Effect of Americium-241 Content on Plutonium Radiation Source Terms

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BACKGROUND

The management of excess plutonium by the US Department of Energy includes a number of storage and disposition alternatives. Savannah River Site (SRS) is supporting DOE with plutonium disposition efforts, including the immobilization of certain plutonium materials in a borosilicate glass matrix.

Surplus plutonium inventories slated for vitrification include materials with elevated levels of Americium-241. The Am-241 content of plutonium materials generally reflects in-growth of the isotope due to decay of plutonium and is age-dependent. However, select plutonium inventories have Am-241 levels considerably above the age-based levels. Elevated levels of americium significantly impact radiation source terms of plutonium materials and will make handling of the materials more difficult. Plutonium materials are normally handled in shielded glove boxes, and the work entails both extremity and whole body exposures.

This paper reports results of an SRS analysis of plutonium materials source terms vs. the Americium-241 content of the materials. Data with respect to dependence and magnitude of source terms on/vs. Am-241 levels are presented and discussed. The investigation encompasses both vitrified and un-vitrified plutonium oxide (PuO_2) batches

OUTLINE OF PLUTONIUM BATCHES

The analysis pertains to weapons grade (WG) plutonium. Isotopic compositions of WG plutonium are given in DOE Standard DOE-STD-3013-96 (Criteria for Preparing and Packaging Plutonium Metals and Oxides for Long-Term Storage, 9/1996). The compositions include 93.6 weight percent (wt%) Pu-239 and 0.4 wt% by Pu-241. In each class batches with low and high Americium-241 concentrations were analyzed.

The un-vitrified batches have an overall weight of 3.5 kg plutonium oxide, and the Am-241 level ranges from comparatively low levels corresponding to in-growth following 15 years decay of WG plutonium and estimated at 0.18 wt% Am-241 to the maximum expected Am-241 level of 5.6 wt%. The vitrified batches have overall weights of 25 kg, plutonium content of 2,490 gm, and Am-241 concentrations range from 0.02 to 0.77 wt%.

ANALYSIS

Parameters investigated include: (a) Neutron and gamma source terms; (b) Contact dose rates, needed to assess to extremity exposures; and (c) Shielded dose rates, applies to whole body exposure of operators at glove box shielded windows.

The RASTA computer code was used to calculate the neutron and gamma source terms of the plutonium masses. RASTA is an SRS in-house computer codes used for engineering calculations. Included in the source terms are neutrons from plutonium and Am-241 spontaneous fission, neutrons from (α, n) reactions in plutonium compounds, and subcritical multiplication neutrons. Gammas from decay of plutonium and plutonium daughter nuclides, prompt and delayed gammas from plutonium and Am-241 spontaneous fission, and gammas from (α, n) reactions in plutonium compounds.

Batches were modeled as spheres and the multi-group neutron/gamma shielding calculations were performed with the ANISN Code.

RESULTS

The more important findings are:

Vitrification Effects – Vitrification of plutonium oxide with a 0.18 weight percent Am-241 yields neutron source terms that are 20 times higher than the un-vitrified source terms. Similarly, vitrification of PuO_2 with 5.6 wt% Am-241 results in neutron source terms that are 40 times higher than the un-vitrified plutonium. The increase is attributed to additional (α, n) reactions with Boron-11 target isotopes present in glass. Gamma source terms are not impacted by vitrification and remain unchanged.

Neutron Source Terms vs. Americium Content - Increasing the Am-241 content of plutonium batches results in significant increases in neutron source terms. Increasing the Am-241 content of un-vitrified plutonium from 0.18% to 5.6% by weight (a 30 fold increase) results in neutron source terms that are 2.2 times higher (120% increase). . The increases are attributed to more alpha particles (α), emitted by Am-241, and additional (α, n) reactions with O-18. In higher concentrations Am-241 surpasses Pu-239 as the dominant alpha emitter. For vitrified plutonium an increase in Am-241 (from 0.02 to 0.77 wt%) results in 4.4 times increase in neutron source terms. For vitrified plutonium most of the (α, n) reactions are with Boron-11 target isotopes present in glass.

Gamma Source Terms vs. Americium Content - Increasing the Am-241 content of plutonium oxide and vitrified plutonium from in-growth levels to maximum expected levels results in significant magnifications in gamma source terms. The analysis shows magnifications of x18 and x23 in gamma source terms for oxide and vitrified materials respectively. Americium-241 decay yields a 0.06 Mev gamma. The majority of gamma photons are in the low energy range of 0.01 to 0.06 Mev.

Contact Dose Rates vs. Americium Content – As expected additional Am-241 result in higher extremity dose rates. For un-vitrified materials gamma photons together with neutrons control extremity exposures. Increasing the Am-241 content of un-vitrified plutonium from 0.18% to 5.6% by weight increases total contact dose rate from 2.3 to 6.1 mSv/hour. For vitrified plutonium neutrons alone control contact dose rates, and the gamma contribution is limited to about 5%, or less. Increasing the Am-241 content of vitrified plutonium from 0.02% to 0.77% by weight causes contact dose rate to increase from 8.3 to 36.5 mSv/hour.

Shielded Dose Rates vs. Americium Content – For both un-vitrified and vitrified plutonium materials dose rates were calculated at the glovebox shielded windows. Independent of the Am-241 content shielded dose rates are controlled by neutrons. This is because of the soft gamma spectrum and attenuation of the photons by the materials that make up the shielded window (i.e. lead glass). Increasing the Am-241 content of un-vitrified plutonium from 0.18% to 5.6% by weight increases total dose rate at the shielded window from 1.4E-3 to 4.0E-3 mSv/hour. Increasing the Am-241 content of vitrified plutonium from 0.02% to 0.77% by weight increases total shielded dose rate from 2.2E-2 to 9.7E-2 mSv/hour.