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Production of Minor Actinides in the Commercial Fuel Cycle

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I. Introduction

The minor actinides are an inevitable by-product of the production of electricity with thermal reactors. This paper will review the mechanisms of production of the principal minor actinides (^{237}Np, ^{241}Am, and ^{243}Am) and their expected concentrations in spent fuel from commercial power reactors as well as in Pu produced from reprocessing these types of spent fuels. All the results shown are derived from calculations done using the ORIGEN2 reactor code.[1]

II. Reactor Production

Neptunium-237 is produced by the relatively rapid (6.75 d half-life) beta decay of ^{237}U. The ^{237}U is generated in a reactor by n,γ reactions either from lighter uranium isotopes (^{235}U, ^{236}U) or from ^{238}U by an n,2n process. Once removed from the reactor, the quantities of ^{237} Np remain essentially unchanged with time because of its long half-life (2.14×10^{6} y).[2]

Americium-243 is produced by the very rapid (4.98 h half-life) beta decay of ^{243}Pu, which is itself formed by successive n, γ reaction starting from ^{239}Pu. While not as long lived as ^{237}Np, ^{243}Am (7950 y half-life) quantities still do not change dramatically after discharge.[2]

Americium-241, however, which is produced by the beta decay (13.2 y half-life) of ^{241} Pu is different in that the quantities of this material grow steadily after discharge.[2] The consequences of this situation will be considered further in Sections III and IV.

Figures 1, 2, and 3 show the concentrations of these three minor actinides in fuel from three different common power reactor types: a pressurized water reactor (PWR), a boiling water reactor (BWR), and a CANDU. The concentrations are shown as a function of burnup out to 50,000 MWd/tU for the BWR and PWR burning 3.5% enriched LEU, and out to 10,000 MWd/tU for the CANDU. In each case the relative concentrations of the actinides are $^{237}\mathrm{Np}>^{243}\mathrm{Am}\geq^{241}\mathrm{Am}$. Also, in each case production shows an induction period while concentrations of the precursors build up to their equilibrium values. At higher burnups large quantities of ^{237}Np and ^{243}Am will be present in spent fuel from PWR and BWR reactors at discharge. The lower concentrations of these materials in CANDU reactors is a result of the lower burnups possible with this type of reactor, and in the case of ^{237}Np the lower initial ^{235}U (CANDU reactors use natural uranium fuel).

III. Spent Fuel

As discussed in the last section, once removed from the reactor the concentrations of ^{237}Np and ^{243}Am remain essentially stable. However, the ^{241}Am concentrations will continue to rise as the ^{241}Pu continues to decay. Figures 4, 5, and 6 show the changes in concentration of ^{241}Pu and ^{241}Am with time for fuel produced by PWR, BWR, and CANDU reactors with burnups of 35,000; 35,000; and 7,500 MWd/tU, respectively. These curves show significant changes in the ^{241}Am content with the maximum levels being achieved more then 70 years after discharge. The concentrations begin to fall after this point as the decay of the ^{241}Am (458 y half-life)[2] comes to dominate the process.

IV. Stored Product Pu

In the normal PUREX process, all of the Am and part of the Np follows the fission products to the high-level waste in the first extraction step. Depending on the process details the remainder of the Np will follow either the U or Pu when these materials are separated. The Np is then usually extracted from these products, although small amounts of Np may remain in the Pu product.

However, as the precursor to ^{241}Am is a Pu isotope, it will not be removed by reprocessing, and ^{241}Am will continue to be produced in the product over time. Figures 7 and 8 show the buildup of ^{241}Am in plutonium separated from 35,000 MWd/tU PWR and BWR spent fuel. As in the parent spent fuel, the maximum levels are found more than 70 years after separation and reaches a significant proportion of the actinide present.

V. Conclusion

From the examples shown above, it is clear that substantial quantities of the minor actinides ^{237}Np, ^{241}Am, and ^{243}Am are produced in the normal operation of common power reactors. Further, these materials remain present (and in the case of ^{241}Am grow) in the spent fuel. Even if reprocessed, the product Pu will soon come to produce significant amounts of ^{241}Am due to the decay of ^{241}Pu.

References

1. ORIGEN2, V2.1, Radiation Shielding Information Center, P.O. Box 2008, Oak Ridge, TN 37831-6362, August 1, 1991.

2. M. Benedict, T. H. Pigford, and H. W. Levi, Nuclear Chemical Engineering, 2nd Edition, McGraw-Hill, New York, 1981.

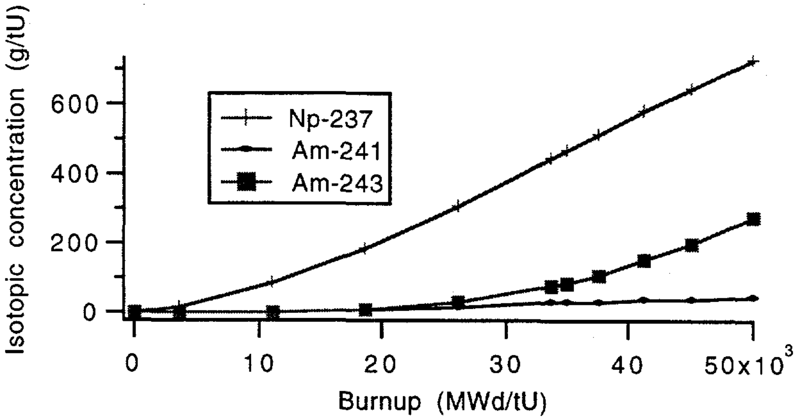

Fig. 1. ^{237}Np, ^{241}Am, and ^{243}Am concentrations at discharge vs. burnup for a PWR.

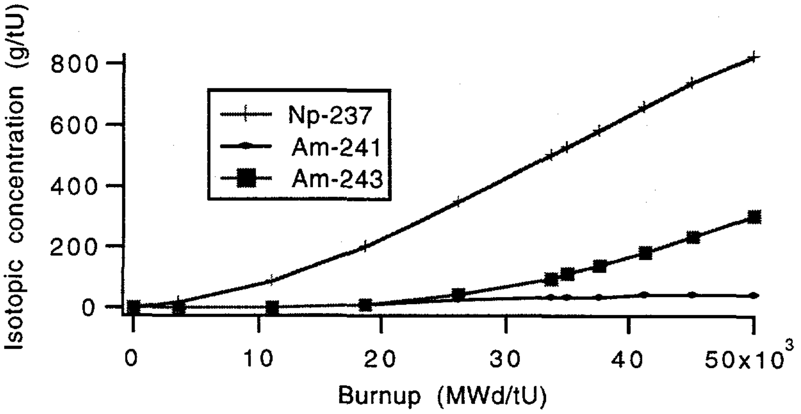

Fig. 2. ^{237}Np, ^{241}Am, and ^{243}Am concentrations at discharge vs. burnup for a BWR.

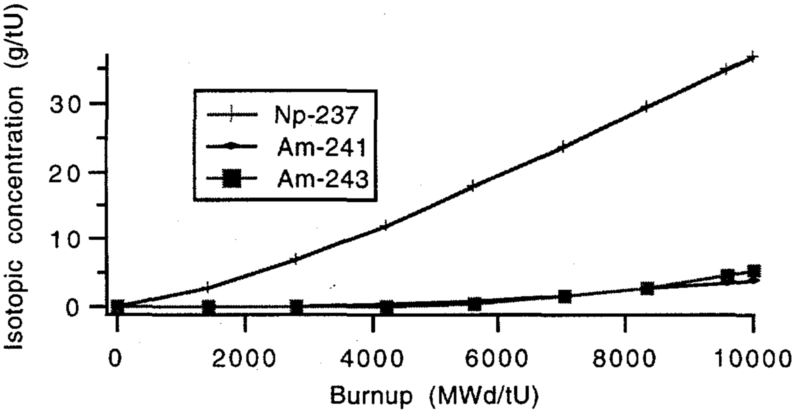

Fig. 3. ^{237}Np, ^{241}Am, and ^{243}Am concentrations at discharge vs. burnup for a CANDU.

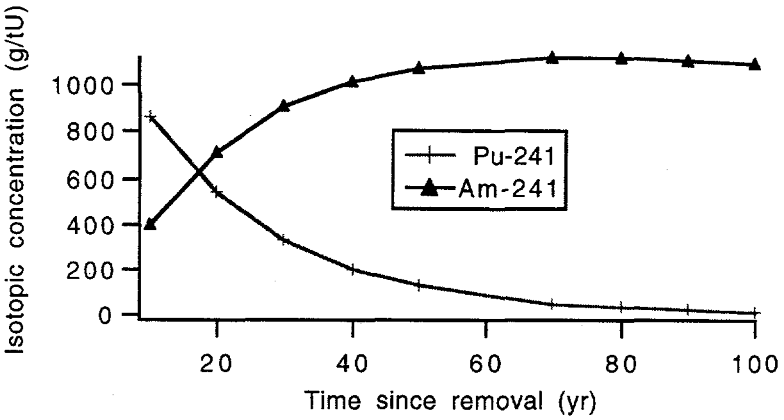

Fig. 4. ^{241}Pu and ^{241}Am concentrations vs. time in 35,000 MWd/tU PWR spent fuel.

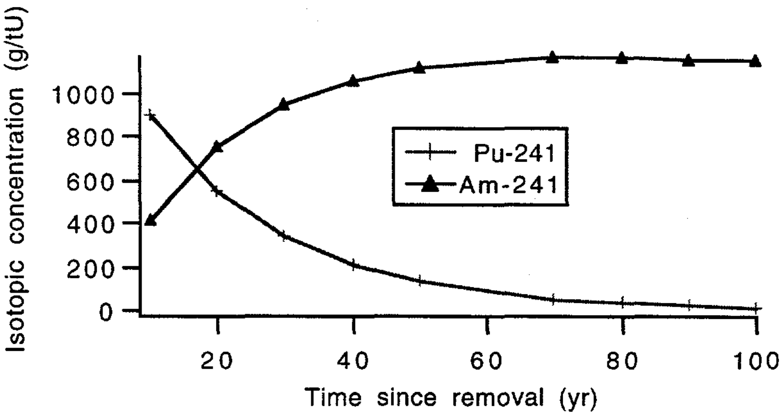

Fig 5. ^{241}Pu and ^{241}Am concentrations vs. time in 35,000 MWd/tU BWR spent fuel.

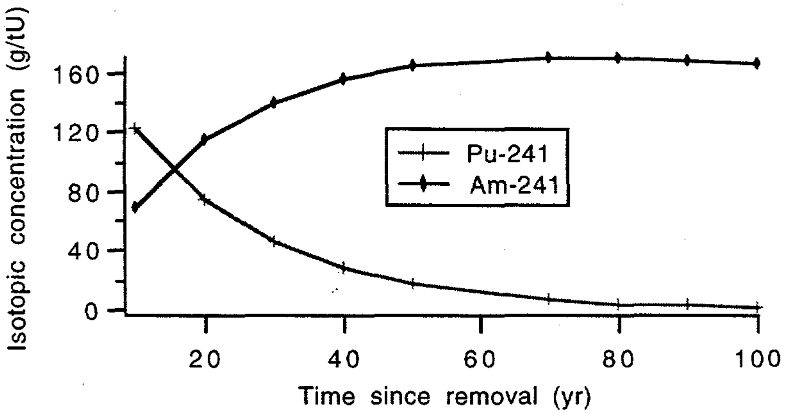

Fig. 6. ^{241}Pu and ^{241}Am concentrations vs. time in 7,500 MWd/tU CANDU spent fuel.

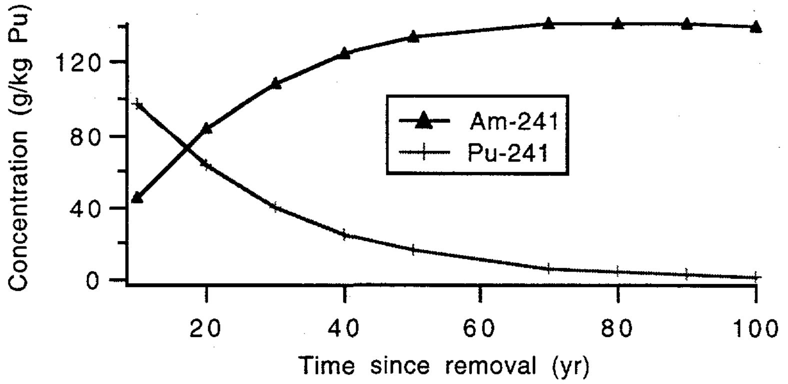

Fig. 7. ^{241}Pu and ^{241}Am concentrations vs. time in Pu separated from 35,000 MWd/tU PWR spent fuel.

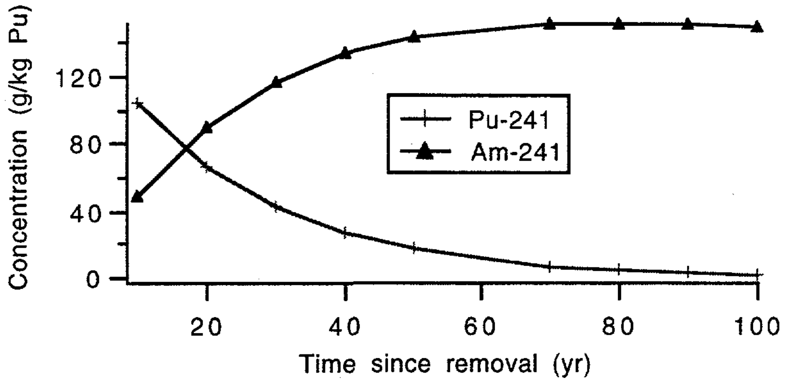

Fig. 8. ^{241}Pu and ^{241}Am concentrations vs. time in Pu separated from 35,000 MWd/tU BWR spent fuel.