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DISCUSSION OF AM-CM ALPHA CORRECTION

INTRODUCTION

Americium and curium isotopes which decay by alpha emission hamper the otherwise simple determination of plutonium content at certain points in the separations process. This study of the production equations for americium and curium was made to determine the practicality of calculating the buildup of alpha background activity in the irradiated metal in lieu of more laborious chemical analyses.

SUMMARY

Accurate calculations of the buildup of americium and curium alpha activity do not appear to be practical at the present time because the useful application would necessitate the segregation of all metal on the basis of length of irradiation and integrated exposure. This "special pickup" would be prohibitively costly if done only for this purpose in the reactor areas at the present time.

For a given type of uranium fuel element in a given reactor, the ratio of americium and curium alpha activity to plutonium alpha activity is to a first approximation given by:

$$R = k_1 M^2 \left[\frac{D}{4} + t \right] + k_2 M D e^{-\lambda_{cm} t}$$

where M is the metal exposure in megawatt days per ton, D is the number of days

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SUMMARY (continued)

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the metal was irradiated, t is the number of days since the metal was discharged, and λ_{cm} is the decay constant for curium 242 in reciprocal days. The factors k_1 and k_2 include decay constants, flux to power ratios, conversion efficiencies, etc., all of which are not necessarily the same for all reactors and fuel elements. More exact relations can be derived; these would include other order terms in the exposure, M .

For low exposure, high specific power metal the amount of americium and curium present is small and the possible error introduced into the determination of the plutonium content is insignificant. For high exposure, low specific power metal the alpha activity due to americium and curium becomes significant and must be considered if the plutonium content is to be determined accurately. For example, a factor of two uncertainty in the americium-curium correction would cause approximately 0.5% uncertainty in the plutonium content at about 300 MWD/T but would cause approximately 5% uncertainty at about 800 MWD/T.

DISCUSSION

Americium, curium, and other transuranic elements are formed during the irradiation of uranium in a reactor. These elements result from successive neutron absorptions and radioactive decay of intermediate nuclei. The abundance of any given isotope as a function of exposure in the reactor can be quite complicated if several paths of formation exist. Because of half lives and cross sections one predominant formation path usually exists which can be used to approximate closely the production of each isotope. The most abundant alpha decaying isotopes (other than plutonium) which are formed during the irradiation of U-238 are Am 241 and Cm 242. Am 241 is formed from U-238 by the absorption of three neutrons plus three beta decay processes (predominantly in the order $n, \beta, \beta, n, n, \beta$). The formation of Cm 242 requires an additional neutron absorption and beta decay process.

1. Plutonium Isotope Production

Plutonium production and the isotopic composition of the product as a function of exposure have been studied extensively. For low exposures (less than about 1500 MWD/T) it is possible to describe the buildup of each plutonium isotope with the relations:

$$(g/t) \text{ of Pu 238} = a_1 M^2 + a_2 M^3 + a_3 M^4 + \dots$$

$$(g/t) \text{ of Pu 239} = b_1 M + b_2 M^2 + b_3 M^3 + \dots$$

$$(g/t) \text{ of Pu 240} = c_1 M^2 + c_2 M^3 + c_3 M^4 + \dots$$

$$(g/t) \text{ of Pu 241} = d_1 M^3 + d_2 M^4 + d_3 M^5 + \dots$$

where M is the metal exposure in MWD/T and the coefficients are constants for the particular type of metal, reactor and operating condition under consideration. By weighting each of the isotopes properly the alpha activity of plutonium as a function of exposure is given by:

$$A (\text{activity/standard sample}) = a M + b M^2 + c M^3 + \dots$$

where the coefficients are the sum of the appropriate coefficients above weighted by the proper decay factor. Because of the long half lives of the plutonium isotopes involved and the short half lives of the parent nuclei, the isotopic composition is independent of irradiation or decay time for most practical applications.

DECLASSIFIEDDISCUSSION (Continued)2. Americium 241 Formation and Decay

An approximate expression for the quantity of Am 241 existing in irradiated uranium is:¹

$$X = \lambda_{41} d_1 M^3 \left(\frac{D}{4} + t \right)$$

where X = grams of Am 241 per ton of metal.

λ_{41} = decay constant of Pu 241 (days⁻¹).

d_1 = g/T per (MWD/T)³, conversion relation for Pu 241 (see above).

D = days of metal irradiation.

t = days of decay (since discharged).

Americium 241 is formed from the beta decay of Pu 241 and consequently is closely related to the production of this isotope. During the irradiation time the quantity of Pu 241 increases from zero to the final value. Concurrently the Pu 241 is decaying to form Am 241. For irradiation times very short compared to the half life of Pu 241 (13 years) the loss of Pu 241 due to decay can be neglected. This quantity, however small, is the Am 241 produced in the reactor. After the metal is discharged the Pu 241 continues to decay to form Am 241. For times very short compared to the Pu 241 half life, the loss of Pu 241 can be neglected even though this quantity is an important consideration in Am 241 production. A more accurate description of Am 241 production would include higher order terms (in M). The alpha activity due to Am 241 can be obtained by applying the proper decay constant to the above relation.

3. Curium 242 Formation and Decay

An approximate relation for the quantity of Cm 242 existing in irradiated uranium is:

$$Y = \lambda_{41} d_1 M^3 (k MD) e^{-\lambda_{cm} t}$$

where Y = grams of Cm 242 per ton of metal.

k = constant for reactor and metal type, tons per MWD. (This term includes cross sections, flux to power ratios, and numerical factors).

λ_{cm} = decay constant for cm 242, days⁻¹.

Curium 242 is formed by the absorption of a neutron in Am 241 followed by the beta decay of the Am 242 nucleus. In most cases the Am 242 beta decays with a 16 hour half life and the production of Cm 242 can be considered to be instantaneous with the production of Am 242. An isomeric state (with a 100 year half life) has been neglected because of the relatively small formation cross section. After the metal is discharged from the reactor the Cm 242 continues to decay exponentially with a 163 day half life. A more accurate description of Cm 242 content would include higher order terms in M. The alpha activity due to Cm 242 can be obtained by applying the appropriate decay constant to the above relation.

1. Personal Communication, R. O. Gumprecht.

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DISCUSSION (Continued)

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4. Ratio of Am-Cm to Pu Alpha Activity

The ratio of Am-Cm alpha counts to those due to Pu can be obtained by combining the relations presented above. The ratio becomes:

$$R = k_1 M^2 \left[\frac{D}{4} + t + k_2 MD e^{-\lambda_{cm} t} \right]$$

where R = ratio of Am Cm alpha counts to Pu alpha counts and k_1 and k_2 are constants for a particular metal, reactor operating condition, and alpha counting arrangement. Other order terms in M have been omitted; these could be included as necessary to increase the accuracy of the calculated ratio.

The ratio is seen to be a function of three variables: metal exposure, irradiation time and decay time. Any attempt to fit laboratory determinations of the ratio simply to exposure would be unsatisfactory unless all metal were irradiated for the same length of time and were permitted to cool for the same length of time. In practice this is not done; peripheral metal is irradiated approximately three times as long as central metal. If each key of metal contained a constant and uniform blend of metal, a simple exposure relation might work. In practice side fringe metal tends to be separated causing gross non-uniformity in the keys. Individual keys of metal which contain an unusual quantity of fringe (or central) metal will have grossly different americium and curium content than an "average" key and individual dissolver batches will vary even more. This probable difference between keys of the same weighted exposure explains why laboratory results are frequently in disagreement with the alpha activity ratio calculated from the formula:²

$$R = 1.45 \times 10^{-9} M^{2.64}$$

5. Numerical Examples

From the power distribution in a reactor the average residence time to obtain a given exposure in the various zones can be estimated. Assuming that all metal in the reactor is irradiated to an exposure of 575 MWD/T, each 100 T can be broken down into four "average" groups as follows: 65 T will have to be irradiated for approximately 82 days, 20 T for 105 days, 10 T for 150 days and 5 T for 205 days. Since this is "average" metal, the ratio, R, would be given by the power dependent relation,

$$R = 1.45 \times 10^{-9} (575)^{2.64} = 2.81 \times 10^{-2},$$

and the constants for the time dependent relation would be:

$$R = 1.42 \times 10^{-10} M^2 \left[\frac{D}{4} + t + 0.0126 MD e^{-0.00425 t} \right].$$

If 100 T of central (82 day) metal were processed, the R factor calculated by the latter equation would be 2.39×10^{-2} . If 100 T of extreme fringe metal were processed, the calculated R factor would be 5.28×10^{-2} . In actual practice the R factor for individual dissolver batches might be expected to be more varied than this. This is true because the fringe metal actually is

2. HW-41274, "Americium-Curium Correction", W. H. Reas, February 2, 1956.

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DISCUSSION (Continued)

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5. Numerical Examples (cont.)

irradiated to a higher goal than the central metal and the exposures are averaged for each key. A hypothetical 575 MWD/T key might contain half fringe metal with an exposure of 900 MWD/T and half central metal with an exposure of 250 MWD/T. In this case the spread of the ratio would be from 0.00162 (for the 250 MWD/T central metal) to 0.294 (for the 900 MWD/T fringe metal). Using the power dependent relation the alpha activity would be adjusted by about 2 1/2% to account for Am-Cm activity; the adjustment should be about 30% for half of the metal and nothing for the other half. The plutonium content for the material would be miscalculated by 10-15%.

6. Conclusions

The am-cm ratio is small for low exposure, high power metal but becomes large for high exposure, low power metal. As goal exposures are increased the ratio must be determined more and more accurately to maintain the same accuracy in the determination of the plutonium content by the method of alpha counting. Even though it is theoretically possible to calculate the ratio for each piece of metal, the identity of the individual piece is lost and the calculation is of no value. It might be possible to develop a numerical biasing technique to determine a useful average value of the ratio for an entire key of metal. If an applicable biasing technique can not be found, it appears impractical to calculate the americium and curium buildup. In such a case the laboratory analysis of each dissolver batch appears to be the most practical method of obtaining the quantity of americium and curium present. If the laboratory analyses are not as accurate as needed for accountability purposes, other physical or chemical methods of determining the plutonium content of the dissolved metal might need to be investigated.

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