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SUBJECT: Estimate of the Ratio of Ta¹⁸² to Co⁶⁰ Activity
Expected in the APPR-1 Core

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ESTIMATE OF THE RATIO OF TA^{182} TO CO^{60} ACTIVITY
EXPECTED IN THE APPR-1 CORE

The Army Package Power Reactor I (APPR-1) is one of the first reactors to use stainless steel as a core structural material. The good high temperature properties, ease of fabrication and availability of stainless steel were felt to override the poor neutron economy expected from its use as a core material. Furthermore, the availability of highly enriched uranium and the nature of the military uses of reactor power made neutron economy a secondary consideration.

Of primary interest in a military (as well as a civilian) reactor is the desirability of quick and easy maintenance of as much of the system as possible throughout the lifetime of the power plant. It is therefore desirable that no serious amounts of long-lived activity build up in equipment outside of the core. Preliminary estimates^{1,2} of the activation products expected from the use of stainless steel as the cladding material for the APPR-1 did not reveal any significant quantities of long-lived activation products. These estimates neglected reactions whose cross sections were unknown and, of course, they also neglected reactions with stainless steel impurities whose concentrations were too small to be reported by the manufacturer.

Recent measurements of long-lived activation products in the APPR-1 water stream³ as well as in the APPR-1 demineralizer⁴ have revealed that there are significant quantities of long-lived (greater than 50 days)

1. W. R. Pearce, "Analysis of Water Activation and Component Shielding for the ORNL Package Reactor Power Plant," ORNL-CF-53-10-168, Nov. 10, 1954.
2. J. L. Meem, and F. B. Fairbanks, "Shielding Requirements for the Army Package Power Reactor," APAE-3, May 1, 1956.
3. W. J. Small et al, "Long-Lived Circulating Activity in the Army Package Power Reactor," APAE-20, Aug. 28, 1957.
4. S. A. Reynolds, "Analysis of APPR-1 Demineralizer Contents," ORNL-CF-58-2-2, Feb. 6, 1958.

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activation products after 700 full power hours of reactor operation (design life is 1.5 years). The principle activation products appear to be Co^{58} , Co^{60} , Fe^{55} , Fe^{59} , Mn^{54} and Cr^{51} . The relative importance of these products depends on the sample observed (water sample or demineralizer sample) as well as on the accumulated time of operation of the reactor.³ The surprising result of these early measurements is the importance of Co^{58} activity which results from the $\text{Ni}^{58}(\text{n,p})\text{Co}^{58}$ reaction. Although this reaction has a negative threshold energy (-0.63 Mev), the reaction is suppressed until the incoming neutron supplies enough energy to overcome the coulomb barrier of Ni^{59} for protons (about 5 Mev). Thus this reaction is not expected to have much of a yield for neutrons with energies less than about 4.5 Mev. Neutrons with energies above this effective threshold energy comprise only about 7% of the fission spectrum.⁵ Together with the lack of knowledge concerning the $\text{Ni}^{58}(\text{n,p})\text{Co}^{58}$ cross section, it is understandable that this activation product was overlooked by the preliminary estimates. The neglect of Co^{60} as an important activation product is also understandable since previous customers for stainless steel have had little or no interest in cobalt as an impurity. Thus the analysis provided by the producer did not include Co^{59} as an impurity and the percentage of Co^{59} in the APPR-1 core material was not known at the time of fabrication of the core. Subsequent analysis⁶ of residual core material has revealed the presence of about 0.05 wt. % of Co^{59} in the APPR-1 stainless steel cladding.

Prior to the above results, the importance of the buildup of long-lived activity due to the use of stainless steel as a core material was not fully appreciated. Recommendations for the choice

5. B. E. Watt, Phys. Rev. 87, 1037 (1952).

6. C. Leitten, Jr., Private communication to R. J. Beaver.

of materials for the second APPR-1 core loading, which were made before the results of the activation measurements, had proposed the use of 347 stainless steel rather than 304L stainless steel as was used in the first core.⁷ The recommendation to substitute 347 for 304L stainless steel was made on the basis of metallurgical considerations.⁸ Following the revelation of the importance of long-lived activation products from stainless steel, Alco Products⁷ pointed out that 347 stainless steel would add still another important long-lived activation product through the impurity Ta. In a desire to reduce the buildup of long-lived activation products, it was therefore decided to fabricate the second APPR-1 core from low cobalt (0.005 wt. %) 304L stainless steel. Thus the production of Co^{60} will be reduced by a factor of ten but the production of Co^{58} activity will remain the same. If the continued buildup of Co^{58} activity should prove troublesome from the standpoint of maintenance of the APPR power plant, then serious thought will have to be given to the elimination of Ni from future APPR cores.

The following is an estimate of the ratio of Ta^{182} to Co^{60} activities expected from the presence of Ta and Co in the core and substantiates the contention of Alco Products. The calculation refers to the buildup of activity in the core cladding by activation of materials in the cladding and does not refer to the buildup of activity in the water, demineralizer, out-of-pile piping, etc. As yet, where and how activation products are deposited is not completely understood.⁹

7. R. C. Woodbury, "Consolidation of Efforts Concerning APPR-1 Core II," Minutes of Oct. 24, 1957 meeting at Washington, D. C., Nov. 8, 1957.
8. R. J. Beaver et al, "Investigation of the Factors Affecting Sensitization of APPR-1 Fuel Elements," ORNL-2312, Sept. 18, 1957.
9. A. L. Medin et al, "Literature Survey for Activity Build-Up on Reactor Primary System Components," APAE-25, Jan. 15, 1958.

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The buildup of Co^{60} atoms in a core will follow the schematic equation:

Rate of Co^{60} atom density buildup = Production of Co^{60} atoms
per cm^3 per sec by thermal
neutron absorption in Co^{59} .

Production of Co^{60} atoms
+ per cm^3 per sec by fast
neutron absorption in Co^{59} . - Loss of Co^{60} atoms per
 cm^3 per sec by radioactive
decay.

Loss of Co^{60} atoms per
- cm^3 per sec by thermal
neutron absorption in Co^{60} - Loss of Co^{60} atoms per
 cm^3 per sec by fast neutron
absorption in Co^{60} .

which may be written in symbolic form:

$$\frac{dN^{60}(t)}{dt} = \sum_{a2}^{59} \phi_2 + \sum_{a1}^{59} \phi_1 - \lambda^{60} N^{60}(t) - \phi_2 \sigma_{a2}^{60} N^{60}(t) - \phi_1 \sigma_{a1}^{60} N^{60}(t) \quad (1)$$

Assuming that the thermal flux, ϕ_2 , the fast flux, ϕ_1 , and the Co^{59} atom density are not functions of time, the solution of Eq. (1) is:

$$N^{60}(t) = \frac{\sum_{a2}^{59} \phi_2 + \sum_{a1}^{59} \phi_1}{\lambda^{60} + \phi_2 \sigma_{a2}^{60} + \phi_1 \sigma_{a1}^{60}} \left[1 - e^{-(\lambda^{60} + \phi_2 \sigma_{a2}^{60} + \phi_1 \sigma_{a1}^{60})t} \right] \quad (2)$$

A similar equation will apply to the atom density of Ta^{182} :

$$N^{182}(t) = \frac{\sum_{a2}^{181} \phi_2 + \sum_{a1}^{181} \phi_1}{\lambda^{182} + \phi_2 \sigma_{a2}^{182} + \phi_1 \sigma_{a1}^{182}} \left[1 - e^{-(\lambda^{182} + \phi_2 \sigma_{a2}^{182} + \phi_1 \sigma_{a1}^{182})t} \right] \quad (3)$$

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Since there is not any information about the fast absorption cross section of Co^{60} and Ta^{182} , the terms involving σ_{a1}^{60} and σ_{a1}^{182} will be ignored in Eqs. (2) and (3). The decay rate of Co^{60} is given by $\lambda^{60} N^{60}(t)$ and the decay rate of Ta^{182} is given by $\lambda^{182} N^{182}(t)$. For each decay of Co^{60} there are two gamma-rays but only one for Ta^{182} . The ratio of gamma-rays from Ta^{182} to Co^{60} should then be given by:

$$\frac{\gamma\text{'s from Ta}^{182}}{\gamma\text{'s from Co}^{60}} = \frac{\lambda^{182} (\sum_{a2}^{181} \phi_2 + \sum_{a1}^{181} \phi_1)}{2 \lambda^{60} (\sum_{a2}^{59} \phi_2 + \sum_{a1}^{59} \phi_1)} \times \frac{(\lambda^{60} + \phi_2 \sigma_{a2}^{60})}{(\lambda^{182} + \phi_2 \sigma_{a2}^{182})} \frac{[1 - e^{-(\lambda^{60} + \phi_2 \sigma_{a2}^{60})t}]}{[1 - e^{-(\lambda^{182} + \phi_2 \sigma_{a2}^{182})t}]} \quad (4)$$

The basic data are summarized as follows:¹⁰

$$\begin{aligned} \lambda^{182} &= 7.14 \times 10^{-8} \text{ sec}^{-1} \\ \lambda^{60} &= 4.21 \times 10^{-9} \text{ sec}^{-1} \\ \sigma_{a2}^{181} &= 18.9 \times 10^{-24} \text{ cm}^2 \\ \sigma_{a2}^{59} &= 32.8 \times 10^{-24} \text{ cm}^2 \\ \sigma_{a2}^{182} &= 20,000 \times 10^{-24} \text{ cm}^2 \\ \sigma_{a2}^{60} &= 6 \times 10^{-24} \text{ cm}^2 \end{aligned}$$

$$\text{Resonance Integral}^{11} \text{ of Ta}^{181} = \text{R.I.}^{181} = 590 \times 10^{-24} \text{ cm}^2$$

$$\text{R.I.}^{59} = 49.3 \times 10^{-24} \text{ cm}^2$$

10. D. J. Hughes and J. A. Harvey, "Neutron Cross Sections," BNL-325, July 1, 1955.
11. R. L. Macklin and H. S. Pomerance, "Resonance Capture Integrals," Progress in Nuclear Energy, Vol. 1, p 179, McGraw-Hill Book Co., 1956.

Thermal cross sections are Maxwellian averages assuming a $1/v$ behavior for the cross sections.

Assuming that all fast absorptions occur in the $1/E$ region of the fast flux, the fast absorption term may be written as:

$$\begin{aligned} \sum_{a1}^i \phi_1 &= N^i \int_{.4 \text{ ev}}^{2 \times 10^6 \text{ ev}} \sigma_a^i(E) \phi_1(E) dE = a_1 N^i \int_{.4}^{2 \times 10^6} \sigma_a^i(E) \frac{dE}{E} \\ &= a_1 N^i R.I.^i \end{aligned} \quad (5)$$

where the fast flux has been assumed to have a $1/E$ behavior and extend from 2×10^6 ev down to 0.4 ev.

$$\phi_1 = \int_{.4}^{2 \times 10^6} \phi(E) dE = a_1 \int_{.4}^{2 \times 10^6} \frac{dE}{E} = 15.40 a_1 \quad (6)$$

The constant a_1 may be determined if the ratio of the fast to thermal fluxes is known. From calculations made by Alco Products¹² this ratio is estimated to be about 7 for the APPR.

$$\frac{\phi_1}{\phi_2} = 7 = \frac{15.4 a_1}{\phi_2}$$

$$\therefore a_1 = .455 \phi_2$$

Assuming the thermal flux for the APPR to be 2×10^{13} neutrons/cm²-sec, then $a_1 = .910 \times 10^{13}$ neutrons/cm²-sec. With this value for a_1 and using Eq. (5) we obtain:

12. F. B. Fairbanks et al, "Predicted Core Performance for the Army Package Power Reactor No. I," APAE-11, Aug. 31, 1956.

$$\sum_{al}^{181} \phi_1 = N^{181} s 5.38 \times 10^{-9} \text{ sec}^{-1}$$

$$\sum_{al}^{59} \phi_1 = N^{59} \times 4.50 \times 10^{-10} \text{ sec}^{-1}$$

Eq. (4) may now be evaluated for any period of reactor operation. For a period of 1.5 years = 4.57×10^7 sec we have:

$$\frac{\gamma's \text{ from Ta}^{182}}{\gamma's \text{ from Co}^{60}} (1.5 \text{ years}) = 2.24 \frac{N^{181}}{N^{59}} = 0.7 \frac{\text{wt. \% Ta}^{181}}{\text{wt. \% Co}^{59}}$$

Thus, this estimate would predict that equal weight percentages of Ta and Co would build up about equal activities in the core after 1.5 years of operation. The use of 347 stainless steel containing a nominal 0.2 wt. % Ta and 0.05 wt. % Co would therefore produce a more serious activation problem than is now being experienced with the present 304L core. A desire to reduce the production of long-lived activities would then require specifying a low tantalum as well as a low cobalt content if 347 stainless steel were used for the second APPR-1 core. Fabrication of a low cobalt 304L core is therefore an attempt to reduce and to simplify the activation problem.

The buildup of gamma-activity in the APPR-1 core predicted by Eqs. 2 and 3 is presented in Fig. 1. The curves are for equal wt. % of Ta¹⁸¹ and Co⁵⁹ in the core.

Gamma-Ray Intensity (Relative)