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THE CALCULATION OF INDUCED GAMMA ACTIVITY
IN COMMERCIAL MATERIALS

by

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MATERIALS TESTING REACTOR PROJECT

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THE CALCULATION OF INDUCED GAMMA ACTIVITY IN COMMERCIAL MATERIALS

Paul J. Persiani

SUMMARY

In many materials it is possible to effect a considerable reduction in pile-induced gamma activity by decreasing the content of one or more impurity. However, there are many factors to consider before setting up preliminary specifications. Some of these factors are the period of irradiation required, the time after irradiation at which the material is to be handled, and the feasibility of decreasing the amount of an impurity. For an example, in a typical sample of extruded Brush beryllium, if the manganese content were reduced by one hundred-fold the induced gamma activity would be decreased proportionately over approximately the first 30 hours after irradiation. However, beyond 30 hours the manganese is ineffective as a contributor to the total activity. If a reduction in activity is desired beyond 30 hours then the content of cobalt, zinc, iron, and chromium, inasmuch as they are the chief contributors, must be reduced.

INTRODUCTION

In handling and storing irradiated component parts of the reactor, such as plugs, removable assemblies, and structural members, problems arise that require a thorough knowledge of the radioactivity induced in the materials involved. An indication as to the elements present in a material, either as alloys or impurities, responsible for the activity, and the type of radiation emitted by the activity is necessary if an efficient design of shielding and storing facilities is to result.

The purpose of this report is to provide gamma activity data for design information of some commercial materials that may be used in the Materials Testing Reactor. A summation of the activities of the more significant elements present in each material is made on a log-log plot. The plots serve to indicate the contribution of the "impurities" to the total activity of a material at a particular time of decay.

METHOD OF CALCULATION

The calculations, wherever possible, were compared with experimental data and were found to be in good agreement, the errors ranging

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Chief, Declassification Branch

well within a factor of ten. Errors will be analyzed in the discussion for the various materials.

The accuracy of the calculations is mainly dependent upon two factors: (1) the qualitative and quantitative analysis of the impurities present in the material; (2) the analysis of the reactions and radiation emitted.

The degree of accuracy required in determining the "impurities" and their amounts present is largely dependent upon the neutron flux in which the material is to be irradiated. Impurities present in micrograms per gram of material come into effect appreciably when irradiated in a high flux pile. If the quantitative analysis of an impurity, which is one of many major contributors to the total induced activity, is in error, the corresponding error in the total intensity is of a lower magnitude.

The spectrochemical analyses were obtained from manufacturers, project literature, and other laboratories. In cases where the information was not available or believed to be inadequate, samples were procured and analyzed by the Metallurgical Division of ANL. In analyzing the reactions and radiations emitted, the energies of radiation and patterns of decay were obtained from the "Table of Isotopes" by Seaborg and Perlman and MonP-405.

The reactions of cobalt and manganese will be used as sample analyses. Cobalt 59 produces an isomeric state of cobalt 60 in an isomeric ratio of 30/0.73 (ratio of activation cross sections). The isotope produced with the greater cross section emits a 0.31-mev beta followed by two photons in a cascading pattern of 1.1 mev and 1.3 mev with a half life of 5.3 years. The other isomer decays by two processes; 90% consists of a 1.3 mev beta followed by a 1.452-mev photon with a half life of 10.7 min., the remaining 10% transforms into the 5.3-year isomer following an emission of a 0.056-mev photon. The decay scheme is represented by the following diagram.

(A) $\text{Co}^{59} \sigma_a = .73 \text{ barns}$

(B) $\text{Co}^{59} \sigma_a = 30 \text{ barns}$

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The amount of isotope A produced is relatively small as compared to isotope B, and its contribution to the activity of Co^{60} after about an hour of cooling is negligible when the period of irradiation is long ($\sim 1/10$ of 5.3 years); however, for short periods of irradiation, when isotope A is just saturated, the activity of isotope B is negligible. The initial intensity will be predominantly that of the 1.452-mev gamma with 5.3-years half life increasingly coming into effect. In view of the above decay scheme and the period of irradiation in which we are interested (one year) the production of isotope A was neglected.

A simple method of treating the radiation emitted by isotope B would be to consider an emission of two 1.2-mev gammas. Due to the varied ranges of energy emitted and patterns of decay throughout the elements under consideration a consistent system of analysis is desirable. Thus isotope B was treated as two separate isotopes, one emitting a 1.3-mev gamma and the other a 1.1-mev gamma.

In the case of manganese 56 the energies of the photons varied in a wide range and the decay pattern is of a cascading and parallel form (see diagram below).

The isotope was treated as three separate isotopes, one emitting 100% of 0.845 mev, another emitting 25% of 1.81 mev, and the third emitting 15% of 2.13 mev.

The decay patterns and intensities of some isotopes were more complex and difficult to investigate. In such cases a decay scheme was assumed in such a way as to introduce the least error.

The neutron capture cross sections for most elements are known with an accuracy within a range of from $\sim \pm 4\%$ to ± 60 or $\pm 70\%$. Thus an error in cross section by, say, 100% will vary the resultant activity of the isotope by a factor of 2. This is well within the accuracy of a spectrochemical analysis which may vary one hundred-fold for some elements in the material.

An indication as to the impurities expected to contribute a major portion of the activity for a particular period of irradiation and decay may be obtained by the following expression

$$\sum a_i \times f \times \left(1 - \frac{1}{2^n}\right) \left(\frac{1}{2^m}\right) \times \bar{E}$$

where

$\sum a_i$ = macroscopic capture cross section for isotope (i) per gram of normal element

f = amount of element present in material in ppm

n = irradiation time (no. of half-life periods)

m = time subsequent to irradiation (no. of half-life periods)

$\bar{E}$ = weighted average energy

In general for a short period of irradiation, isotopes with a short half life will dominate the activity. For a long period of irradiation the degree of activity will increase due to long half-life isotopes and the total effect of the resultant curve is a decrease in the rate of decay.

The activity was determined by the following formula:

$$N_0 = \lambda N = \Phi \times \sum a_i \times \rho \times f (1 - e^{-\lambda_i t_1}) e^{-\lambda_i t_2}$$

where

N_0 = no. of disintegrations per sec. per cc = λN

Φ = neutron flux (neutrons/cm² sec.)

ρ = density of material

λ_i = decay constant

t_1 = irradiation period

t_2 = time subsequent to irradiation

In high neutron fluxes there is a possibility that the products are further converted into isotopes that have a very high neutron capture cross section. Neutron reactions of several orders usually prevail among the heavier isotopes. The contribution to the total activity of second order reactions is found however to be negligible. The second order reaction for

the europium isotope 153 will be used to illustrate the point. The thermal neutron capture cross section for the reactions are:

$$\text{Eu}^{153} (n, \gamma) \text{Eu}^{154} \quad \sigma_{153} = 240 \text{ barns}$$

$$\text{Eu}^{154} (n, \gamma) \text{Eu}^{155} \quad \sigma_{154} = 880 \text{ barns}$$

$$\text{Eu}^{155} (n, \gamma) \text{Eu}^{156} \quad \sigma_{155} = 7900 \text{ barns}$$

The concentration of Eu^{154} at any time, t , during irradiation is obtained as follows:

$$a) \text{ production of } \text{Eu}^{154} = \phi \times \Sigma_{a153} \times \rho \times f = K \text{ (constant)}$$

$$b) \text{ loss of } \text{Eu}^{154} \text{ due to natural decay} = \lambda_{154} N_{154}$$

$$c) \text{ loss of } \text{Eu}^{154} \text{ due to neutron capture} = \phi \sigma_{154} N_{154}$$

The rate of production of Eu^{154} is then

$$1) \frac{dN_{154}}{dt} = K - (\lambda_{154} + \phi \sigma_{154}) N_{154}$$

The expression $(\lambda_{154} + \phi \sigma_{154})$ includes the burning out of the isotope and may be referred to as the effective decay constant represented by the symbol λ_e^{154} .

Equation 1) reduces to

$$2) N_{154}(t) = \frac{K(1 - e^{-\lambda_e^{154} t})}{\lambda_e^{154}}$$

where $N_{154}(t)$ is the concentration of Eu^{154} for an irradiation time, t .

Now the rate of production of Eu^{155} is

$$3) \frac{dN_{155}}{dt} = \phi N_{154}(t) \sigma_{154} - (\lambda_{155} + \phi \sigma_{155}) N_{155}$$

where $\phi N_{154}(t) \sigma_{154} = \text{production of } \text{Eu}^{155}$

As above, $(\lambda_{155} + \phi \sigma_{155}) = \lambda_e^{155} = \text{effective decay constant for } \text{Eu}^{155}$.

We then have, by substituting equation 2) into equation 3)

$$4) \frac{dN_{155}}{dt} = \frac{\phi \sigma_{154} K (1 - e^{-\lambda_e^{154} t})}{\lambda_e^{154}} - \lambda_e^{155} N_{155}$$

equation 4) reduces to

$$5) N_{155}(t) = \frac{K \phi \sigma_{154}}{\lambda_e^{154}} \left[\lambda_e^{155} (1 - e^{-\lambda_e^{154} t}) + \lambda_e^{154} (e^{-\lambda_e^{154} t} - e^{-\lambda_e^{155} t}) \right]$$

From formulas 2) and 5) and the conditions of operation assumed in the preceding section, the concentration of Eu^{155} is lower than that of Eu^{154} by a factor of $\sim 10^{-9}$.

In converting from activity to equivalent biological effect, the geometry of the source must be considered. By definition of a roentgen, it is necessary to determine the number of incident photons striking a unit area at the surface of a receptor. The intensity obtained for various source geometries and positions of the receptor have been derived and are included in Appendix I.

If we let I_0 be the number of incident photons striking a unit area then the equivalent biological effect expressed in roentgens per hour is as follows,

$$I = I_0 \times E \times 10^6 \times \mu_a \times 1/32 \times 4.8 \times 10^{-10} \times 3.6 \times 10^3$$

where

I = roentgens per hour

I_0 = number of photons per cm^2 per sec.

E = energy of the photons in ev

* μ_a = absorption coefficient of ionizing photons in air

$1/32$ = number of ion pairs produced per electron volt

4.8×10^{-10} = esu per charge

3.6×10^3 = seconds per hour

A plot of I_0 (number of photons per cm^2 per sec.) required to produce one roentgen per hour for various photon energies is included in Appendix II (Figure 17).

* See Appendix II (Figure 16)

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The intensity at the surface of a one cubic centimeter sphere was calculated for commercial grades of various materials. The calculations were based on a thermal neutron flux of 10^{14} neutrons/cm² sec. and an irradiation period of 300 days. To utilize the curves for conditions other than the above, the following relation is set up.

$$D_T = \sum D_i$$

$$D_i = D_{i_{t_1}} \times \frac{I_2}{I_1} \times V \times \frac{\Phi_2}{\Phi_1} \times \left[\frac{(1 - e^{-\lambda_i t_2})}{(1 - e^{-\lambda_i t_1})} \right]$$

D_T = total intensity in roentgens per hour

D_i = intensity (r/hr) of isotope (i) for new condition

$D_{i_{t_1}}$ = intensity (r/hr) for isotope (i) at decay time (t_1)

I_2 = (I_0) number of photons per cm² per sec at surface of receptor expressed in N_T

$I_1 = \frac{N_T R}{2} = 0.31 N_T$ where R = radius of a 1 cc sphere

N_T = total source strength

V = volume of material irradiated

t_2 = new time of irradiation

DISCUSSION

1. Beryllium (Figures 1, 2, 3, 4)

The analyses for Brush Hot Pressed Beryllium were obtained from Mr. Greggs' report on corrosion tests (ORNL 49-4-144). The maximum values were used. The analyses of all materials referred to in this report are included on the graphs.

It is interesting to note that the intensity and decay of the two lots of powdered beryllium for the first 1,000 hours are approximately the same although they contain only three common impurities, Mn⁵⁶, Zn⁶⁵, and Fe⁵⁹. The combination of Na²⁴, Cr⁵¹, and Ag¹¹⁰ seems to simulate that portion of the intensity in lots 3 and 4 due to Co⁶⁰ and Cu⁶⁴.

The analysis for the extruded beryllium was obtained from Charles Egger. It is a compilation of analyses run on 20-30 different specimens. In a general comparison the extruded beryllium has a

Fig. 1

Fig. 2

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Fig. 3

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Fig. 4

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higher induced activity than the hot pressed beryllium. Some impurities are common in all three specimens.

In shielding design the cooling period as well as the grade of material becomes a factor to consider, e.g. the thickness of shield required for the extruded beryllium after 1,000 hours of cooling is approximately two half-value layers greater than that for the powdered beryllium; whereas the variation in shield thickness is relatively negligible after a day of cooling.

The only data available at present on the activity of beryllium is that of Metcalf as reported in Shielding Calculations (AMW CL-2-080447). A 1.9 gm sample of beryllium was irradiated in a slow neutron flux of 9×10^{12} for a period of 23 days. The intensity measured at 15.3 cm after 8 days of cooling was 1.6 mr/hour. The analysis and grade of beryllium used were not given; however, upon inquiry it was found that the sample was an extruded grade of beryllium approximating the analysis upon which the calculations were based. Assuming that a 1.9-cm specimen acts as a point source at a distance of 15.3 cm and correcting for the experimental conditions ($D_T = \sum D_i$), the calculated intensity is 1.4 mr/hour. The values are in close agreement.

2. Aluminum (Figures 5, 6, 7, 8)

The analyses of the three grades of aluminum were obtained from the Aluminum Company of America by the Metallurgical Division.

The manganese content in the 2S aluminum is twice that of the 3A grade and after one hour cooling the activity of 2S is greater by a factor of 2 than that of the 3A grade. This factor is gradually reduced within the next two hours. The activity thereafter is approximately the same for the two grades. Gallium, sodium, and zinc are present in the two grades in equal amounts. Hoopes refined aluminum is free of gallium and zinc, and the manganese content is much lower than the 2S and 3A grade. The resulting activity is therefore much lower.

A sample of 2S aluminum was exposed 71 days in a neutron flux of $\sim 10^{12}$ neutrons per cm^2 sec., and measurements were taken at a distance of one foot for a period of 70 days. Calculated and experimental results are compared in the following table.

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Fig. 6

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Fig. 8

19 TABLE I
Induced Activities in Aluminum

<u>Days after Removal</u>	<u>Activity in mr/hr gm</u>	
	<u>Exp.</u>	<u>Calc.</u>
2	10	7.7
4	2	4.8
6	0.9	3.2
8	0.56	2.2
10	0.46	1.75
20	0.28	0.61
30	0.21	0.38
50	0.136	0.174
70	0.1	0.078

The computations are in good agreement with the experimental data. Thus the 4-hour to 20-hour decay period values vary as much as by a factor of 4. The spectrochemical analysis of the sample used was not available. Speculatively, probably the gallium or sodium content was not as high as that upon which the calculations were made.

3. Magnesium (Figures 9, 10, 11, 12)

The Domal High Purity Magnesium is a product of the Dominion Magnesium Limited of Canada. The high purity magnesium is the result of essentially a distillation process carried out under a vacuum; the vapor is condensed directly to a solid under controlled conditions. The total iron, copper and nickel content is guaranteed not to exceed 0.005 per cent.

The remaining two grades are Dowmetal products. The magnesium designated 58135 is an Oak Ridge specification. The 58135 magnesium appears to be a highly purified form of the FS#1 grade. If the zinc, cobalt, and sodium content were further reduced, then its activity would approach that of the Domal - approximately in the same order of magnitude.

The analyses were obtained from the manufacturers. Experimental data on the activity of magnesium is not available.

4. Graphite (Figures 13, 14, 15)

The analyses of AGOT K III and KC-GBF grades of graphite were obtained from L. Fuchs and J. R. Gilbreath. The two analyses did not specify the content of rare earth elements which were found to be present in the unpurified graphite (Report HW-12780). The concentrations were

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Fig. 9

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Fig. 10

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Fig. 11

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Fig. 12

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Fig. 13

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Fig. 14

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Fig. 15

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considered to fall within the range of 0.1 to 0.3 ppm. Gadolinium and samarium have very low activation cross sections; however, Eu^{152} and Eu^{154} add considerably to the activity. In view of this, an assumed 0.2 ppm of europium was included in the analysis of the AGOT K III (unpurified) grade of graphite. The report further stated that the concentration of europium and samarium in the best quality purified graphite was reduced a 100-fold.

Experiments are being conducted at Oak Ridge National Laboratory to determine the gamma activity induced in CS and GBF graphite. The program calls for samples to be irradiated for periods of two weeks, four weeks, two months, and four months. Gamma activity data have been accumulated for the two- and four-week radiation tests, and the decay has been followed for 900 hours. These data are included in ORNL-529. The periods of irradiation thus far investigated are short. Long half-life elements such as europium and cobalt (~ 5.3 years) are not sufficiently activated to influence the total activity of the graphite. A one-year irradiation will yield $\sim 12\%$ of saturation, and this amount greatly influences the activity of the CS graphite.

The experiments indicated elements present in the CS and GBF graphite having 15-hour, 33-hour, 50-day, and >100 -day half-lives.

The experimentalist suspects the elements corresponding to these half-lives are Cu^{64} for the 15-hour, Br^{82} , La^{140} , or As^{76} for the 33-hour, and Fe^{59} for the 50-day half-life. The calculations would indicate Na^{24} for the 15-hour, As^{76} for the 33-hour, and Zn^{65} , Hg^{203} , Cr^{51} or Fe^{59} for the 50-day half-life. The 100-day half-life may be attributed to zinc. It may also be a combination of isotopes that simulate a half-life decay of 100 days. However, it is somewhat difficult to analyze a decay curve that is nearly horizontal.

CONCLUSION

The results presented in this study mainly serve to emphasize the degree of pile-induced gamma activity contributed by the impurities in several materials and the reduction in activity that may be obtained if certain impurities were decreased or completely eliminated. The information may be used as a preliminary basis for metallurgical specifications.

Further, it should be noted that if the activity dictates the choice of material then the absence of certain isotopes must be definitely established.

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APPENDIX I

The formulas to obtain the number of incident gammas per cm^2 per sec. are derived from various geometries and positions. For a more extensive treatment refer to:

1. TNX-7 - C. W. J. Wende
2. Acta Radiologica - Sivert
3. ORNL CF-48-10-44 - Shielding Symposium

1. Point source

$$I_0 = \frac{N_T}{4\pi r^2}$$

where N_T = Total number of photons emitted per sec.

2. Line source viewed broadwise.

let N_L = source strength per unit length $\frac{N_T}{L}$

a = distance to observer

then

$$I_0 = \int_{x_1}^{x_2} \frac{N_L}{4\pi r^2} dx, \quad r^2 = a^2 + x^2$$

$$I_0 = \frac{N_L}{4\pi a} \left(\tan^{-1} \frac{x_2}{a} - \tan^{-1} \frac{x_1}{a} \right)$$

$$I_0 = \frac{N_L}{4\pi a} (\theta_1 - \theta_2)$$

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3. Line source viewed endwise.

$$I = \int_{x_1}^{x_2} \frac{N_L}{4\pi x^2} dx$$

$$I = \frac{N_L}{4\pi} \left[-\frac{1}{x} \right]_{x_1}^{x_2}$$

$$I_0 = \frac{N_L}{4\pi} \left[\frac{1}{x_1} - \frac{1}{x_2} \right]$$

4. Disc source viewed from a point on a normal to its center.

R = radius of disc

a = distance from receptor

let N_A = source strength per
unit area $\frac{N_T}{\pi R^2}$

elemental area = $\rho d\theta d\phi$

$$\rho = a \tan \phi$$

$$d\rho = a \sec^2 \phi d\phi$$

$$r^2 = \frac{a^2}{\cos^2 \phi} = a^2 \sec^2 \phi$$

$$I_0 = \int_0^{2\pi} \int_0^{\tan^{-1} \frac{R}{a}} \frac{N_A}{4\pi a^2 \sec^2 \phi} \cdot a^2 \tan \phi \sec^2 \phi d\phi d\theta$$

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$$I_0 = \frac{N_v R}{2}$$

6. Spherical source with self-absorption receptor at surface.

$$I_0 = \int_v \frac{N_v}{4\pi r^2} e^{-\mu r} dv$$

$$I_0 = \int_0^{2R} \int_0^{\cos^{-1} \frac{r}{2R}} \int_0^{2\pi} \frac{N_v}{4\pi r^2} e^{-\mu r} r^2 dr \sin \theta d\theta d\phi$$

$$I_0 = \frac{N_v}{2} \int_0^{2R} \int_0^{\cos^{-1} \frac{r}{2R}} e^{-\mu r} dr \sin \theta d\theta$$

$$I_0 = \frac{N_v}{2} \int_0^{2R} \left[-\cos \theta \right]_0^{\cos^{-1} \frac{r}{2R}} e^{-\mu r} dr$$

$$I_0 = \frac{N_v}{2} \int_0^{2R} \left(1 - \frac{r}{2R} \right) e^{-\mu r} dr$$

$$I_0 = \frac{N_v}{2} \left[\int_0^{2R} e^{-\mu r} dr - \frac{1}{2R} \int_0^{2R} e^{-\mu r} r dr \right]$$

$$I_0 = \frac{N_v}{2} \left[\left(-\frac{e^{-\mu r}}{\mu} \right)_0^{2R} - \frac{e^{-\mu r}}{2R\mu^2} \left(-\mu r - 1 \right)_0^{2R} \right]$$

$$I_0 = \frac{N_v}{4\mu^2 R} \left[2\mu R + e^{-2\mu R} - 1 \right]$$

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APPENDIX II

That portion of radiation traversing the tissue without loss of energy produces no biological effect. The absorption coefficient for air, to determine equivalent dosage, is therefore comprised of the Compton absorption (electron recoil) process, photoelectric effect and pair production. A plot of τ , σ_a and σ_p was obtained from the Caldwell Lectures by G. Failla, reprinted from the American Journal of Roentgenology and Radium Therapy. The values of σ_a represented by this graph were derived from the Klein-Nishina formula and the value of τ and σ_p were obtained from other published tables and literature.

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Fig. 16

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Fig. 17

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