Mobility, solubility and biokinetics of ^{238}Pu

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1 Introduction

Plutonium-238 is commonly used in radioisotope thermoelectric generators for space missions beyond Jupiter, where the sunlight is too weak to use as a power source. The workers who prepare the plutonium heat sources are at risk for intakes of the material, and require routine monitoring. Although chemically identical, experience has shown that the high specific activity of ^{238}Pu dramatically increases its mobility through a laboratory, and alters its solubility both in the laboratory and in vivo. This effect on solubility, in which recoil nuclei gradually increase the solubility of a particle, is particularly important for the biokinetics of ^{238}Pu oxides, which initially exhibit very low solubility in the respiratory tract.

2 Mobility in the laboratory

While the mobility of ^{238}Pu is well known to those who work with it, many of the details are limited to personal correspondence and anecdotes. At the Savannah River National Laboratory, personnel who worked with ^{238}Pu described it as "lifelike," stating that fines appear to "walk" down pipes and "fly" through the air. It was also reported that surfaces were difficult to decontaminate because particles were easily re-suspended into the air and previously decontaminated surfaces were later found to be re-contaminated after hours or days. [1] The United States Department of Energy warms that "microspheres have been observed to climb walls of glass beakers and spread throughout the glovebox" and that, because of internal heating, 100-200 micron spheres can melt through glove material. [2] Correspondence with Los Alamos workers describe instances of ^{238}Pu hot particles dislodging themselves from wipes or tape. In one instance, after decontaminating a wall, the contamination was found to have migrated to a different wall (presumably driven by electrostatic forces). Quantitative studies have shown that ^{238}Pu particles trapped in HEPA filters are re suspended back into the laboratory at a rate of 5% per month by weight, compared to 0.02% per month for ^{239}Pu. [3] Particles can also be transported through HEPA filters by aggregate alpha recoil at a rate dependent on specific activity and number of atoms per aggregate. [4]

3 Solubility and fragmentation

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When a nucleus decays, the recoil nucleus from an alpha decay can damage a particle and increase its solubility. This is particularly important in plutonium oxides, which, when intact, have extremely low solubility. A cone of 50x50 angstroms approximates the volume damaged by a recoiling nucleus of PuO_2. [5]. The ratio of dissolution between $^{238}PuO_2$ and $^{239}PuO_2$ is about 200, consistent with the ratio of decay rates. When aged $^{239}PuO_2$ was dissolved in water, a large amount of dissolution was observed in the first 24 hours, dropping by about 2 order of magnitude the next day. The high initial solubility was attributed to accumulated damage. In $^{239}PuO_2$, the release material was due to damaged regions in the original particle which were subsequently exposed to water [6,7]. In contrast, $^{238}PuO_2$ has been shown to fragment. For example 34-micron particles have been shown to fragment at 0.001% per day. [8]

4 Biokinetics

Unusual biokinetics for plutonium oxides were first observed in 1971 following an exposure to a $^{238}PuO_2$ ceramic. Excretion was below the detectable limit for about 100 days following the incident, and eventually increased to large values, peaking about 3 years following the incident. The observed excretion could be predicted by starting with an initial lung solubility of 1 X 10^{-6} d^{-1}, increasing gradually to 4 X 10^{-3} d^{-1}. A mechanistic model was developed for non-ceramic plutonium oxides in beagles [9], and was eventually adapted to fit the urinary excretion observed in cases involved in the 1971 incident [10, 11]. The ICRP66/67 models were calibrated to fit the excretion and autopsy data of a worker who was exposed to ceramic $^{238}PuO_2$, but since this was only based on one individual, this modified ICRP model is not broadly applicable [12]. The latest ICRP publication (in draft) includes dissolution models for ceramic and non-ceramic ^{238}Pu-oxides [ICRP 2018]. However, neither of these is able to predict the observed excretion from a more recent incident at LANL involving several individuals, in which the excretion peaks at about 10 days, then remains constant for many years. We consider various ICRP-compatible dissoloution models which are consistent with all of the observed excretion patterns.

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