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PRODUCTION OF HIGH SPECIFIC ACTIVITY COBALT-60 IN HIFAR

by

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Summary

High specific activity cobalt 60 sources, as used in teletherapy work, may be produced in HIFAR. Approximate calculations indicate that a specific activity of 30 curies per gram per year can be made in the 2V and 6V experimental holes, and that a total activity of 21,000 curies per year in these holes corresponds to using 1% excess k.

February, 1959.

CONTENTS

1. INTRODUCTION

A survey of hospitals and radiotherapy clinics in Australia has shown that the requirements of high specific activity cobalt–60 in the future may amount to as much as 10 or 20 kilocuries per annum. There is also a world wide demand for this material which will be difficult to meet, particularly in the sterling area, and Australian production may be able to assist by producing cobalt–60 for export.

There is also likely to be a considerable demand for large quantities of lower specific activity material for industrial purposes.

Sources of cobalt–60 of specific activity of 20 to 50 curies per gram and total activity 500 to 2,000 curies are used for teletherapy work. Higher specific activity sources are desirable, but the main limitation is the high thermal neutron flux required for activation.

Cobalt–60 is produced in a nuclear pile by the reaction; Co^{59} (n, γ) Co^{60}. It has a half-life of 5.25 years, emits γ–rays of energy 1.17 and 1.33 MeV, and a principal beta of 0.31 MeV (1).

It is the aim of this report to investigate:

(i) the specific activity of cobalt–60 obtainable in HIFAR,

(ii) the total activity of high specific activity cobalt–60 which may be produced per year.

2. SPECIFIC ACTIVITY versus TIME

2.1 Discussion

To produce cobalt–60 of specific activity 30 to 50 c/g. a high thermal neutron flux, as available in the 2V and 6V holes in HIFAR is required. This flux is of the order of 5×10^{13} neutrons/cm^2/ sec.

A calculation of the rise of activity with irradiation time is not simple, and various authors (2, 5) have found that theory and experiment do not agree well. The simple activation equation (1) does not hold. Various other factors have to be considered: "self protection", flux depression, and the reduction of the number of cobalt 59 nuclei in the sample due to the formation of cobalt 60 nuclei. Each of these factors reduces the specific activity which can be produced.

"Self protection" is the shielding of the inner layers of the sample being irradiated by the outer layers, and consequently the neutron flux is depressed within the sample. Cobalt 59 has a high neutron absorption cross–section of ~ 30 barns at 2,200 m/sec (3) and so this effect is large in cobalt samples. The neutron linear absorption coefficient μ is given by:

$$\mu = \frac{N \sigma \rho}{A} \qquad (1)$$

where N is Avagadro's number, σ the neutron absorption cross–section, ρ the density and A the atomic weight of the sample. For cobalt of density 8.7 g/cc and neutron absorption cross–section 30 barns, the linear absorption coefficient is 2.65 per cm, and the half thickness ~ 2.6 mm. The "self protection" may be reduced by making one dimension of the sample small, e.g. cobalt discs of 2.5 mm thickness are used in DIDO, and 1 mm and 2 mm thickness in MTR.

Local depressions of flux in a reactor are caused by the presence of other absorbers near the sample being irradiated. Ideally it would be best to space absorbers so that no interaction occurred. In practice there is a limited volume of space available in high flux positions, and it is necessary to compromise between number of samples to be irradiated and the specific activity which is desired. In DIDO cobalt discs are supported in a stringer with each disc placed 2 cm apart (3). The depression of flux in the 2V holes about one disc due to the other discs in the stringer is such that the resulting flux is about 0.86 of the undisturbed flux (3).

With the buildup of cobalt 60 nuclei in a sample being irradiated, there is a corresponding reduction in the number of cobalt 59 nuclei , and so the effective neutron absorption for cobalt 60 production becomes less. At specific activities being considered in this report, the ratio of cobalt 60 to cobalt 59 atoms is roughly 0.03, and so this effect is small.

2.2 Specific Activity versus Time for Flux ϕ

The rise of activity with irradiation time is calculated, allowing for flux depression and self protection, but neglecting the small effect of the nuclei formed reducing the effective cobalt 59 absorption. If a sample of surface area α is placed, with other absorbers, in a position of undisturbed flux ϕ thermal neutrons/cm^2/sec, then the number of neutrons absorbed/sec by the sample is:

$$g \times \frac{\phi}{2} \times f \times \alpha \qquad (2)$$

where g = flux depression factor, and is the ratio of the flux about the sample (depressed due to the presence of other absorbers) to the undisturbed flux.

and f = fraction of neutrons entering the sample absorbed in traversing it.

The activity of the sample produced in an irradiation time τ is given by:

$$\text{Activity} = g \times \frac{\phi}{2} \times \frac{f \times \alpha}{3.7 \times 10^{10}} \left(1 - e^{-\lambda\tau}\right) \text{ curies} \qquad (3)$$

where λ is the decay constant of the isotope.

The activity of cobalt 60 produced will depend on the dimensions of the cobalt sample, and the effect of neighboring absorbers. For this report it will be assumed that the samples are similar to those irradiated in DIDO, i.e. discs of diameter 1.7 cm, thickness 2.5 mm, and density 8.7 g/cc, 30 of which are supported in a 2 ft. long stringer, with each disc spaced 2 cm apart (3). The flux depression factor, g, has been measured for the stringer in a 2V hole in DIDO, and is approximately 0.86 (3).

The fraction, f, of neutrons absorbed in traversing the disc may be estimated if the angular distribution of the thermal neutrons is known. With no absorbers present in HIFAR, the distribution near the core can be assumed approximately isotropic, but strong absorbers as cobalt alter the local flux distributions. Two flux distributions will be considered: isotropic, and a beam of neutrons travelling parallel to the axis of the disc. The value of f calculated for these two distributions will probably be near maximum and minimum values and so an activation equation can be calculated within limits of two values of f. Both calculations of f assume that the absorption cross-section of cobalt 59 for thermal neutrons >> scattering cross-section [36 and 6 barns respectively (4)]. The calculation for the isotropic distribution assumes that the percentage of neutrons absorbed in traversing the disc is the same as that for an infinitely extended plane sheet of thickness that of the disc. If f_i (x) is f for an isotropic neutron distribution, where x is the thickness of the disc, Green (5) has shown that:

$$f_i(x) \simeq 1 - 2 \int_1^{\infty} \frac{e^{-\mu xy}}{y^3}\, dy \qquad (4)$$

For a beam of neutrons traversing the disc parallel to its axis, then if $f = f_b(x)$,

$$f_b(x) \simeq 1 - e^{-\mu x} \qquad (5)$$

The disc as irradiated in DIDO is 2.5 mm thick, and from equation 1, $\mu = 2.65$/cm, and so

$$f_i(x) \simeq 0.65$$
$$f_b(x) \simeq 0.48 \qquad (6)$$

With the calculated values of f and experimental values of g, the activity of cobalt 60 produced in an irradiation time τ may be calculated. The surface area α of the discs $\sim$ 5.54 cm^2, and the half life of cobalt 60 is 5.25 years. Irradiation times to produce high specific activity cobalt 60 will be of the order of a year, and equation (3) assumes the activation is continuous. HIFAR will be operating approximately 82% of the year, and if t is normal time, then the activity, as in equation (3), may be calculated to a good approximation, if $\tau = 0.82\,t$ is substituted. If t is time in years, then the activation equation for the cobalt disc, is, from equation (3):

$$\text{Activity} \simeq \begin{pmatrix}3.44\\2.53\end{pmatrix} \times 10^{-11}\, \phi \left(1 - e^{-0.107t}\right) \text{curies/disc} \qquad (7)$$

where the two figures 3.44 and 2.53 result from the two calculated values of f. These figures do not necessarily determine the limits within which the activity is calculated, as errors also arise from uncertainties in ϕ, g and τ.

2.3 Specific Activity versus Time in HIFAR

To produce high specific activity cobalt 60, it is necessary to place the cobalt samples near the reactor core, i.e., in the 2V or 6V holes in HIFAR. Most available experimental holes with a high flux are at 55.5 cm from the centre of the core, and the rise of activity with time will be calculated for 2V holes at this distance. There is little published information of neutron fluxes in experimental holes in DIDO for full power reactor operation. Fenning (6) gives an experimentally measured value of the thermal neutron flux in an empty 6V hole (55.5 cm from the centre of the core) in DIDO as 0.41×10^{14}n/cm^2/sec. The flux will be higher in 2V holes at the same distance, mainly due to less streaming of neutrons up the smaller experimental hole. An approximate value of flux in the 2V hole may be calculated from Fenning's value in a 6V hole, by multiplying it by the ratio of the fluxes in empty 2V and 6V holes at the same distance from the centre of the core for clean core conditions, given as 0.66 : 0.56 (7). The value of flux in the empty 2V hole, for full power reactor operation, will then be approximately 0.48×10^{14} n/cm^2/sec. Substituting this value of ϕ in equation (7), the activity per disc per year will be between 124 and 168 curies, i.e. a specific activity of between 25 and 34 curies/g/year. Published values for DIDO in 2V holes, with an unspecified neutron flux, are 20 – 40 c/g (3), and so the calculated value agrees reasonably well with the experimental one.

The main purpose of the calculation has been to give an indication of the specific activity of cobalt 60 produced per year in a certain neutron flux, i.e. 30 c/g/year in a flux of approximately 5×10^{13} n/cm2/sec. Figure 1 shows the rise of specific activity with time, assuming an activity of 30 c/g per first year's irradiation.

3. TOTAL ACTIVITY PRODUCED PER YEAR IN HIFAR

It is desirable to estimate the total activity of high specific activity cobalt 60 which may be produced per year in HIFAR. A limit to this total activity will be determined by either the excess k used in producing the cobalt or the number of experimental holes available for cobalt 60 production in high flux positions.

A very approximate method of calculating the cobalt 60 produced corresponding to absorbing neutrons equivalent to 1% excess k is as follows. HIFAR will operate at 10 MW power, and since the energy per U235 fission is ~ 200 MeV and the number of neutrons produced per fission ~ 2.5, then the number of neutrons produced/sec in HIFAR is:

$$\sim \frac{10\ \text{MW}}{200\ \text{MeV}} \times 2.5$$

$$\sim 7.8 \times 10^{17}\ /\text{sec}$$

The number of neutrons corresponding to 1% excess k is then 7.8×10^{15}/sec.

Substituting this number of neutrons absorbed per second in equation (3) i.e. $g\frac{\phi}{2}fA \approx 7.8 \times 10^{15}$, then a total of 21,000 curies per year corresponds to an excess k of 1%. This calculation should underestimate the total activity of cobalt 60 which may be produced in experimental holes far from the core, since some of the neutrons which normally would have escaped the reactor would be absorbed by the cobalt. However, for activation in holes very near the reactor core, this calculation overestimates the total activity. C. McKenzie (7) has estimated, based on an experimental figure for the reactivity controlled by a cobalt stringer in a 2V hole 45.7 cm from the core in a DIDO/PLUTO mockup in DIMPLE (8), that the total number of neutrons absorbed per second by the stringer is 6.4×10^{15}, i.e. a total of 17,500 curies per year corresponds to using 1% excess k. The reactivity controlled by a stringer falls off more quickly than the neutron flux as the stringer is placed in positions further from the reactor core, and consequently a higher value than 17,500 curies corresponding to 1% excess k will be produced per year in the 2V holes 55.5 cm from the centre of the reactor core.

The number of cobalt stringers which control 1% excess k may be estimated. An experimental figure for the specific activity of cobalt 60 produced per year in 2V holes is approximately 30 c/g (Section 2.3). Each stringer holds approximately 150 grams of cobalt and so 4,500 curies per stringer per year are produced. A total activity of approximately 21,000 curies per year corresponds to using 1% excess k, so 5 stringers will control approximately 1% excess k.

The limitation of the total amount of high specific activity cobalt 60 which may be produced per year will probably be determined by the available reactor space in high flux positions. There are 8 2V holes in HIFAR. If 5 of these are used for cobalt 60 production, then, very approximately, 21,000 curies per year will be produced.

4. CONCLUSION

High specific activity cobalt 60 sources may be produced in HIFAR and activities of ~ 30 c/g per year may be obtained in the 2V holes. A total of 21,000 curies per year, in holes close to the core (2V and 6V) corresponds, very approximately, to 1% excess k.

The calculations in this report are very approximate, and experimental work is required to determine the results more accurately. The total activity of high specific activity cobalt 60 produced per year will probably be limited by the number of available high flux experimental holes. Experimental work should be undertaken to determine whether, by variation of disc configuration, etc., a higher total activity of high specific activity cobalt 60 could be produced per experimental hole.

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6. REFERENCES

(1) A.E.R.E. "Radioactive Materials and Stable Isotopes – Catalogue No. 4" (1957).

(2) Levin, J.S. et al "Flux depression and self protection in production of radio cobalt". Nucleonics 11, 7, 9 (1953).

(3) Eastwood, W.S. et al "Cobalt 60 production in the U.K.A.E.A." – International Conference on the Peaceful Uses of Atomic Energy A/CONF/15/P/288 (1958).

(4) Hughes, D.J. and Harvey, J.A. "Neutron cross-sections." BNL.325 (1955).

(5) Green, D.T. et al "Production of multicurie γ – ray teletherapy sources." Nucleonics 11, 5, 29 (1953).

(6) Fenning, F.W. "The First Year of DIDO Operation" Symposium on the Peaceful Uses of Atomic Energy in Australia, 1958.

(7) C. McKenzie, private communication.

(8) Hicks, D. "Experiments with DIDO–PLUTO Mock–Ups in Dimple" – A.E.R.E. R/R 2337 (1957).

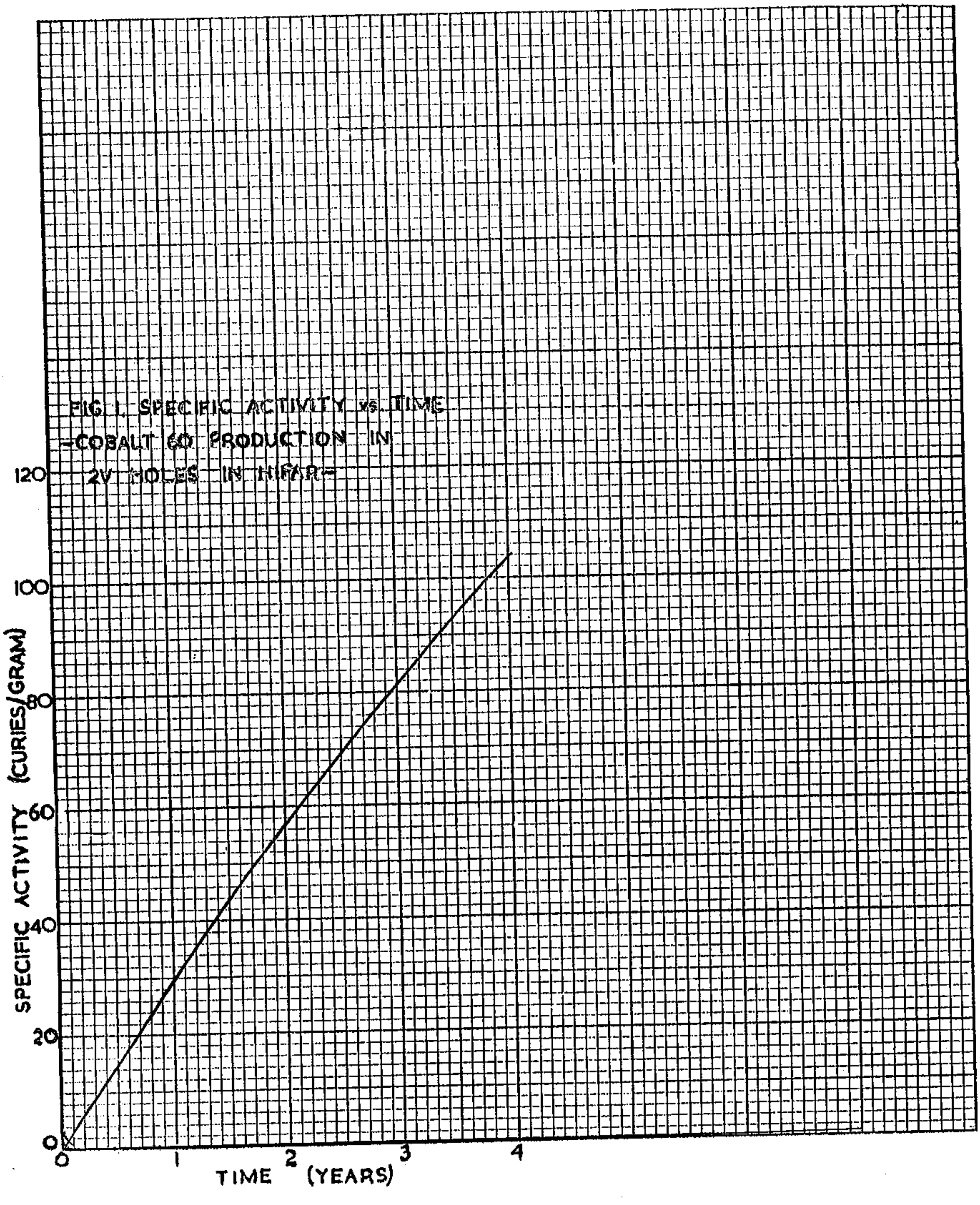

FIG. 1. SPECIFIC ACTIVITY vs. TIME
-COBALT 60 PRODUCTION IN
2V HOLES IN HIFAR-