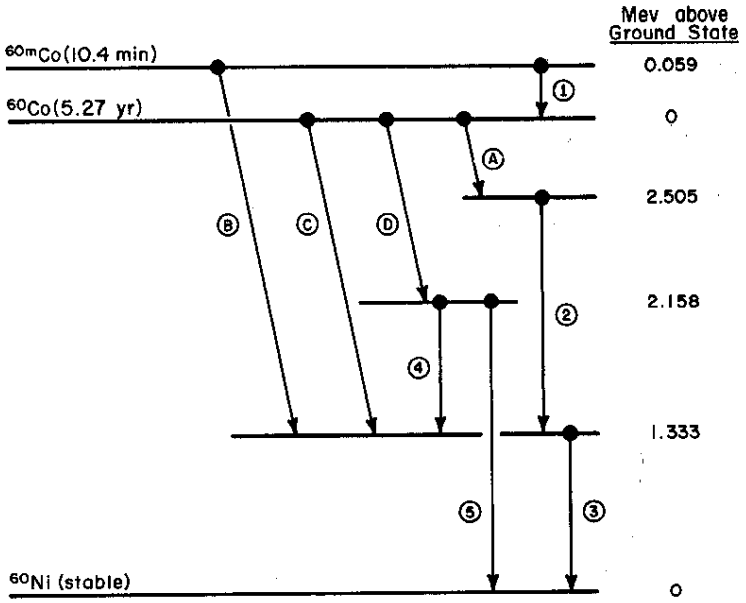


FIG. 1 PRODUCTION PROCESS FOR COBALT-60[1]

Two isomers of cobalt-60 are formed by n,γ reactions but more than 99% of the short-lived cobalt-60m undergoes an isomeric transition to cobalt-60. Little error is introduced by neglecting the formation of cobalt-60m and assuming that capture of a neutron by an atom of cobalt-59 produces an atom of cobalt-60 and a 7-Mev gamma ray.

$$^{59}Co + n \rightarrow {}^{60}Co + \gamma$$

Cobalt-60 decays to stable nickel-60 by the emission of beta particles with a maximum energy of 0.312 Mev and gamma rays with energies of 1.172 and 1.333 Mev. Small amounts of other radiations are also emitted, as illustrated in Figure 2[2,3], but these emissions have negligible influences on shielding requirements or heat generation. Properties of radioactive cobalt are summarized in Table I.



Beta Particles	Max. Energy, Mev	Abundance, %
A	0.312	99+
B	1.480	0.15
C	1.478	0.01
D	0.63	0.004
Gamma Rays	**Energy, Mev**	**Abundance, %**
1	0.059	99+ (isomeric transition)
2	1.172	99+
3	1.333	99+
4	0.825	<0.003
5	2.158	0.001

FIG. 2 DECAY OF COBALT-60[2, 3]

TABLE I

Properties of Radioactive Cobalt

Cobalt form	Metal
Shapes	Pellets, wafers, rods, slabs
Half-life, years	5.27
Activity, Mev	
β (max)	0.312
γ	1.172
	1.333

Radioactive cobalt can be produced with a specific activity and power to match its intended application.

	Typical Cases				Pure ^{60}Co
curies/g	100	200	400	700	1140
watts/g	1.6	3.1	6.2	11	17.7
watts/cm^3	14	27	55	96	156

Activity - power conversion

1 megacurie = 15.6 kw

1 curie = 0.0156 watt

1 watt = 64.2 curies

1 kw = 0.0642 megacurie

Some of the cobalt-60 atoms in the reactor capture neutrons to form cobalt-61 atoms. The cobalt-61 decays rapidly to stable nickel-61 by the emission of beta particles with a maximum energy of 1.22 Mev and gamma rays with an energy of 0.072 Mev. Because of its 99-minute half-life, essentially all of the cobalt-61 is converted to nickel-61 within a day after the irradiation is completed. The neutron absorption cross section of cobalt-60 is much less than the cross section of cobalt-59, so the amount of cobalt-61 and nickel-61 formed during irradiation is small except for extended irradiations or irradiations at extremely high flux.

The net formation of cobalt-60 during irradiation is described by the equation:[4]

$$\frac{N_{60}}{N_{59}} = \frac{\alpha e^{-\alpha t}}{\beta} (1 - e^{-\beta t})$$

where N_{60} = number of atoms of cobalt-60 at time t

N_{59} = initial number of atoms of cobalt-59

$\alpha = \phi\sigma_{59}$

ϕ = neutron flux in cobalt, $n/(cm^2)(sec)$

σ_{59} = absorption cross section of cobalt-59, 36.3×10^{-24} cm^2/atom

$\beta = \lambda_{60} + \phi\sigma_{60} - \phi\sigma_{59}$

λ_{60} = decay constant of cobalt-60, 4.16×10^{-9} sec^{-1}

σ_{60} = absorption cross section of cobalt-60, 5.5×10^{-24} cm^2/atom

t = time in reactor, seconds

Solutions to this equation, converted to specific activities and powers, are plotted in Figure 3 for several neutron fluxes. These data illustrate the effects of cobalt-60 burnup and decay on limiting the maximum activity that can be achieved by irradiation.

The activity of pure cobalt-60 is 1140 curies/gram but the maximum activity of radioactive cobalt after irradiation will be limited to about 700 curies/g. In theory, higher cobalt specific activities are possible by chemical or isotopic separation after irradiation. If cobalt is irradiated for an extended period at high flux so that almost all of the cobalt-59 is converted to cobalt-60 and nickel, chemical separation of nickel and cobalt could yield almost pure cobalt-60. In practice, the separation would be difficult because of the likelihood of radiolytic damage to the chemical compounds needed for the separation. For cobalt of lower specific activity, isotopic separation might also be used to increase the cobalt-60 content[5].

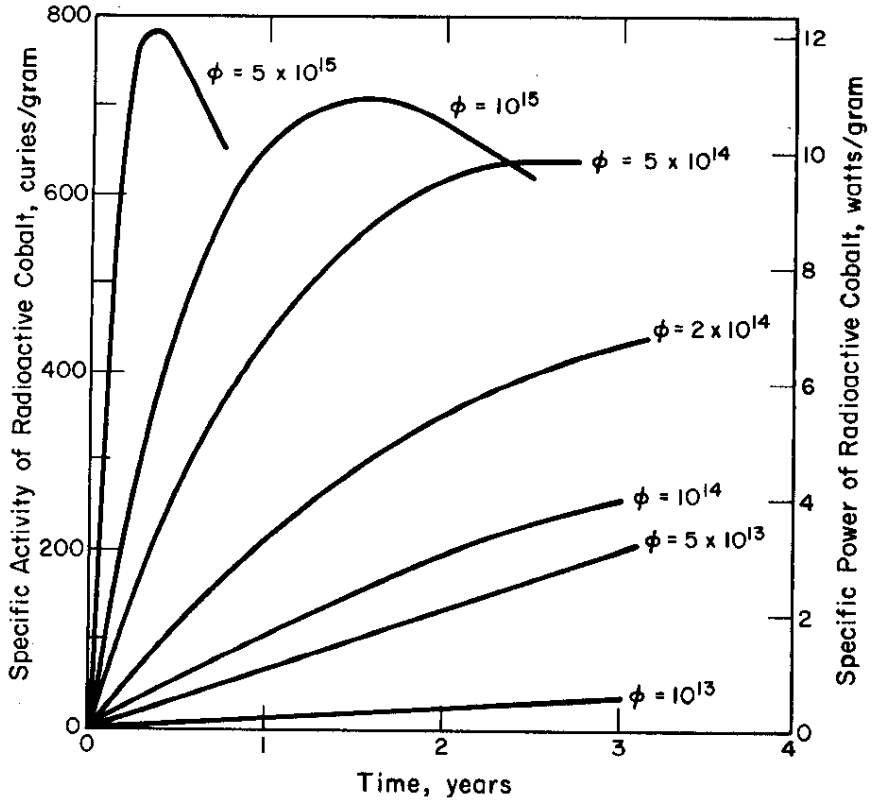


FIG. 3 PRODUCTION OF COBALT-60
ϕ = thermal neutron flux in cobalt, $n/(cm^2)(sec)$

Nickel Content

Radioactive cobalt is an almost pure cobalt-nickel alloy with only trace amounts of impurities. The nickel content increases with time, but because the two metals are very similar the effect on properties is probably very small.

The nickel content at the completion of irradiation is a function of the reactor neutron flux level and the irradiation time. Not all radioactive cobalt of the same specific activity will have the same nickel content.

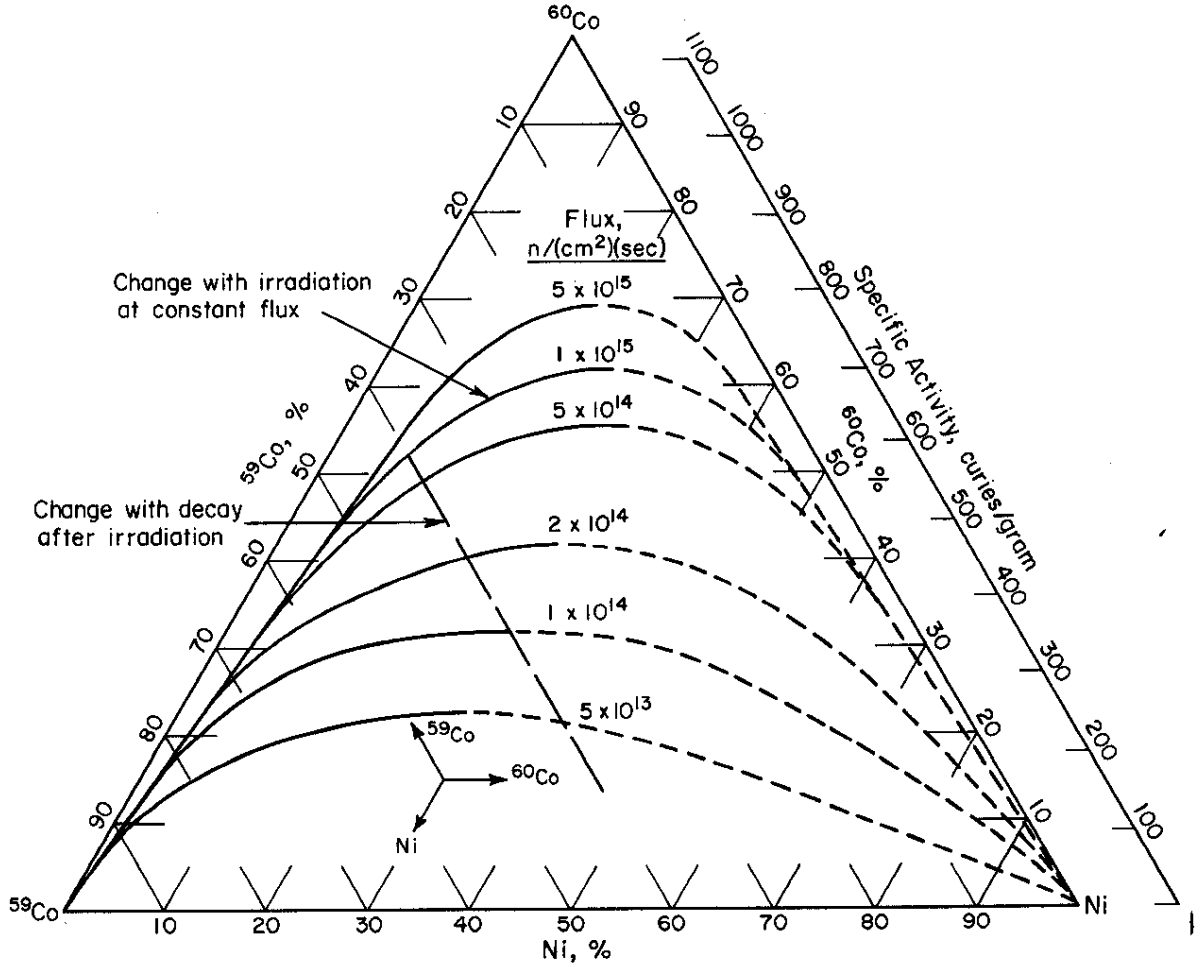


FIG. 4 ISOTOPIC CONTENT OF RADIOACTIVE COBALT

The alloy composition of radioactive cobalt is shown in Figure 4 as a function of irradiation flux. The solid portions of the lines give the composition for practical irradiations (irradiation times less than those required to produce the maximum cobalt-60 content possible at the irradiation flux level). The composition at any time after irradiation is completed lies on a line of constant cobalt-59 content. As an example of determining the composition of radioactive cobalt, material that had been irradiated at a flux of 10^{15} n/(cm^2)(sec) to a specific activity of 600 curies/g would consist of 52.6% ^{60}Co, 39.4% ^{59}Co, and 8.0% Ni. After decaying to 300 curies/g the composition would be: 26.3% ^{60}Co, 39.4% ^{59}Co, 34.3% Ni. Decay curves for radioactive cobalt of several specific activities are included in Figure 5.

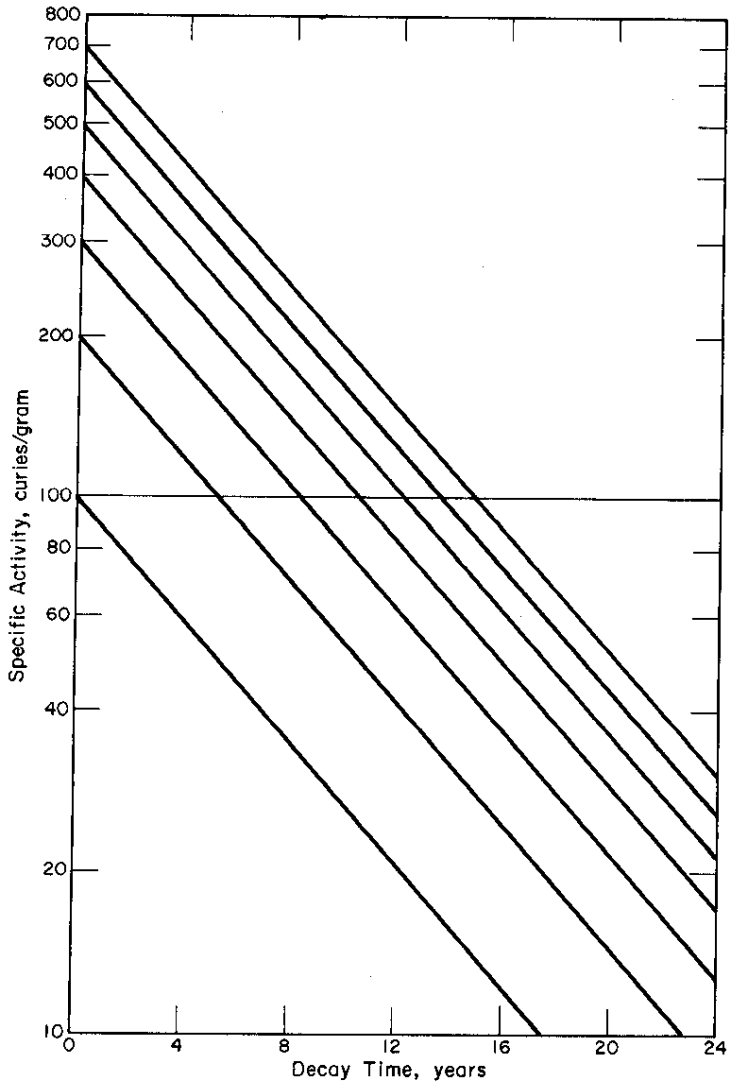


FIG. 5 EFFECT OF COBALT-60 DECAY ON ACTIVITY OF RADIOACTIVE COBALT

The neutron flux in the cobalt is a function of the amount of cobalt in a reactor position, the geometry of the cobalt, and the neutron flux in reactor fuel positions adjacent to the cobalt. In general, high and uniform cobalt activities are best achieved by irradiating cobalt in tubes that have large diameters and thin walls. The large diameter exposes the cobalt to a greater number of neutrons, thus resulting in maximum "blackness"[4] for a given weight of cobalt, and the thin wall minimizes the flux depression in the cobalt caused by self-shielding.[6]

Production at the Savannah River Plant

More than 4.2 megacuries of cobalt-60 have been produced in Savannah River Plant reactors during the last ten years. Cobalt irradiations currently in progress will produce another 5.5 megacuries during the next 18 months. Irradiations proposed for FY-1966 may add as much as 7 megacuries of cobalt-60.

All of the radioactive cobalt produced to date has been incidental to the production of other materials. The maximum diameter of the aluminum cans that contain the cobalt has been 0.94 inch, but it is possible to irradiate targets as large as 3 inches in diameter. Cobalt has been irradiated in a variety of sizes and shapes as shown in Figures 6 through 9.

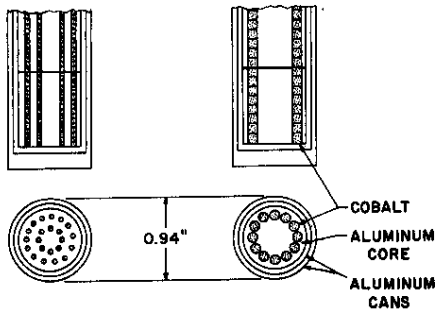
Some of the design criteria for these targets are:

- Materials - Reactor-grade cobalt as described in the next paragraph; type 1245 aluminum and type 304 stainless steel are used for canning.

- Cobalt protection - The cobalt is plated with nickel (0.0004 to 0.0006 inch thick) or canned in stainless steel to reduce cobalt corrosion and possible contamination of facilities during postirradiation handling.

- Canning - The cobalt is usually double-canned with either two aluminum cans or one aluminum can over a stainless steel can. Both cans are welded and tested for leaks. Single aluminum cans are also used.

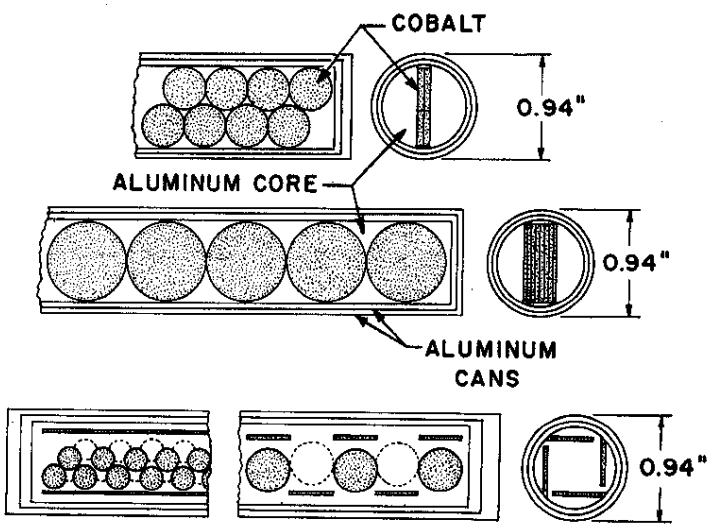
- Density - The amount of cobalt in a target is specified to produce the desired activity in the cobalt after irradiation in a specific location in the reactor for the time required to meet the desired delivery date.



Pellet Sizes

1 mm x 1 mm
1/16 inch x 1/16 inch
1/8 inch x 1/8 inch
1/8 inch x 1/4 inch

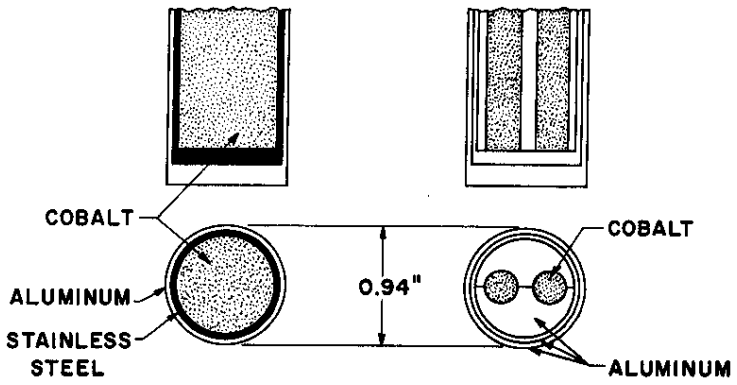
FIG. 6 TARGET DESIGNS FOR COBALT PELLETS



Wafer Sizes

1/2 cm x 1 mm
1 cm x 1 mm
1 cm x 2 mm
2 cm x 1 mm
0.750 inch x 0.036 inch

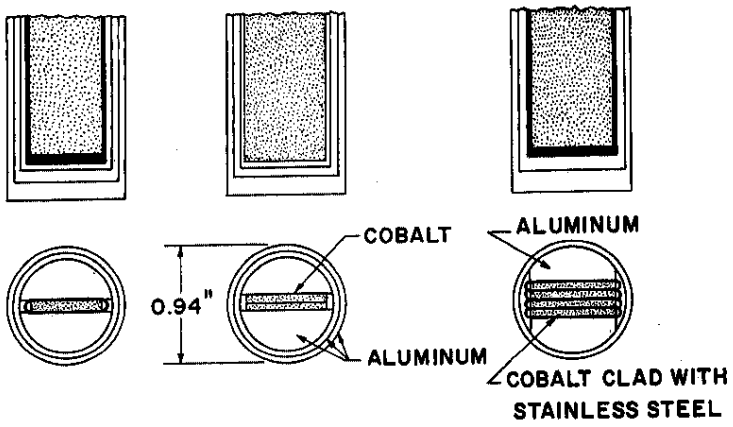
FIG. 7 TARGET DESIGNS FOR COBALT WAFERS



Rod Sizes

0.725 inch x 4.375 inch
0.725 inch x 10.25 inch

FIG. 8 TARGET DESIGNS FOR COBALT RODS



Slab Size, inches		
Width	Thickness	Length
0.745	0.033	9.9
0.745	0.059	9.9
0.745	0.063	9.9
0.745	0.260	9.9
0.760	0.130	10
0.640*	0.060	12
0.640	0.060	11

* This size is used in the BNL standard cobalt-60 source.

FIG. 9 TARGET DESIGNS FOR COBALT SLABS

Reactor-grade cobalt, as received from the vendor, contains at least 99.87% cobalt and nickel, no more than 0.37% nickel, and small amounts of carbon, iron, manganese, and silicon. Analyses of representative cobalt slabs and wafers are included in Table II. The impurities are in solid solution and remain in solid solution during irradiation. Limited data indicate that the cobalt microstructure is not altered by irradiation to an activity of 100 curies/g, as shown in Figure 10.

Deliveries of 30 curies/g cobalt can usually be made within a year after an order is placed if standard shapes are used (an inventory of unirradiated cobalt is maintained). The production of high-activity cobalt may take longer unless suitable high-flux reactor charges are available.

TABLE II

Analysis of Reactor-Grade Cobalt, ppm (wt)

Element	Slab	Wafer	Element	Slab	Wafer
U	0.44	0.12	Ag	<0.13	0.13
Th	<0.12	430	Pd	<0.23	<0.23
Bi	<0.11	<0.11	Rh	<0.19	<0.19
Pb	0.77	2.1	Ru	<0.19	<0.19
Tl	<0.38	<0.38	Mo	2.1	0.70
Hg	<0.37	0.37	Nb	<0.05	<0.15
Au	<0.10	<0.10	Zr	<0.11	0.11
Pt	<0.36	<0.36	Y	<0.05	<0.05
Ir	0.52	0.17	Sr	<0.18	<0.18
Os	<0.22	<0.22	Rb	<0.16	<0.16
Re	<0.16	<0.16	Br	<0.09	<0.09
W	<0.34	0.34	Se	0.29	0.09
Hf	<0.33	<0.33	As	0.14	0.42
Lu	<0.09	<0.09	Ge	<1.5	<1.5
Yb	<0.32	<0.32	Ga	<0.23	<0.23
Tm	<0.09	<0.09	Zn	3.0	9.0
Er	<0.31	<0.31	Cu	5.2	16
Ho	<0.08	<0.08	Ni	1600	1600
Dy	<0.36	<0.36	Fe	110	330
Tb	<0.08	<0.08	Mn	0.31	3100
Gd	<0.35	<0.35	Cr	3.5	10
Eu	<0.18	<0.18	V	<0.10	1.0
Sm	<0.33	<0.33	Ti	<1.6	1.6
Nd	0.32	0.32	Sc	<0.03	<0.03
Pr	<0.07	<0.07	Ca	2.2	66
Ce	<0.09	<0.09	K	0.22	<0.07
La	<0.07	<0.07	Cl	0.83	24
Ba	<0.12	<0.12	S	5.4	16
Cs	0.74	0.74	P	0.58	1.7
I	0.71	0.07	Si	10	150
Te	<0.24	<0.24	Al	1.5	45
Sb	0.14	4.2	Mg	4.5	4.5
Sn	<0.22	6.6	Na	0.43	<0.04
In	<0.06	0.18	F	0.11	0.11
Cd	25	<0.25	B	0.02	0.60

Analyses determined by spark source mass spectrometry of samples of wrought cobalt wafers and slabs as illustrated in Figures 7 and 9. Samples were representative of current production of wrought cobalt shapes.

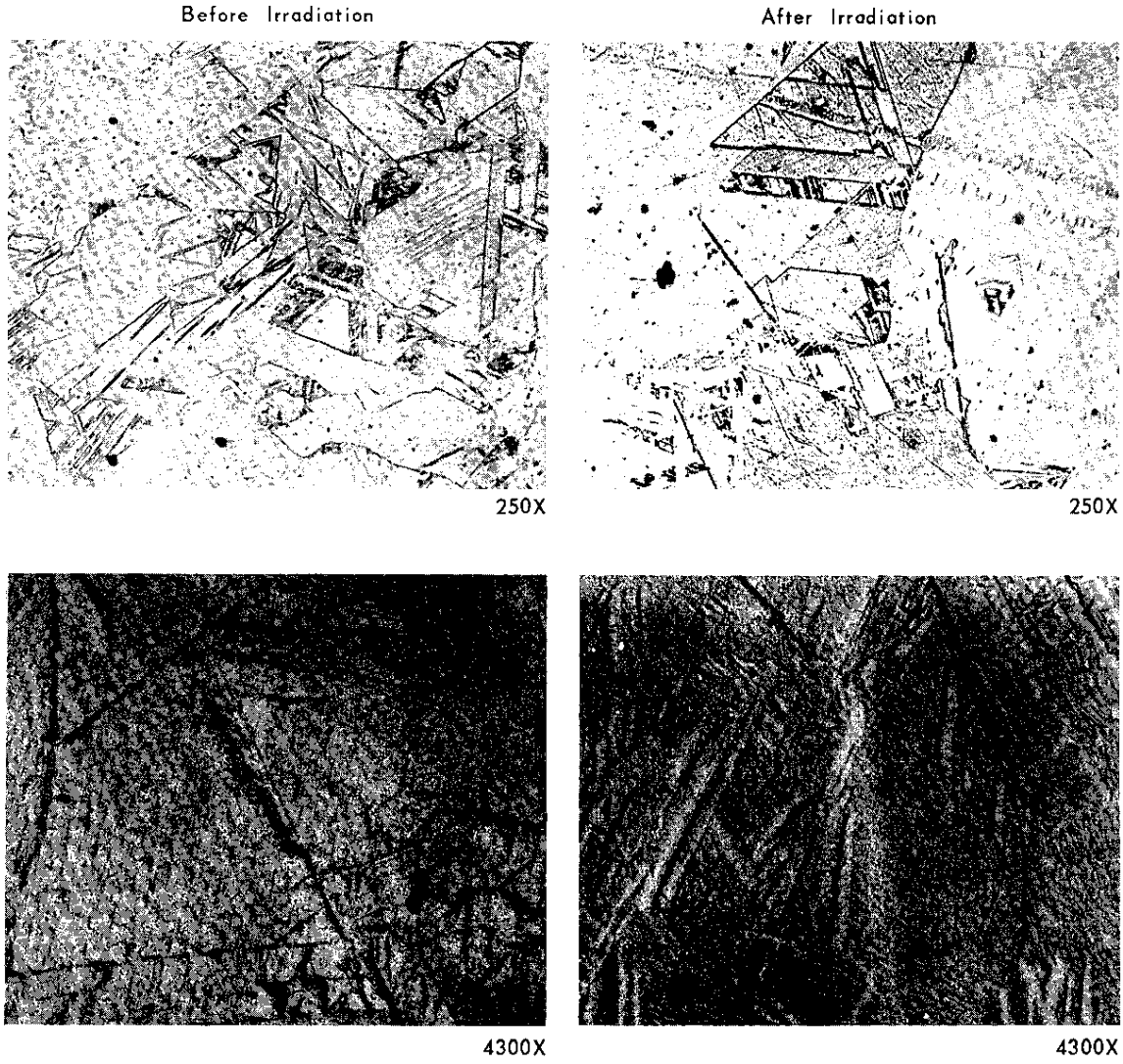


FIG. 10 EFFECT OF IRRADIATION ON MICROSTRUCTURE OF COBALT

(Cobalt slab as shown in Fig. 9)

Original structure of heavily twinned, relatively equiaxed grains is not altered by irradiation to an activity of 100 curies/g. Impurities originally present in solid solution are not precipitated during irradiation.

Production Capacity and Price

The Savannah River Plant (SRP) capacity for producing radioactive cobalt is very large. As much as 8 megacuries (125 kw) of cobalt-60 can be made per reactor-year as incidental production without interfering with the production of other materials[4]. Thus, the incidental output of the four SRP reactors could reach 32 megacuries (500 kw) per year. If the demand for radioactive cobalt exceeds the amount that can be supplied by incidental production, cobalt can be coproduced in charges that will also produce other isotopes. Larger amounts of cobalt-60 can be produced in full reactor charges specifically designed for cobalt production. Hundreds of megacuries of cobalt-60 can be produced per year by this mode of operation[7]. The technology required for full-scale production of cobalt-60 at Savannah River is presently available.

Annual production capacities, in kw per year, for cobalt-60 and several fission products are compared in Table III. The production capabilities for fission products in this comparison are those predicted in Nucleonics Week, July 29, 1965.[8] Even after the Isochem Plant is operating and with the very conservative assumption that cobalt-60 production is limited to 100 megacuries per year (1560 kw/yr), the heat available from cobalt-60 will be about equal to that available from all fission products combined.

TABLE III

Annual Production Capacities for Cobalt-60 and Fission Products

Isotope	^{60}Co	^{90}Sr	^{137}Cs	^{144}Ce
Half-life, years	5.3	28	30	0.78
Production capability, kw thermal/yr[a]				
Present and near future	>1560	35	20	30
After Isochem Plant (1968)	>1560	200	140	1600

(a) Initial thermal output. Because of rapid decay, ^{144}Ce capacity decreases rapidly after production.

Most of the predicted fission product heat is generated by ^{144}Ce, the only fission product that can be produced in large quantities. The half-life of ^{144}Ce is short (0.78 yr); hence, its use in heat sources will be restricted. Only cobalt-60 can supply a requirement for large long-lived heat sources.

The sale price for large quantities of cobalt-60 at the present time is 50¢/curie ($32/watt) for low specific activity cobalt (no more than 30 curies/g) to customers other than the Federal Government. The price for transferring radioactive cobalt to other Government agencies is less than 33¢/curie; this price is more than sufficient to compensate for the direct cost of producing the cobalt, including administrative overhead and depreciation. Each cobalt transfer is negotiated individually and the price depends on the amount of cobalt ordered, the desired specific activity, the required delivery dates, and the current operating schedule for the reactors. In the past, these transfer prices have ranged between less than 10¢/curie and 25¢/curie for incidental production of cobalt with an activity up to 120 curies/g. The price of radioactive cobalt with a specific activity of 400 curies/g will be about 20% higher than the price of low-activity cobalt, when produced incidental to other products.

The estimated prices for cobalt-60 and several fission products are compared in Table IV. Fission product prices, before and after the Isochem Plant is in operation, are those cited in Nucleonics Week, September 2, 1965[8]. The cobalt price is indicated as a range between 10¢/curie and 25¢/curie, representative of transfer prices to Government agencies.

TABLE IV

Prices of Cobalt-60 and Fission Products

Isotope	^{60}Co	^{90}Sr	^{137}Cs	^{144}Ce
Price, ¢/curie				
Present and near future	10-25	200	100	200
After Isochem Plant (1968)	10-25	20	12.5	15
Price, $/watt thermal				
Present and near future	6.4-16	300	210	250
After Isochem Plant (1968)	6.4-16	30	26	19
Price, $/kw-yr[a]				
Present and near future	1,700-4,200	15,000	10,000	430,000
After Isochem Plant (1968)	1,700-4,200	1,500	1,200	26,000

(a) Assumes service life equal to half-life of isotope.

PROPERTIES OF RADIOACTIVE COBALT

Expected Differences from Natural Cobalt

Radioactive cobalt is a mixture of cobalt-59, cobalt-60, and nickel. The properties of cobalt-59 are well documented, primarily in two excellent monographs[8,10]. The properties of radioactive cobalt may differ from the properties of cobalt-59 for several reasons:

- Differences between the properties of cobalt-59 and cobalt-60 - These differences are expected to be small.
- The effect of neutron and gamma irradiation during the production of cobalt-60 - Neutron irradiation causes some changes in the physical and mechanical properties of iron and nickel[11] and probably has similar effects on cobalt.
- The effect of beta and gamma irradiation from the decay of cobalt-60 during service - These radiations can cause structural changes resulting in physical and mechanical property changes in metals, but the most significant effect of beta and gamma radiation is on reaction rates[12].
- The effect of the gradual increase of the nickel decay-product - Because nickel forms a complete series of solid solutions with cobalt, as shown in Figure 11[8], the property changes with increasing nickel content due to radioactive decay are expected to be small and gradual.

The properties of unirradiated cobalt that are useful for the design of heat sources or radiation sources are described in the following pages with some comments on how these properties may be different for radioactive cobalt.

Physical Properties

The melting point of cobalt-59 is 1495°C (2723°F), and the melting point of radioactive cobalt is expected to be about the same. The addition of up to 25% nickel to cobalt will decrease the melting point by no more than 10°C, as shown by the cobalt-nickel phase diagram in Figure 11. The impurities that are present in typical reactor-grade cobalt as received at Savannah River have a negligible effect on the melting point. The heat of fusion is 61.8 cal/g.

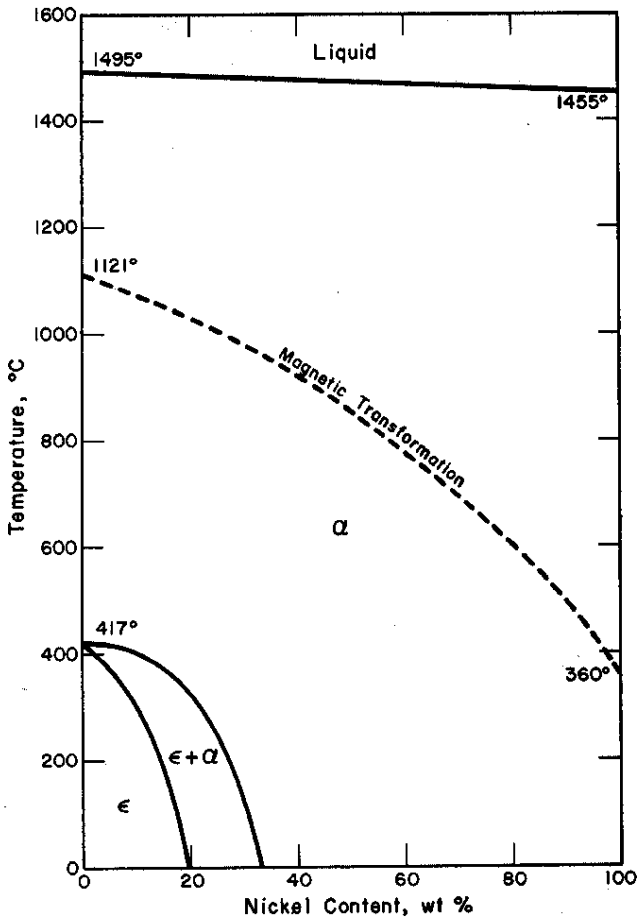


FIG. 11 COBALT-NICKEL PHASE DIAGRAM[9]

Cobalt is an allotropic metal that, under equilibrium conditions, has a close-packed hexagonal crystal structure, the ϵ phase, at temperatures below 417°C (785°F) and a face-centered cubic structure, the α phase, at temperatures between 417°C and the melting point. The transformation is sluggish and typically occurs at 390°C on cooling and at 430°C on heating[8]. The addition of 20 wt % nickel lowers the equilibrium transformation temperature to about room temperature. Since the high temperature (α) phase is frequently retained as a metastable phase at room temperature, the properties of cobalt are influenced by the relative amounts of the α and ϵ phases present. The heat of transformation is 1.0 ±0.5 cal/g. There is controversy regarding the existence of a second allotropic transformation from the cubic (α) phase to another hexagonal phase at or near the curie temperature of 1121 ±3°C (2050°F). Many phase transformations are sensitive to neutron irradiation[12] but there is no information on this effect in cobalt.

A volume increase of 0.30 ±0.06% occurs on heating cobalt-59 from the ϵ phase to the α phase. Rapid thermal cycling through the phase transformation temperature could cause internal cracking, as occurs in uranium and many other anisotropic metals. However, because the volume expansion is small (compared to the 3.0% expansion that occurs during the α to β transformation in uranium) and since the frequency and amplitude of the thermal cycles are expected to be small in a cobalt source, this cracking is expected to be negligible. If cracking becomes serious, some relief may be achieved by strengthening the cobalt by alloying, as has been done with uranium; or alternatively, the transformation temperature may be lowered by alloying so that the cobalt always operates in the α phase.

The boiling point of cobalt at 760 mm Hg has been reported to be between 2375 and 3550°C. A value of 3100°C (5612°F) is generally accepted. The heat of vaporization is 1550 cal/g. Data on vapor pressure are summarized in Table V.

The specific heat of cobalt increases with increasing temperature from about 0.09 cal/(g)(°C) at room temperature to a maximum of about 0.204 cal/(g)(°C) at 1100°C and then decreases to 0.141 cal/(g)(°C) for the molten state.

The thermal conductivity of cobalt-59 decreases from 0.22 cal/(sec)(cm)(°C) [53 Btu/(hr)(ft)(°F)] at room temperature to about 0.13 cal/(sec)(cm)(°C) [32 Btu/(hr)(ft)(°F)] at 500°C (932°F). The conductivity has been measured only at low temperatures[13]. The authors have calculated the conductivity for temperatures greater than 150°C using the Wiedemann-Franz Law and data for the electrical resistivity of cobalt[14]. Calculated data are plotted in Figure 12 along with the latest experimental determinations. The calculated and measured data agree fairly well. The accuracy of the predicted values can be estimated from the comparison of the calculated and measured thermal conductivities of iron and nickel in Figure 13. The effect of increasing nickel content in radioactive cobalt will be to decrease the thermal conductivity slightly at low temperatures and to increase it slightly at temperatures above 600°C. The effect of irradiation on the thermal conductivity of cobalt is expected to be small because reactor irradiation is known to have a negligible effect on the conductivity of pure nickel[15].

TABLE V

Summary of Physical Properties of Cobalt-59

(Data from Reference 9 except as noted)

Melting point	1495°C Impurities, including nickel decay product, decrease mp by <20°C as shown in Figure 11.
Heat of fusion[26]	61.8 cal/g
Transformation[27] temperature	417°C Transformation is sluggish and typically occurs at 390°C on cooling, 430°C on heating. Many properties are influenced by relative amounts of phases present.
Crystal structure	Close-packed hexagonal (ϵ) at <417°C (a_o = 2.524Å, c/a = 1.624Å) Face-centered cubic (α) at >417°C (a_o = 3.567Å)
Volume change[27]	0.30% increase with hcp to fcc transformation
Heat of transformation[26]	1.0 ±0.5 cal/g
Curie temperature	1121 ±3°C
Boiling point	3100°C
Heat of vaporization[26]	1,500 cal/g

Vapor pressure

Temperature, °C	Pressure, atm
1050	7.1×10^{-9}
1200	2.9×10^{-7}
1595	9.6×10^{-6}
2027	1.0×10^{-3}
2327	1.0×10^{-2}
3097	1.0

Specific heat

Temperature, °C	Specific Heat, cal/(g)(°C)
0	0.099
100	0.106
500	0.126
1100	0.204
1400	0.168
Molten	0.141

Thermal conductivity[13]

Temperature, °C	Conductivity, cal/(sec)(cm)(°C)
50	0.22
100	0.20
150	0.19
500	0.13

Coefficient of thermal expansion[28]	14.2 microinches/(inch)(°C) between 20 and 300°C 15.7 microinches/(inch)(°C) at 400°C 16.8 microinches/(inch)(°C) at 750°C
Density	8.85 g/cm³ solid forms 8.0 g/cm³ molten

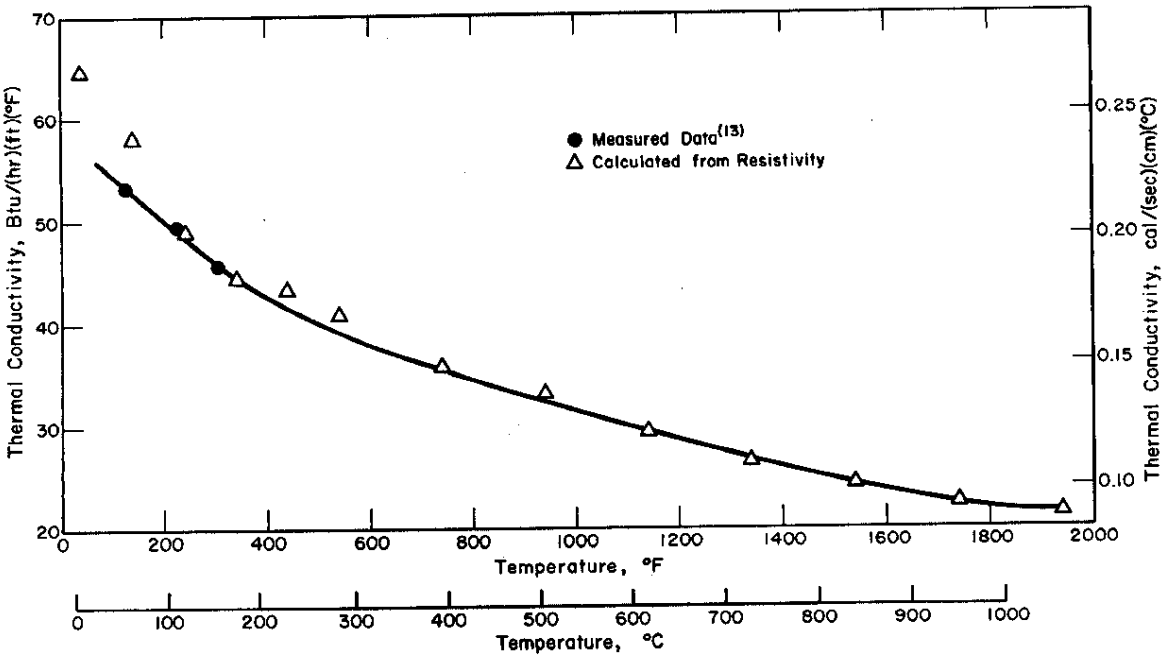


FIG. 12 THERMAL CONDUCTIVITY OF COBALT

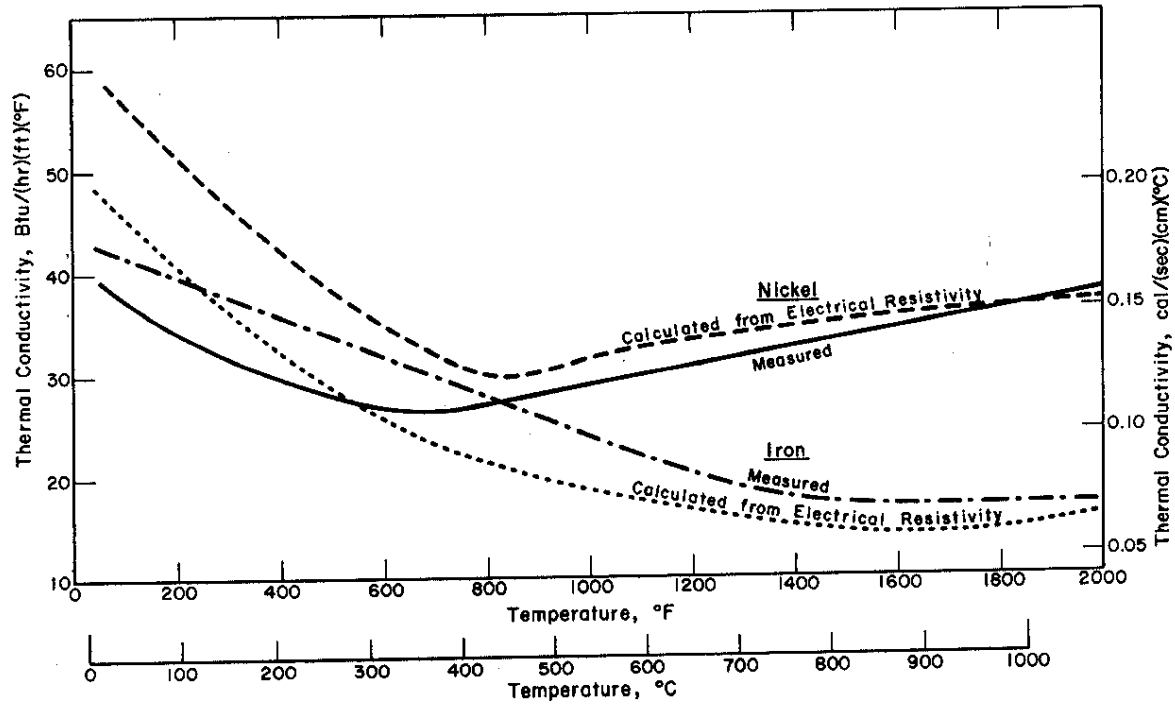


FIG. 13 THERMAL CONDUCTIVITY OF IRON AND NICKEL

The most recent data on pure cobalt indicate that the coefficient of linear thermal expansion is 14.2 microinches/(inch)(°C) between room temperature and 300°C and increases to 15.7 microinches/(inch)(°C) at 400°C and to 16.8 microinches/(inch)(°C) at 750°C. Different values reported by various investigators may be due to the effects of impurities or to the effects of anisotropy associated with the hexagonal ϵ phase. The coefficient of thermal expansion parallel to the hexagonal axis has been measured to be 28% greater than the coefficient of expansion perpendicular to the hexagonal axis at room temperature. Thus, the effects of preferred orientation must be considered when the properties of fabricated cobalt are characterized.

The densities of ϵ- and α-cobalt are, at room temperature, about 8.85 g/cm^3 (0.32 lb/in^3) and 8.80 g/cm^3, respectively. The density of molten cobalt is 8.0 ±0.2 g/cm^3.

The physical properties of cobalt are summarized in Table V.

Mechanical Properties

The tensile strength of cobalt at room temperature varies from about 120,000 psi for wrought metal of normal purity to 137,000 psi for hot-rolled and zone-refined metal. Strengths as low as 35,000 psi have been reported for cast and annealed material. Tensile strength decreases linearly with increasing temperature as indicated by the data in Table VI[16,17]. Proportions of the α and ϵ phases present have relatively little effect on the tensile strength of cobalt but they have a large effect on the ductility; the room-temperature elongation of wrought cobalt increases linearly from 4 to 26% as the amount of α cobalt increases from 7 to 50%[17]. The ductility gradually increases with increasing temperature to reach a maximum near 500°C and then decreases rapidly with further temperature increases. Irradiation causes a slight increase in the tensile strength of nickel alloys, a large increase in the yield strength, and a large decrease in the ductility[11]. Similar trends are anticipated in irradiated cobalt.

Although creep data for cobalt are extremely limited, a design stress of 12,000 psi at 500°C (932°F) appears practicable. Under these conditions, the elongation at the end of first-stage creep, after about 100 hours, is 2.7% and the minimum creep rate is about 10^{-6} in./(in.)(sec)[18].

The elastic modulus (Young's modulus) for cobalt decreases from about 30 x 10^6 psi at room temperature to about 24 x 10^6 psi at 700°C (1292°F). The shear modulus decreases from about 12 x 10^6 psi at room temperature to about 8 x 10^6 psi over the same temperature range.

Poisson's ratio for cobalt is about 0.32. The relative amounts and orientation of the phases present are important for measurements of elastic moduli because of the anisotropic behavior of the hexagonal crystals of ϵ cobalt.

The mechanical properties of cobalt are summarized in Table VI.

TABLE VI

Summary of Mechanical Properties of Cobalt-59

Tensile Properties[16,17]

	Ultimate Strength, 10^3 psi		Uniform Elongation, %	
Temp, °C	Zone Refined[a]	Wrought[b]	Zone Refined[a]	Wrought[b]
25	137	120	-	4-26[c]
100	85	100	17	20
400	50	60	32	20
500	35	35	63	40
700	15	15	10	15
1,000	-	7	-	5

Creep Properties[18]

Temp, °C	Stress, 10^3 psi	Minimum Creep Rate, in./(in.)(sec)
400	23	10^{-7}
500	28	10^{-6}
600	23	10^{-4}
750	11	10^{-4}

for 99.999% cobalt in vacuum

Elastic Characteristics[9]

Temp, °C	Elastic Modulus, 10^6 psi	Shear Modulus, 10^6 psi
25	30	12
100	30	12
400	27	10
500	26	9
700	24	8

Poisson's ratio = 0.32

(a) Hot rolled and stress relieved 1 hr at 350°C.
(b) Roll compacted powder, sintered, hot rolled, cold rolled, and annealed 1 hr at 930°C.
(c) Elongation increases linearly from 4 to 26% as amount of fcc phase increases from 7 to 50%.

Corrosion Properties

The corrosion of natural cobalt in various media has been studied by many investigators. Cobalt is attacked slowly by both fresh water and sea water; the corrosion rates are low, about 0.0001 inch/yr in fresh water and about 0.0050 inch/yr in sea water. Cobalt is attacked vigorously by nitric acid at temperatures above 25°C. The addition of nickel slightly improves the corrosion resistance in nitric acid, but the effect in water is unknown. The corrosion resistance of cobalt alloys in liquid mercury is fair[19], and cobalt-base alloys are being studied for possible use in metal-cooled systems for aerospace applications. The effect of irradiation on the corrosion of cobalt is a major uncertainty; there is some evidence that irradiation may cause the corrosion rate to increase, as described in the next paragraph.

Cobalt is not resistant to oxidation and scaling and generally compares with zirconium in this respect. Oxidation follows the parabolic time relationship and becomes severe at temperatures above 700°C. There is some evidence that ϵ cobalt oxidizes more rapidly than α cobalt. Radioactive cobalt has been observed to deteriorate rapidly when stored bare in air at the Oak Ridge National Laboratory.[20] This deterioration may be caused by the radiolytic formation of nitrogen oxides by gamma rays from the radioactive cobalt, and subsequent attack of the cobalt by nitric acid produced by the reaction of the oxides with moisture in the air. Further studies of the oxidation resistance of radioactive cobalt are required to determine the seriousness of this problem.

Diffusion Properties

The temperature dependence of the diffusion of one material into another is described by the equation:

$$D = Ae^{-\frac{Q}{RT}}$$

where D = diffusion coefficient, cm^2/sec

T = temperature, °K

A = diffusion constant, cm^2/sec

Q = activation energy, cal/mole

Values of these constants for the diffusion of cobalt in various metals are summarized in Table VII.

TABLE VII

Summary of Corrosion and Diffusion Properties of Cobalt-59

Corrosion rate (typical conditions of exposure)	~0.0001 in./yr in fresh water ~0.005 in./yr in sea water

Oxidation Rate in Air[29]

Temp, °C	Parabolic Scaling Constant, $(mg/sq.cm)^2/hr$
400	0.0004
600	0.1
800	2.1
1000	70.0
1200	400.0

Oxidation follows parabolic rate law; more rapid for hcp than for fcc form. Some evidence that irradiated cobalt oxidizes more rapidly than unirradiated cobalt at room temperature.

Diffusion Characteristics[9]

System	Temp Range, °C	Diffusion Constant, sq.cm/sec	Activation Energy, cal/mole
Co	1040-1250	0.3	64,000
Fe_α	700-1200	0.2	54,000
Fe_γ	700-1200	300	87,000
Ni	700-1200	1.46	68,300
Cu	700-1200	1.93	56,500
Al	350- 600	1.1×10^{-6}	19,900

One design requirement for power sources is the ability of the cladding material to contain the cobalt during the life of the source. This containment is controlled by the rate of diffusion of the cobalt through the cladding. The diffusion data in Table VII[9] were extrapolated, by a nomographic technique described by Seith,[21] to predict the distance from the original cobalt-cladding interface at which the concentration of cobalt would reach 0.01 wt % after 53 years at 600°C. This time was selected because it corresponds to ten half-lives, the frequently quoted time period for safety considerations. The results of this extrapolation follow.

Diffusion of Cobalt in Various Materials

Material	Distance from Original Interface, inch[a]
Co	0.00012
Fe_γ	0.000016
Ni	0.00024
Cu	0.0079
Al	0.079

(a) Assumed conditions were a time of 53 years at a temperature of 600°C. The distance is that at which the concentration would reach 0.01 wt % Co.

These data indicate that cobalt itself or almost any alloy whose matrix is austenite (Fe_γ), such as 300 series stainless steel, or whose matrix is Ni, such as "Hastelloy"* or "Inconel"**, would be a suitable cladding material from the standpoint of containing the cobalt. Copper would be less satisfactory and aluminum would probably be unsuitable.

SAFETY

Shielding

The intense and energetic gamma activity that results from the decay of cobalt-60 requires heavy shielding to reduce the radiation to tolerable levels. For shielding calculations, it is sufficient to consider that the radiation consists entirely of the following three constituents:

Beta particle

Maximum energy, Mev	0.312
Average energy, Mev	0.095

Gamma rays

Energy, Mev	1.172	1.333
Emission rate, photons/watt-sec	2.385×10^{12}	2.385×10^{12}

* Trademark of Haynes Stellite Co.
** Trademark of International Nickel Co.

The results of shielding calculations[22] for idealized cobalt heat sources are plotted in Figure 14; the radioactive cobalt is assumed to be a solid sphere surrounded by a spherical shield of depleted uranium of uniform thickness. Since no provision for cooling the source is included, the data in Figure 14 may be used for comparative purposes only. Analysis of the data shows that the thickness of the required shield is not influenced strongly by the power of the heat source. Increasing the source power and maintaining a constant cobalt specific power increase the amount of cobalt required, and hence, the dimensions of the cobalt core. Thus, the shield thickness occurs at a larger diameter and the shield weight increases with increasing source power. This effect limits the maximum practical size of a single cobalt heat source. If weight is an important consideration, the use of high-activity cobalt can result in appreciable weight reductions, as indicated in Figure 14, but this weight reduction must be balanced against the higher cost of the high-activity metal as described previously.

The shielding required for cobalt heat sources producing between 100 watts and 20 kw has been summarized by Arnold.[23]

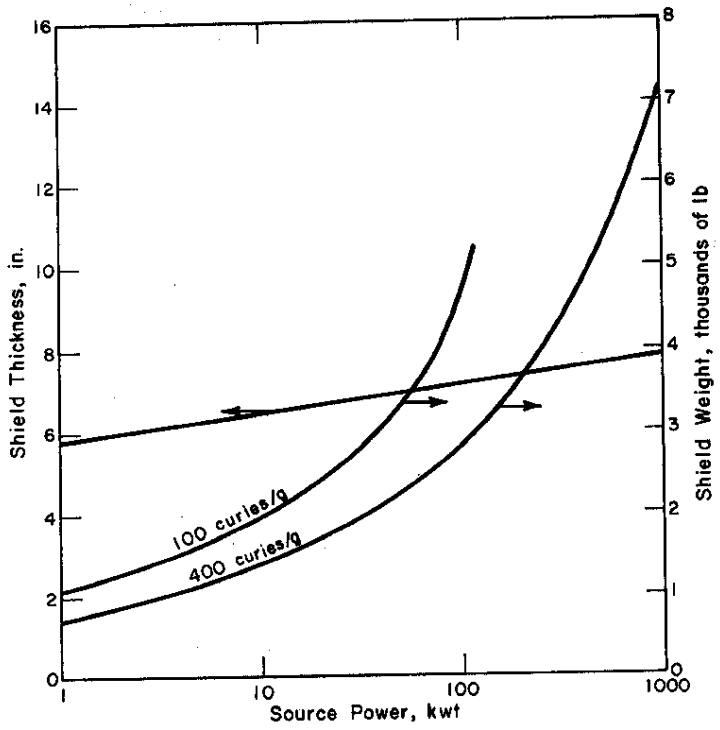


FIG. 14 SHIELDING REQUIRED FOR COBALT HEAT SOURCES
(For comparative purposes only)

Depleted uranium shield to reduce radiation to < 200 mr/hr at shield surface, < 10 mr/hr at 1 meter. Solid spherical source at 100 curies/g and 400 curies/g. Spherical shield.

About 95% of the heat produced in a cobalt heat source is due to the gamma radiation. Most of this radiation must be absorbed so that the heat can be transferred to the working fluid. Self-absorption of the gamma rays within the cobalt can be increased by increasing the cobalt thickness (at the cost of higher cobalt temperatures) as shown below:

Self-Absorption of Gamma Energy in Cobalt

Diameter of Cobalt Cylinder, in.	Estimated Gamma Energy Absorbed, %
0.5	15
1	26
2	42
3	54

Most of the radiation that escapes from the cobalt is absorbed in the innermost layers of the shield. About 74% of the incident cobalt gamma energy is absorbed in the first 0.5 inch of a depleted uranium shield and about 90% is absorbed in the first inch. Thus, only the inner layer of the shield must be cooled by the working fluid to achieve efficient utilization of the radioactive cobalt.

Ingestion Hazards

Cobalt-60 is less toxic biologically than most other isotopes that are potentially useful in heat sources. The relatively large tolerances for the ingestion of cobalt-60 result from several factors:

- Soluble cobalt-60 is removed rapidly from the body by normal metabolic processes. Many other isotopes are concentrated within the body and have long effective metabolic half-lives.
- Within the body, sources of beta and gamma radiation are less damaging than internal sources of alpha radiation.
- The decay of cobalt-60 produces only stable nickel-60 as shown in Figure 1. Decay of several of the other isotopes produces radioactive daughter products whose activities also must be considered.

The factors contributing to the maximum permissible body burdens and the relative hazards of the isotopes that are considered for use in heat sources are compared in Table VIII[24].

TABLE VIII

Ingestion Hazards of Isotopes for Heat Sources

Isotope	Effective Metabolic Half-Life, days[a]	Critical Organ	Activity	Max Energy, Mev[b]	Relative Hazard per watt[d]
^{60}Co	9.5	Gastrointestinal	β γ	0.312 1.333	1.0
^{210}Po	46	Spleen	α	5.305	350
^{137}Cs	138	Total body	β γ(c)	1.18 0.662	8.0
^{242}Cm	162	Gastrointestinal	α	6.11	1.1
^{144}Ce	243	Gastrointestinal	β γ	0.31 0.134	9.5
^{147}Pm	570	Gastrointestinal	β	0.23	10.7
^{244}Cm	6,100	Kidney	α	5.801	2.6
^{90}Sr	6,400	Bone	β	0.545	1150
^{238}Pu	23,000	Bone	α	5.495	45.3

(a) Maximum metabolic half-life for critical organ[18].
(b) Maximum energy of individual particle or photon.
(c) Gamma from decay of ^{137}Ba daughter product.
(d) Relative hazard of one watt of isotope in soluble form. Based on MPC in water for 168-hr week converted to equivalent watts/cm³[24].

TABLE IX

Maximum Permissible Concentrations of Cobalt-60 for Occupational Exposure[24]

Cobalt Form	Organ of Reference	Max Permissible Body Burden (Total Body), microcuries	Max Permissible Concentration, microcuries/cm³ For 40-hr Week Water	For 40-hr Week Air	For 168-hr Week Water	For 168-hr Week Air
Soluble	Gastrointestinal		10^{-3}	3×10^{-7}	5×10^{-4}	10^{-7}
	Total body	10	4×10^{-3}	4×10^{-7}	10^{-3}	10^{-7}
	Pancreas	70	0.02	2×10^{-6}	7×10^{-3}	6×10^{-7}
	Liver	90	0.03	10^{-6}	9×10^{-3}	5×10^{-7}
	Spleen	200	0.05	4×10^{-6}	0.02	2×10^{-6}
	Kidney	200	0.07	6×10^{-6}	0.03	2×10^{-6}
Insoluble	Lung		-	9×10^{-9}	-	3×10^{-9}
	Gastrointestinal		10^{-3}	2×10^{-7}	3×10^{-4}	6×10^{-8}

Maximum permissible concentrations of soluble and insoluble forms of cobalt-60 in water and air are tabulated in Table IX[24]. The limiting concentrations in drinking water are approximately the same for soluble and insoluble cobalt compounds, but the limiting concentrations in air are much smaller for insoluble compounds than for soluble compounds. The lower values should be used for design purposes.

A standard for the maximum permissible concentration of cobalt-60 in sea water has not been adopted to date. A recent study group proposed a maximum permissible concentration of 5.0×10^{-9} $\mu c/cm^3$ for disposal of cobalt-60 into Pacific coastal waters[25]. Higher and lower values have been proposed by other study groups; the differences reflect the use of different assumptions, including diet habits, sea water chemistry, and degree of dispersal in the sea[25].

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