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Properties and Behavior of ^{238}Pu Relevant to Decontamination of Building 235-F

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Acronym	Definition
CBI	Cellular Bioengineering, Inc.
D&D	Decontamination and Deactivation
GPHS	General Purpose Heat Source
HEPA	High Efficiency Particulate Air
MMAD	Mass Median Aerodynamic Diameter
NASA	National Aeronautics and Space Administration
PuFF	Plutonium Fuel Form Facility
RTG	Radioisotope Thermoelectric Generator
SRNL	Savannah River National Laboratory
SRS	Savannah River Site

1.0 EXECUTIVE SUMMARY

This report was prepared to document the physical, chemical and radiological properties of plutonium oxide materials that were processed in the Plutonium Fuel Form Facility (PuFF) in building 235-F at the Savannah River Plant (now known as the Savannah River Site) in the late 1970s and early 1980s. An understanding of these properties is needed to support current project planning for the safe and effective decontamination and deactivation (D&D) of PuFF.

The PuFF mission was production of heat sources to power Radioisotope Thermoelectric Generators (RTGs) used in space craft. The specification for the PuO₂ used to fabricate the heat sources required that the isotopic content of the plutonium be $83 \pm 1\%$ Pu-238 due to its high decay heat of 0.57 W/g. The high specific activity of Pu-238 (17.1 Ci/g) due to alpha decay makes this material very difficult to manage. The production process produced micron-sized particles which proved difficult to contain during operations, creating personnel contamination concerns and resulting in the expenditure of significant resources to decontaminate spaces after loss of material containment.

This report examines high ²³⁸Pu-content material properties relevant to the D&D of PuFF. These relevant properties are those that contribute to the mobility of the material. Physical properties which produce or maintain small particle size work to increase particle mobility. Early workers with ²³⁸PuO₂ felt that, unlike most small particles, Pu-238 oxide particles would not naturally agglomerate to form larger, less mobile particles. It was thought that the heat generated by the particles would prevent water molecules from binding to the particle surface. Particles covered with bound water tend to agglomerate more easily. However, it is now understood that the self-heating effect is not sufficient to prevent adsorption of water on particle surfaces and thus would not prevent agglomeration of particles.

Operational experience at PuFF indicates that the Pu-238 contamination was observed to move along surfaces and through High Efficiency Particulate Air (HEPA) filters over time. Recent research into the phenomenon known as alpha recoil offers a potential explanation for this observed behavior. Momentum is conserved when an alpha particle is ejected from a Pu-238 atom due to radioactive decay. Consequently, the entire particle of which that Pu-238 atom is a constituent experiences a movement similar to the recoil of a gun when a bullet is ejected. Furthermore, the particle often fractures in response to Pu-238 atom disintegration (yielding an alpha particle), with a small particle fragment also being ejected in order to conserve momentum. This process results in the continuous size reduction and transport of particles containing Pu-238 atoms, thus explaining movement of contamination along surfaces and through HEPA filters.

A better understanding of the thermal behavior of ²³⁸PuO₂ particles is needed to inform the planning process for the PuFF D&D project at the 235-F facility. There has been a concern that the surface temperature of individual particles may be high enough to cause problems with decontamination equipment and materials as a result of heat generation due to radioactive decay. A calculation under conservative assumptions shows that the surface temperature of particles less than about 100 μm diameter is not appreciably above ambient. Since most particles in PuFF are on order of 1 μm in diameter, the effect of particle surface temperature on decontamination equipment and materials is expected to be minimal. The result of this calculation also indicates that thermal imaging, which has been under consideration as a method to monitor the progress of system decontamination efforts would not likely be effective.

The use of strippable coating was suggested as a possible alternative to other decontamination techniques. One particular system (i.e., Decon Gel 1101) may offer significant advantages over conventional liquid decontamination solutions. Previous trials are discussed and have been used successfully at SRS on various surfaces. Some areas showed minimal improvement, while others presented complete (100%) decontamination. However, small scale testing is recommended prior to the full scale use in the cells of PuFF in building 235-F.

2.0 PU-238 PRODUCTION PROCESS– OVERVIEW

The Savannah River Plant began producing Pu-238 in the late 1950s by irradiating Np-237. Approximately 300 kg were produced between 1959 and 1988. In 1961, Building 235-F was converted to produce Np-oxide-aluminum targets for irradiation in the reactors. The target design evolved into thin tubes fabricated in the 321-M building. Separations were initially performed in the high-level caves. The process was scaled up and mounted onto frames installed in the 221-H canyon.¹ Initially, oxide powder was shipped elsewhere for fabrication into heat sources. In 1971, however, the Atomic Energy Commission made a decision to transfer plutonium containing cermet fuel form preparation to Building 235-F at the Savannah River Site (SRS).

2.1 Pu-238 Properties

Plutonium is a man made radionuclide that exists in several different isotopes having molecular weights ranging from 228 to 247. In metallic form, 6 different crystal structures (allotropes) exist between room temperature and its melting point (640 °C). Its oxide is very stable and can exist in a range of different stoichiometries. Pu-238 has a very high activity relative to its other isotopes and hence a shorter half-life (i.e., 87.7 years).

Various stoichiometries exist for the oxide; the composition of which is largely dependent on temperature and oxygen concentration. In an oxygen rich atmosphere, the primary oxide phase is observed to be PuO₂, but other intermediate stoichiometries are also observed. The oxide structure is based on the CaF₂ structure (See Figure 1), which is a face centered cubic lattice with cations (i.e., Pu) at corners and faces of the unit cell and anions at tetrahedral sites (i.e., O). Ordered vacancies on 25% of the anion sites occupied by oxygen produce a different stoichiometry of oxide (i.e., Pu₂O₃)². PuO₂ is the most stable form of the oxide at ambient temperature. Its high melting point (2400 °C) and defect tolerance makes it a good choice for reactor and radiological applications. In addition, uranium, americium and neptunium oxides are stable in the same oxide to metal ratio and crystal structure. Hence, single phase, multi-component solid solutions exist between these elements, although chemical segregation has been observed under certain conditions.³ Table I shows some of the relevant physical properties of PuO₂.

The physical properties of ²³⁸PuO₂ were reviewed by Congdon⁴ in relation vitrification technologies for safe storage and transportation. Congdon pointed out that the unique properties of Pu-238 prevent the use of simulant oxides for process development and property investigations. Its relatively high activity and self-heating properties were cited as the primary reasons for this conclusion.

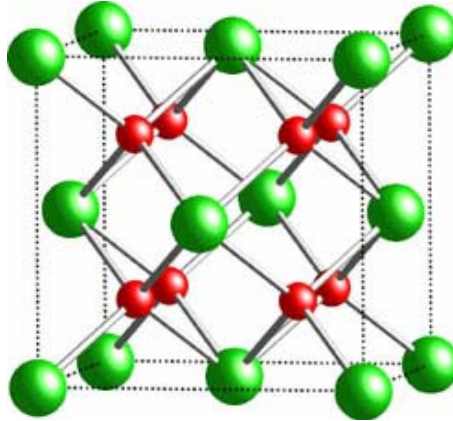


Figure 1: Schematic of the CaF₂ structure which is the basis for oxide phases in the plutonium-oxide system (Pu in green, O in red).

Table I: Physical Properties of ²³⁸PuO₂

Property	Value
Melting Point	2400 °C
Theoretical Density	11.46 g/cm ³
Typical GPHS Density	82-85% of theoretical
Half life	87.7 years
Decay Mode	α particle to U-234
Energy	5.5 MeV
Specific Activity	17.1 Ci/g
Decay Heat	0.57 W/g

2.2 235-F Operational History

The Savannah River Site made a significant contribution to the National Aeronautics and Space Administration (NASA) space missions through its support of the Pu-238 heat source program. Construction of PuFF began in October 1973 and was completed in mid-July 1977. In total, nine cells were fabricated for the production of General Purpose Heat Sources (GPHS's) which included facilities to operate the process which started with oxide powder and produced encapsulated heat sources. Production of iridium-encapsulated 100-W Pu-238 spheres for multi-hundred watt RTGs commenced in 1978 and was completed in April 1980. In June 1980, production of the 62.5-W GPHSs for NASA's Galileo and Ulysses missions began. By December 1983, all heat source production was completed for these missions. The last heat sources were shipped off-site in February 1984 for final assembly. During production, PuFF processed approximately 165 kg of Pu-238. When production was completed, the PuFF facility was placed in standby mode.

2.3 Baseline Process

Pu-238 is produced by irradiating a target containing Np-237, which was recovered from existing SRS processes where it had been produced as a byproduct in the reactor irradiation of uranium. The

Np-237 was fabricated into targets and irradiated in SRS reactors to form Np-238. The Np-238 isotope beta decayed to produce Pu-238. Pu-238 was separated from the neptunium and fission products in H-Canyon and processed to form plutonium oxide in HB-Line. The isotopic content of the feed material is shown in Table II and was controlled to be approximately $83.5 \pm 1\%$ Pu-238 oxide with most of the remaining material as Pu-239 oxide. Although Pu-238 and Pu-239 are different radiologically they behave similar to one another, chemically. Also in Table II, an estimate of the composition of feed material is presented. Over time, in-growth of decay products (i.e., U-234 and Am-241) is expected to remain in the same oxide phase but increase the defect density due to alpha decay. The physical properties of the oxide particles will be changed as a result of the new composition. However, at least in the present case, the physical properties of the daughter products are not significantly different from the parent. For example, one study compared the thermal properties of various actinides and found them to be relatively similar to one another (see Figure 2).⁵ In cases where the properties of the individual components of a blended oxide differ, the property may be estimated using the rule of mixtures.

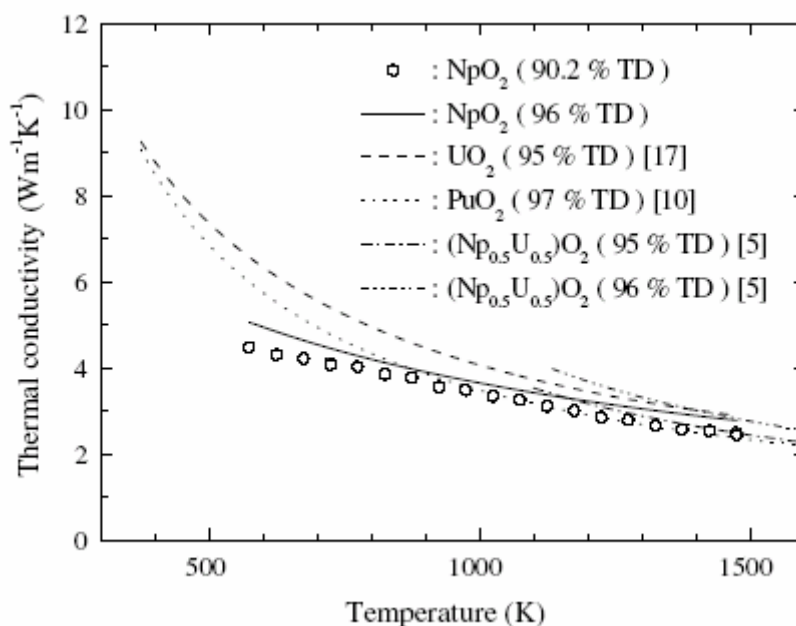


Figure 2: Thermal conductivity of NpO_2 (corrected for differences in theoretical density) together with those for UO_2 , PuO_2 and $(\text{U}_{0.5}\text{Np}_{0.5})\text{O}_2$ ⁵

The production process for encapsulation from raw feed powder may be separated into 10 characterization and processing steps. These steps are shown in Figure 3 and summarized below. As shown in the figure, the steps can be grouped into two parts: granule production and pellet production. Plutonium (III) oxalate is produced by adding plutonium nitrate to oxalic acid. After precipitation as plutonium oxalate, separation by filtration and drying, the material is converted to plutonium oxide by heating in an oxygen rich environment in HB-Line. The remainder of the process was conducted in the cells of PuFF.

Table II: Typical isotopic composition Pu-238 oxide produced for heat sources in PuFF⁶

Isotope	Initial Weight Percent (%)	Specific Activity (Ci/g)	Activity per gram of feed material (Ci/g)	Relative Activity (%)	Current Weight Percent*	Activity per gram of feed material (Ci/g)	Relative Activity (%)
Pu-238	84.5	1.71×10^1	1.40×10^1	95.7	66.7	11.533	98.7
Pu-239	12.3	6.21×10^{-2}	8.89×10^{-3}	0.060	12.3	0.008	0.1
Pu-240	2.6	2.27×10^{-1}	6.82×10^{-3}	0.047	2.6	0.006	0.1
Pu-241	0.5	1.03×10^2	6.19×10^{-1}	0.423	0.1	0.127	1.1
Pu-242	0.1	3.94×10^{-3}	3.94×10^{-6}	2.69×10^{-3}	0.1	0.000	0
Pu-236	$<2 \times 10^{-6}$	5.32×10^2	1.06×10^{-5}	7.24×10^{-3}	0	0	0
Am-241	0	3.47	0	0	0.4	0.014	0.1
U-234	0	6.32×10^{-3}	0	0	17.8	0.001	0

*Allowed to decay for 30 years⁷

After the PuFF facility received Pu oxide from HB-line, the powder would be heated in O-16 rich oxygen to remove any fraction of O-17 and O-18 isotopes present in the feed. This step was performed to reduce neutron radiation generated as a result of the reaction between alpha particles emitted by Pu-238 and these light neutron-rich atoms (i.e., α , η reactions). The powder would then be ball milled to a target particle size of $<1 \mu\text{m}$, cold pressed into pellets and granulated into a bi-modal particle size distribution (this was done to control shrinkage in the final sintering step). The granules were pre-sintered at different temperatures, blended, hot pressed and sintered again. The resulting product was a high density, cylindrical pellet with stringent acceptance criteria. This pellet would eventually be clad in iridium and packaged as a GPHS.

2.4 Baseline Particle Size Distribution

PuO₂ was received in the PuFF cells as calcined powder. This processing step involves heating the oxalate precipitates, driving off any remaining moisture and oxalic ions. Nominal furnace temperature during calcining is 735°C.⁸ This dries the plutonium oxalate precipitate and converts the plutonium oxalate precipitate to plutonium-oxide, PuO₂. The initial particle size has been observed to be sensitive to both oxalate precipitation technique and calcination temperature. Figure 4 shows the particle size distribution of the PuO₂ produced from calcination of various types plutonium oxalate feeds. The histograms show a weight averaged particle size distribution of calcined PuO₂ baseline process. In general, two particle shapes are usually produced under these conditions: Lath shaped precipitates and larger rosette-shaped agglomerates. Additionally, a large amount of variability can be observed within these two categories. For example, the aspect ratios of the lath dimensions were observed to vary up to 50-fold¹⁰ and Pu valence state can determine the symmetry and shape of the particles. Figure 5 shows the morphology of typical lath particulates and agglomerates produced by two different methods. In general, the baseline process produced as-received calcined powder with a mass-median diameter of about 3-5 μm (see Figure 4A) but is reported to be as high as 30 μm .¹⁰ For the purposes of the D&D or 235-F a lower bound estimate of particle size distribution should be assumed.

Due to the wide variability of particulate morphology, ball milling the feed powder was required to produce the desired particle shape and size for initial cold pressing. This is, to some extent, a

normalizing step to eliminate differences in surface activities and powder packing from one lot of feed powder to the next. The as-received powder had a mass-median diameter of about 3 μm . Data during the Cassini program showed mass-median particle size of 20 μm . The lath-shaped particles have an extremely high surface activity, low packing density which causes the material to shrink excessively when sintered. The results of developmental studies show the reactivity of the feed would be adequate for bonding during hot pressing if the powder is milled to produce a mass-median diameter of about 1 μm with a significant mass fraction below this value.⁹

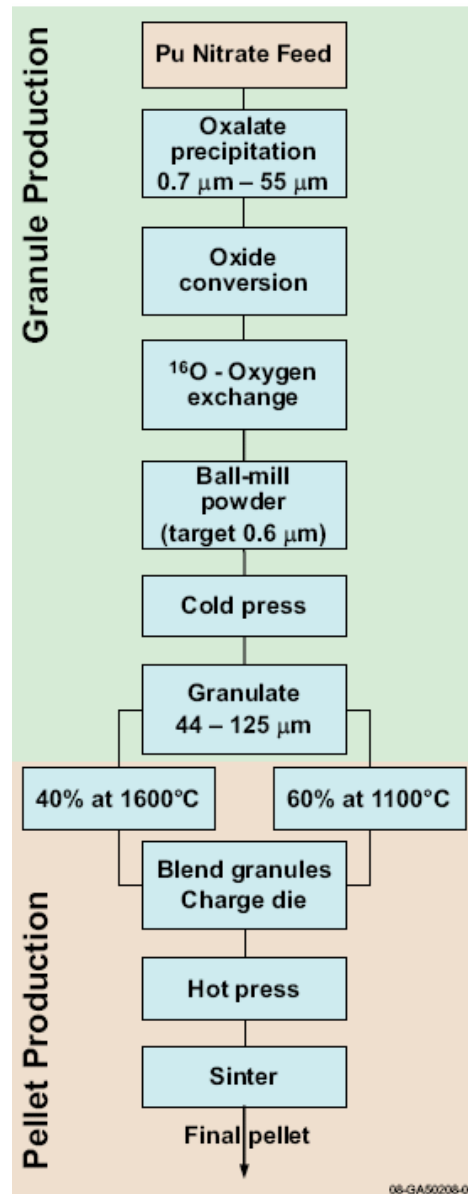


Figure 3: Schematic Flow Chart of Heat Source Production Process¹

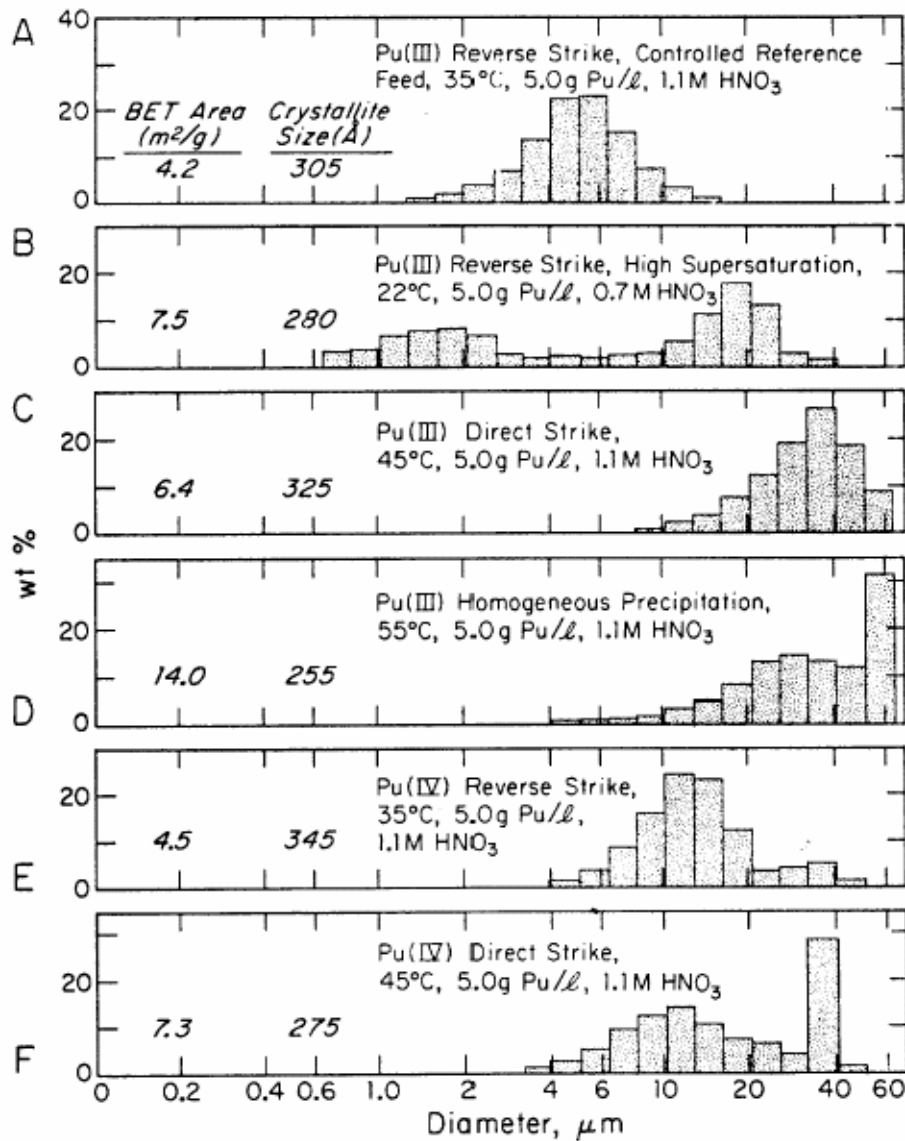


Figure 4: Typical mass particle size histograms of calcined Pu oxide powder (at 735 °C) for various oxalate precipitation methods. For the reverse strike precipitation procedure, Pu feed solution was added with mixing to saturated (0.9M) oxalic acid solution; for direct strike, the acid was added to the feed.¹⁰

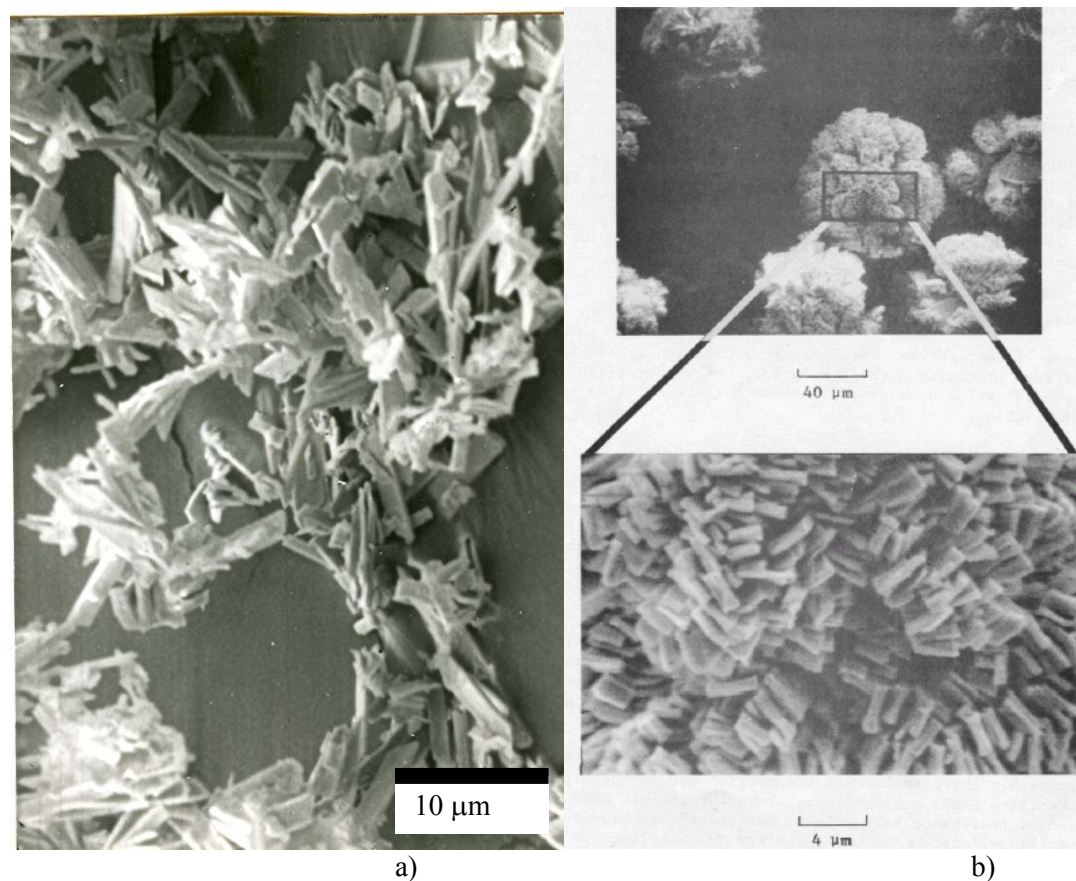


Figure 5: Particle morphology of Pu oxide produced by the reverse strike method (a) and direct strike method (b)^{11, 12}

2.5 Agglomeration

The force of attraction between two colloidal particles is a function of the distance separating those particles and more importantly, the nature of the surface of those particles¹³. It is strongly believed that no water can exist on the surface of Pu-238 particles; however, there is extensive literature evidence that there is an adsorbed layer (or multilayer) of water on the surface of each PuO₂ particle in the PuFF facility. In general, inorganic oxide surfaces in vacuum possess an equal number of positively and negatively charged sites, either metal or oxygen. When exposed to air, which usually contains at least a small relative humidity (15%), these charged sites react with water to create surface hydroxyls, denoted M-OH. Haschke and Ricketts (1997) have stated that the strongest interaction on the Pu-oxide surface is that between a Pu atom and the oxygen atom in a water molecule. The cycle of water adsorption to the PuO₂ surface is shown in Figure 6. The dry Pu-oxide surface comes in contact first with water in the form of a gas which condenses onto the surface. Oxygen then catalyzes the subsequent oxidation of the surface to the higher oxide. Oxygen and hydrogen are released upon water desorption, but are said to quickly recombine on the surface. As more moisture in the air is seen by the particle surface, the molecular water layers build on the surface until equilibrium is reached. Water is removed from the surface with heating at three distinct kinetically favorable temperatures, although complete water removal was not achieved until temperatures of over 1200 °K were reached.

The potential for radiolysis of water has been raised by researchers at SRS and other sites, although significant evidence of this phenomenon has not been obtained.² Some researchers have reported the evidence of hydrogen gas in strong excess of oxygen as a result of water radiolysis, but the presence of water vapor in the air would quickly replace any water lost in this manner. Gas generation may be a concern for storage purposes, but not for agglomeration of the particles.

Self-heating of PuO₂ particles is also thought to contribute to water removal from the surface. However, as mentioned above, a surface temperature of nearly 1000°C would have to be reached before 100% removal can be achieved of both physically and chemically adsorbed water layers¹⁴. It has been estimated that nearly three molecular layers of water exist at 60% relative humidity, which most likely represents the current conditions of the PuFF facility without environmental control. In regard to a parallel issue, in reference to corrosion of container materials of construction by PuO₂ under long term storage conditions, Haschke and Martz¹⁵ noted that, “the evidence for the radiolysis of water (*due to alpha radiation*) is absent. If, in fact, water is dissociated by alpha radiation, the O₂ and H₂ products are transient and immediately recombine on the catalytic oxide surface.”

There are several mechanisms for agglomeration, which are all enhanced by the presence of water on the surface of the oxide particles. First, Brownian agglomeration describes the aggregation of particles due to random movement and subsequent collision while suspended in a given medium (in this case, air). Due to dipole-dipole interactions and capillary forces between water molecules on opposing particle surfaces, agglomeration is most likely irreversible. Agglomerated particles will then settle onto a surface according to Stokes Law, which will occur faster for larger particles due to gravity. Gravitational agglomeration then occurs due to the size dependence of the terminal velocity of small particles, which are captured by faster settling larger particles or agglomerates. Lastly, turbulent agglomeration can occur if particles are aligned in the same flow path or trajectory, due to eddy currents or airflow¹⁶.

With respect to particles in the PuFF facility, there is a good chance that few if any particles remain suspended in the air due to water adsorption and agglomeration by Brownian collisions, settling, and/or air turbulence. De-agglomeration may occur due to alpha recoil and is covered in section 2.7; however, it is believed that adsorbed water/enhanced agglomeration by the mechanisms suggested here are much stronger and more frequent than the spallation of small particles and agglomerate break-up by alpha recoil. The rate at which particles fragment from PuO₂ was described by a simple model as follows¹⁷

$$\frac{dV}{dt} = \frac{\lambda n V}{4} \left(\frac{\delta}{r} - \frac{\delta^2}{r^2} + \frac{\delta^3}{3r^2} \right),$$

where V is the volume of the particles, λ is the decay constant, n is the number of atoms ejected per recoil, r is particle radius, and δ is the recoil nucleus range. The rate of ²³⁸PuO₂ is estimated to be 200 times greater than that of ²³⁹PuO₂. For ²³⁸PuO₂, this rate was reported to be about 0.001%/day. However, sub-microscopic alpha recoiled particles are likely to obtain an adsorbed water layer within minutes of ejection and subsequent agglomeration due to van der Waals attractive forces at small distances to other particles.

In summary, it has been discussed that water exists on the surface of the particles in the PuFF facility and that water will not be removed significantly by water radiolysis or self-heating. These molecular water layers will enhance the affects of agglomeration by collision in air by making the particles

“sticky” and facilitate the gravitational settling of aerosolized particles present in the facility. Small particles ejected from larger particles by alpha recoil will quickly be covered by the same water multilayer experienced by the parent particle and suffer the same agglomeration and gravitational settling. The potential for these particles to be entrained in filters is discussed in another section.

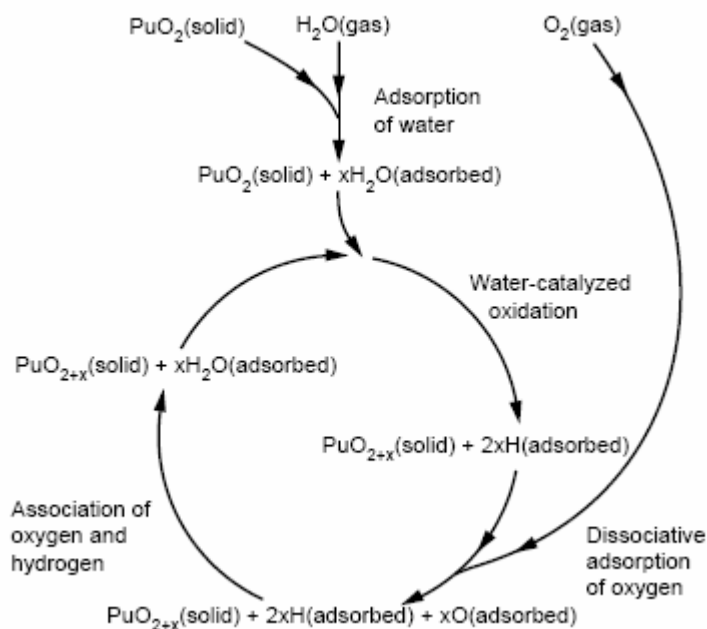


Figure 6: Cycle of adsorption/oxidation reactions when PuO₂ is exposed to water vapor²

2.6 Alpha Recoil Driven Decrepiation

The alpha fluence of ²³⁸PuO₂ is ~ 300x greater than ²³⁹PuO₂. This can lead to differences in physical behavior. For example, research has shown that PuO₂ solubility is not zero and there is a significant difference in the rates of dissolution between ²³⁸PuO₂ and ²³⁹PuO₂ which was thought to be due to the higher specific activity of the ²³⁸PuO₂. These rates of dissolution are small and dependent on factors such as the pH, temperature, the presence of oxidizing, reducing or complexing agents, the surface areas of the particles, and the history of the sample.¹ Fleisher¹⁷ proposed an explanation for this behavior based on alpha recoil. In this model, subparticles would be produced as recoiled aggregates. The differences in “dissolution rates” between ²³⁸PuO₂ and ²³⁹PuO₂ were postulated to be controlled by radiation damage and that alpha decay occurring near the surface of the particle ejects a certain number of atoms from the particle reducing its overall size. Figure 7 illustrates this model with a schematic. An alpha particle can only travel a short distance in PuO₂. The range of an alpha particle was determined to be approximately 12 microns in UO₂, PuO₂ and ThO₂ phases.¹⁸ Hence, a dotted line denotes the recoil range which is the zone around the outside surface of the particle where it would be possible for the recoil from an alpha decay to generate a fragment.

Several studies on dispersion of materials by the alpha recoil mechanism were reviewed by Icenhour.¹⁹ The review concluded, "The recoil energy resulting from alpha decay is sufficient to re-suspend

nanometer-sized particles from a filter fiber. Particles with densities of 10 g/cm^3 and up to 20 nm in diameter can be dislodged. While alpha-recoil energy is not sufficient to re-suspend micron or submicron particles, fragmentation can lead to the production of even smaller particles that can be re-suspended."

The reason alpha recoil is insufficient to dislodge larger particles is due to conservation of momentum. The recoil energy is inversely proportional to the particle's mass. For example, a recoiled daughter product of Pu^{238} (i.e., U^{234}) by alpha decay of a 5.5 MeV alpha particle has been determined to have an energy of $\sim 100 \text{ keV}$ (the mass ratio of U^{234} : alpha particle is $\sim 59:1$) and a 10 nm recoiled particle fragment would have an energy of approximately 1eV. As mass increases and recoil energy drops, particle ejection from a surface becomes implausible. However, fragments up to 20 nm could be dispersed in air and facilitate the release of PuO_2 to the environment if filtration technology is not adequate re-capture these particles. The decrepitation and re-distribution of materials has been modeled based on the activity of the species concerned and determined that release rate is dependant on lattice damage by recoil nuclei and alpha particles, as well as radiolytic effects.^{19,}
²⁰ The range of recoiling particles in air resulting from alpha decay with energies of 6 MeV is about 0.12 mm, so fragments re-distributed by this mechanism will not travel large distances without the assistance of other transport mechanisms (e.g., entrainment in air currents, dissolution or transfer by contact). The settling of particles in this range of sizes is very slow and dominated by Brownian motion²¹ (see Section 3.0). However, it is not expected that the phenomenon of alpha recoil could result in transport of contamination of significant distance against air currents.

An alternative explanation that would cause the higher release rate of $^{238}\text{PuO}_2$ is an enhanced reaction rate of dissolution and a subsequent production of colloids in the presence of moisture. Alpha decay and recoil nuclei can damage the lattice structure of $^{238}\text{PuO}_2$. As a result, higher defect density in the structure would lead to an enhanced release rates to the environment. Patterson, *et al.* studied the long term release of $^{238}\text{PuO}_2$ in simulated terrestrial environments (air, water and soil).²² They noted that the principal mechanism for plutonium release from these sources is the removal of the submicroscopic oxide particles on the surfaces, by dissolution and transport in water, and not airborne fines. Some larger particles were observed to be released to air from large chunks of $^{238}\text{PuO}_2$ during the initial period of rain. The release rate of plutonium to the environment during these periods was observed to increase by 10,000 times. Thermal shock of the larger, self-heated chunks was proposed to be the controlling mechanism behind the increase in release rate. Once the chunk cooled to ambient temperatures, by evaporative cooling, the release rate dropped relatively quickly. This behavior was **not** observed when release rates were measured over "fines" which were said to be at ambient temperatures already.

Based on these observations, it appears that the redistribution of Pu-238 contamination could occur by a number of mechanisms. The spread of contamination of the $^{238}\text{PuO}_2$ will most likely depend on environmental conditions (moisture content, temperature, particle size distribution, etc.). Fine particles of $^{238}\text{PuO}_2$ are believed to continue to break up as a result of alpha recoil. While the initial distance of this recoil is limited, the ability for entrainment in air currents may cause them to be transported farther. The spread of contamination from larger particles of $^{238}\text{PuO}_2$ may occur by the process of wetting with liquid water and fragmentation by thermal shock. More analysis is needed to determine the mechanisms that will dominate the redistribution of plutonium during decontamination activities in the PuFF facility.

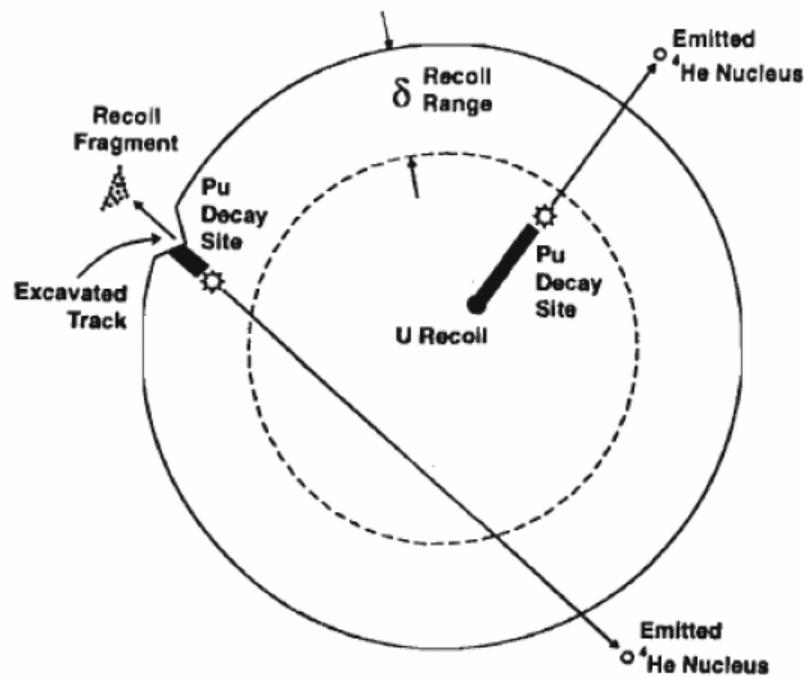


Figure 7: Plutonium fragmentation related specific alpha activity¹⁷

3.0 PU-238 PARTICLE SETTLING AND FILTRATION

The probable particle size distribution of Pu-238 oxide in the PuFF facility cells is difficult to estimate. However, other properties are not. As shown in Figure 5, the basic morphology of PuO₂ is known to be a lath (thin, narrow strip) with a thickness: width: length ratio of approximately 1:4:16. Table I gives the theoretical density of 11.46 g/cc. The density of the powder would be bounded by this. Stokes law of settling is an effective method of estimating the terminal velocity (or settling velocity) of particles in air. In general, the settling rate of particles is dependent on a balance of forces exerted on them: the buoyancy forces that result from the density of the fluid of suspension, the frictional drag on the surface of the particle and the acceleration due to gravity. The law is expressed for hard spheres as:

$$V_t = \frac{gd^2(\rho_p - \rho_m)}{18\mu}$$

where V_t is the terminal velocity, g is the acceleration due to gravity, d is the particle diameter, ρ_p and ρ_m are the densities of the particle and medium, respectively, and μ is the viscosity of the fluid.

Figure 8 illustrates the behavior of particles with the same density of PuO₂ by plotting settling time (i.e., time for a particle to settle through a distance of 1 ft of static air) vs. particle diameter. From Stokes law, the settling time for a 10 μm particle is ~9 seconds; for a 1 μm particle it is ~25 minutes and for a 0.1 μm particle it is ~25 hours. Radiological hoods are required to have a face velocity of at least 100 fpm, which would be enough air velocity to entrain a maximum particle diameter of about 30 μm . The actual size of particles entrained in ventilation would depend on the direction of the flow

in relation to gravity and other factors. In addition, particle shape would play a role in settling rates. The lath shape particles should settle faster than spheres. In stagnant flow conditions, the particles will settle at a rate of approximately V_t .

Practically, particles with settling velocities below 0.1 ft/min (e.g., 1 μm) will never settle out because settling of particles in this range is dominated by Brownian motion.²¹ Gas molecules are in constant motion. Small particles are more affected by collisions with gas molecules than large particles. Below a certain size the terminal velocity is so low that these collisions will offset the gravitational settling and the particle will never settle out of the air. Hence, controlling the movement of these particles is best achieved by controlling the air itself. At a face velocity of 100 ft/min with particles of 30 μm or less the flow conditions are laminar and would be expected well defined and not prone to spread as a result of turbulence. The presence of eddie currents near surfaces or obstacles could affect this transport and lead to deposition or spread of contamination but to what degree is not clear. It is expected, however, that these particles will not behave substantially different than other particles of similar size, shape and density in like air flow conditions.

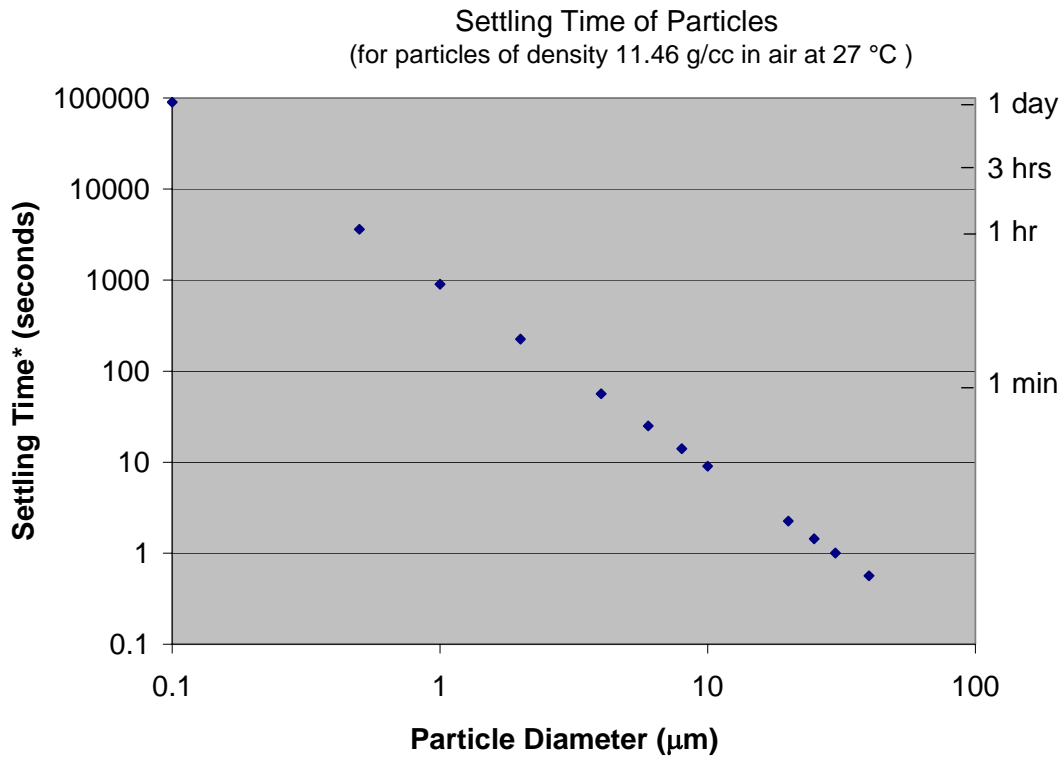


Figure 8: Settling Time of PuO_2 Particles as a Function of Particle Diameter²³

3.1 Impact of Particle Size Distribution on Filter Efficiency

Given the limited information²⁴ about the probable particle size distribution of Pu-238 particles (~1 μm) in the PuFF facility cells, there are several implications to using filters for particle filtration and capture that must be considered. Figure 9 provides a graphical representation of the mechanisms of

particle capture with respect to particle size and filter efficiency. It has been reported that for particles less than 100 nm, Brownian diffusion is the main mechanism for filter capture and this diffusion increases with decreasing particle size. The random-walk diffusion mechanism provides increased probability of collision (and adhesion via Van der Waals forces) with a filter fiber. Larger (>400 nm) particles are thought to collide and adhere with the filter due to inertial impaction due to larger mass²⁵.

There is currently much debate on the efficacy of a HEPA-type filter for airborne nano-particulates. However, the current National Institute of Safety and Health (NIOSH) document with recommendations on safe handling of nanoparticles has suggested that a HEPA-type filter will effectively remove airborne nanoparticles.²⁶ HEPA filters will collect 99.97% of particles down to and including 0.3 μm (300 nm).²⁷ It is recommended that a multi-stage or graduated filter system with progressively small filter media be used for added protection. Additionally, an ultra low penetration air filter may be used with effectiveness down to 0.12 μm with 99.999% efficiency. However, this does not include the predicted size range of ball-milled particles which include particles in the 10-30 nm range. *There are no filters currently rated for particle collection in this size range.* However, the current suggestions for safe handling of nanoparticulate materials include the use of a HEPA-like filter system for capture of all airborne particulates. This information also does not take into account the effects of alpha recoil, which has been suggested by some to cause nanoparticles to deviate from normal classical penetration models²⁸

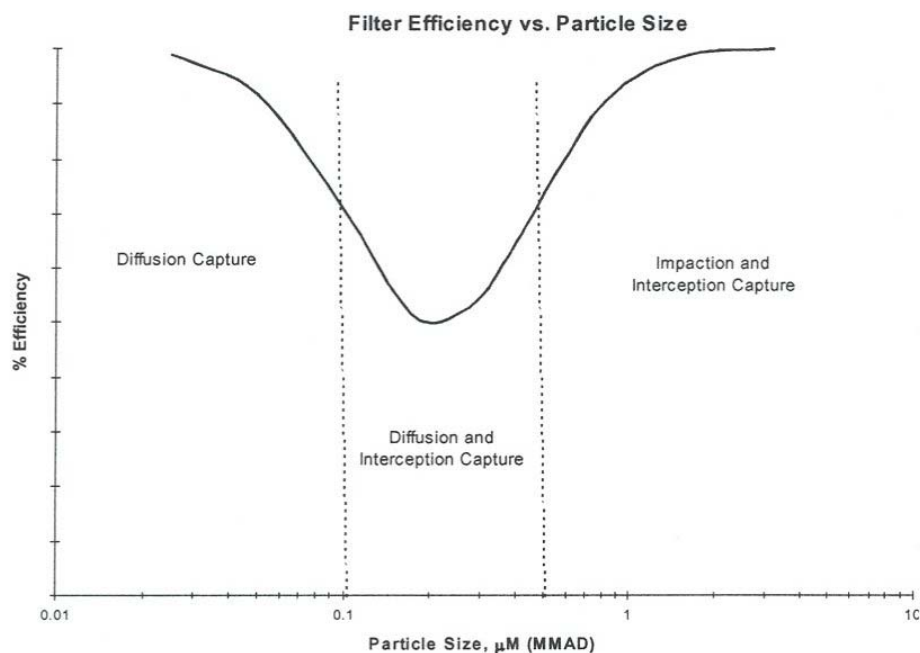


Figure 9: Schematic of filter efficiency with respect to particle size (MMAD is mass median aerodynamic diameter).

3.2 Alpha Recoil as it Relates to Filter Efficiency

Pu-oxide particles are known to adsorb large quantities (see section 2.6) of water on a given surface area (surface area typically 10-20 m^2/g). Wang, et al state that this adsorbed water layer is responsible for the interfacial adhesion between nanoparticles and the filter media²⁹. If that water

layer can be removed by thermal heating, then there would be little interfacial interaction between the particle and filter, thus resulting in the particles penetrating the filter. As mentioned in section 2.6, the physisorbed water layer can be removed at lower temperatures (50 - 100°C), while the chemisorbed layer cannot be removed until heated above 1000°C. Physisorption includes weakly bound water molecules while chemisorption represent more strongly bounds molecules, usually including a covalent chemical bond. Since approximately 66% of the water is physisorbed and the particle surface temperature will never reach 1000°C (unless a sufficiently large particle existed, which would be filtered easily), there will still be a ~33% water layer on the Pu particle surface for filter adhesion. This layer may be discontinuous, but still present. The structure of the remaining water layer after the initial desorption step is uncertain. Particle surface heat generated by alpha recoil will not prevent filter media adhesion, but may decrease its efficiency.

The above discussion concludes that particles will be caught in a series of HEPA-like filters; however, the additional alpha recoil phenomena has not yet been factored into the filter efficiency consideration. Some have concluded that alpha-emitting particulates penetrate HEPA filters much more readily than Beta-emitting or non-radioactive particles.¹⁹ This trend has been observed for Pu-238 specifically at SRS where multiple grams of particles were detected beyond the first HEPA filter layer³⁰. Aggregate recoil particles, which are produced from larger particles, are re-entrained into the airflow and deposited deeper into the filter, or onto a subsequent filter in the series. However, it is still believed that sub-micron sized particles will eventually be entrained in the filter due to Brownian motion collisions with the filter media and the adsorbed water layer, which enhances adhesion with the filter. One HEPA filter is obviously not sufficient to capture all particles and subsequently ejected particles due to alpha recoil. It is also necessary to change filters frequently so that particles ejected from the last filter layer cannot become re-entrained into the air.

4.0 PU-238 BINDING ENERGY WITH METALS AND OXIDES

Most data on corrosion and reactivity with other oxides can be found in support of safe interim storage of Pu containing materials. In general, corrosion of storage containers is often controlled by the impurities in the oxide, not the oxide itself. Safe storage of Pu compounds requires that the materials be treated to stabilize the material (i.e., 2hr @ 950 °C in oxidizing atmospheres). This stabilization treatment is required to reduce the reactivity of the materials such that gas generation and subsequent pressurization is not a problem. Specifically, this treatment is designed to reduce the volatile species content, especially water. Although this treatment tends to reduce the corrosive nature of the stored material, the degradation of stainless steel container materials is still a concern. Container materials will be exposed to ionizing radiation, elevated temperatures, embrittling agents, chloride-containing compounds, and a limited quantity of moisture. In general stainless steel is ideally suited for this type of environment, but has limitations for use. For this reason, the moisture content for all materials stored in the Department of Energy 3013 canisters is limited to restrict the amount of hydrogen generated by radiolysis of water and the amount of water available to dissolve reactive impurities in the oxide.³¹ Certain conditions can exist that will lead to stress corrosion cracking. This phenomenon can lead to the existence of internal contamination in stainless steel by exposing localized crevices in the matrix. However, Pu-238 is not generally considered soluble in the surface films of stainless steel.

Aluminum, however, is noted as exhibiting unsatisfactory corrosion performance in the presence of ²³⁸PuO₂ contamination. Several sources have noted that the presence of extensive corrosion taking place as a result of the high specific activity of Pu-238 contributing to spallation of its protective oxide film. One illustrative example¹ is that of the manipulators in 235-F PuFF facility that had to be

replaced after the facility was taken off standby. The reason that was given was that the contamination contributed to the accelerated corrosion rate of all aluminum inside the cells. Figure 10 illustrates this phenomenon by showing the erosion rates of He^+ ions for various substrates. As can be observed, the erosion rate for aluminum is much higher than that of stainless steels. Severe exfoliation of blister skins is observed to occur at lower temperatures in aluminum alloys which will increase surface area and roughness.³² In addition, the corrosion performance of aluminum alloys is severely reduced in the presence of fluorides and chlorides (two common impurities in Pu processing). Therefore, the use of aluminum in the presence of Pu-238 should be limited to components that can be adequately maintained or disposed of easily. It is expected that the potential for internal contamination in aluminum alloys is high and extensive scrubbing may be required to decontaminate aluminum components.

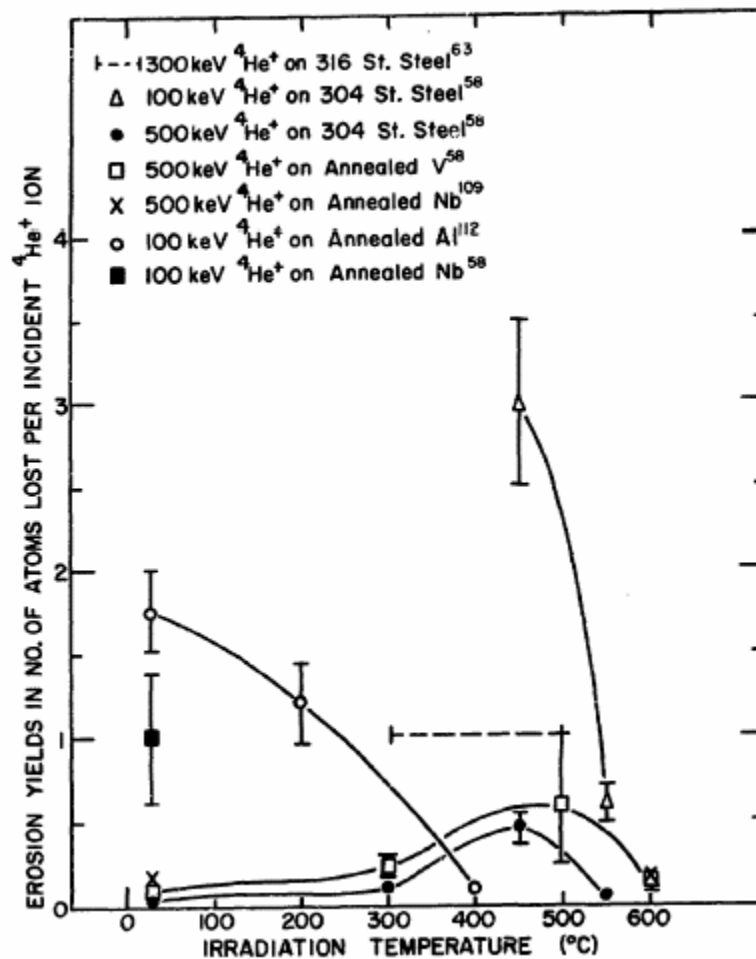


Figure 10: Erosion rates for different metals and alloys as a function of irradiation temperature for different projectile energies and total doses.³²

5.0 PU-238 THERMAL RADIATION AND PREDICTED TEMPERATURE

Pu-238 is an alpha emitter with a relatively short half-life, making alpha heating a problem for the storage and handling of large amounts of this isotope. Gram quantities of Pu-238 are said to generate enough heat to melt plastic bags.¹ Large quantities must be handled with insulated gloves, and special precautions must be taken to ensure a good thermal heat sink during shipping and storage. The surface temperature of ²³⁸PuO₂ particles can be conservatively estimated by performing an energy balance on spherical particles of various diameters. The energy balance can be summarized as:

$$\begin{array}{l} \text{Heat gained} \\ \text{from radioactive} \\ \text{decay} \end{array} = \begin{array}{l} \text{heat lost} \\ \text{by radiation} \end{array} + \begin{array}{l} \text{heat lost} \\ \text{by convection} \end{array} + \begin{array}{l} \text{heat lost} \\ \text{by conduction} \end{array}$$

Losses due to convection and conduction can be neglected to estimate a conservative bounding temperature. Radiant heat transfer is relatively easy to calculate and is summarized by the equation,

$$q = \sigma (\epsilon T_1^4 - \alpha T_2^4)$$

where q is the heat flux (heat/unit area), ϵ is the emissivity and α is the absorptivity (experimental parameters)³³, σ is the Stefan–Boltzmann constant ($5.67 \times 10^{-8} \text{ J/s m}^2 \text{ K}^4$) and T is temperature (K). The emissivity is the ratio of energy radiated by a particular material to energy radiated by a black body at the same temperature. The absorptivity is defined as the fraction of incident radiation that is absorbed by a surrounding body. The surface area of a spherical Pu oxide particle is assumed and an emissivity of 0.8 is assumed for Pu oxide.³³ Using the Kirchoff's Law as an approximation for the absorptivity of surrounding surfaces (i.e., $\alpha = \epsilon$)³⁴, the temperature required for the surface to radiate the decay heat is calculated as a function of particle diameter (see Figure 11). In this model, the temperature is determined by the overall thermal loading present as a result of alpha decay (i.e., 0.57 watts/g) for a particle with a density equal to that of fully dense ²³⁸PuO₂ (i.e., 11.46 g/cm³). This model neglects convection and conduction, and hence, is conservative. As is evident, the particle surface temperature begins to rapidly increase as the particle size increases above 50-100 μm . The actual results would exhibit lower temperatures if convection and conduction with surrounding components were considered.

6.0 EFFECTIVENESS OF DECON GEL STRIPPABLE COATINGS FOR REMOVAL OF PU-238 OXIDE

Conventional decontamination, fixation and encapsulation techniques were assessed and summarized in a previous review.³⁵ Advantages, disadvantages and recommendations were made for decontamination of 235-F. These techniques were noted as having the disadvantage of being labor intensive and difficult to apply to all areas of a glovebox or shielded cell environment. In addition, newer formulations of decontamination agents and fixatives have been developed since the issuance of the report. Given the current data known about the ²³⁸PuO₂ contamination in the 235-F facility and the conclusions drawn in this technical report, it can be suggested that a strippable coating, such as Decon Gel by Cellular Bioengineering, Inc., (CBI) would be preferable for decontamination of the particles within the facility. Based on the discussion presented in this report, it can be assumed that

most or all of the particles have settled from the air due to water adsorption and agglomeration and would be present on the surfaces contained in the cells.

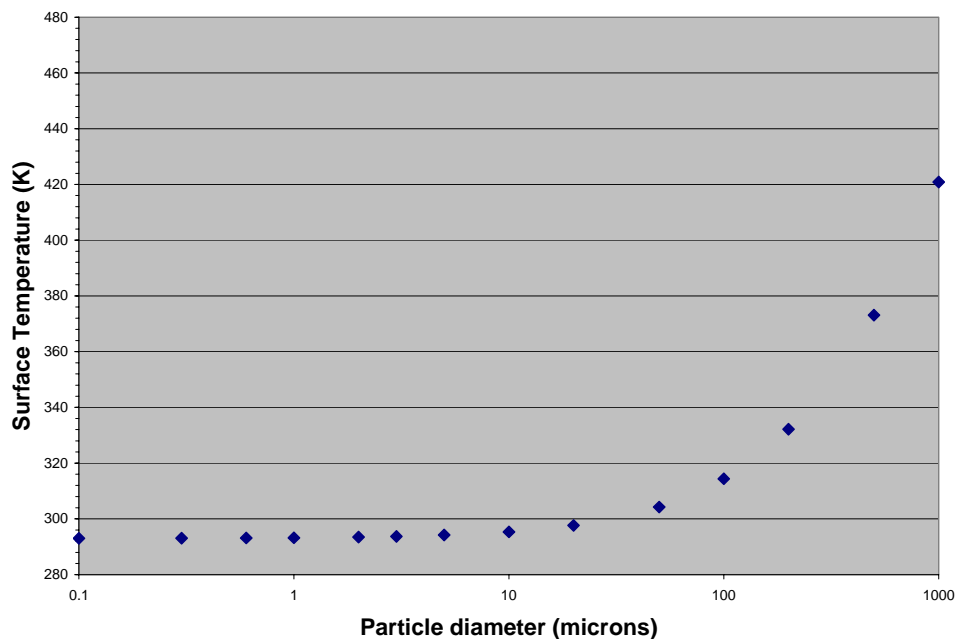


Figure 11: Surface Temperature Required to Maintain a Constant Temperature by Radiant Heat Transfer

The Decon Gel product can be obtained from Cellular Bioengineering, Inc. in a purely water-based form or with added ethanol to decrease drying times. Both grades will be tested at SRNL before a decision can be made as to their use. The gel consists of the water/ethanol solvent mixture, water-soluble polymer gel, a chelating agent, a surfactant and other small amounts of organic additives. The ethanol content (i.e., 6%) may present some challenges to meet the waste acceptance criteria, but calculations of the volatiles emitted from such a mixture may prevent such issues. The water/ethanol azeotrope and likely waste storage conditions must be taken into account.

The Decon Gel strippable coating has been used successfully at SRS in H canyon on tanks 6C and 6E on various surfaces that had been decontaminated previously with radiac wash. Some areas showed minimal improvement, while others presented complete (100%) decontamination.³⁶ This product has also been used by other facilities with limited success for decontamination.^{37, 38}

The issues of using such a strippable coating include surface heating of the particles, radiolysis of the water contained in the gel, and waste storage. Heat generated by small (< 1 micron) particles should not present an issue for use with this gel, although very large particles may degrade the gel and reduce its effectiveness. Conversely, particle self-heating may decrease the gel drying time and drive off any volatiles in the product before waste storage. Water radiolysis will most likely produce hydrogen (and oxygen) gas, and further calculations are necessary to determine if this will present an issue for waste storage.

With respect to the product itself, water already present on the particle surface will have a strong affinity for the water-based gel and should increase wettability of the Pu-particles, thus increasing product effectiveness. It is recommended that testing of this product in a small contaminated area,

such as a glovebox, be performed to assess the performance of this product before large-scale use in building 235-F.

7.0 SUMMARY

The physical, chemical and radiological properties of $^{238}\text{PuO}_2$ materials that were processed in the Plutonium Fuel Form Facility (PuFF) in building 235-F at the Savannah River Site were reviewed. A better understanding of the behavior of $^{238}\text{PuO}_2$ is needed to inform the planning process for the PuFF D&D project at the 235-F facility. The properties, as they pertain to D&D activities, are summarized in this report.

The current work focuses on the properties of $^{238}\text{PuO}_2$ particulates, in relation to its handling, containment, stability and airborne release to the environment during D&D activities. Several items of interest were discussed from legacy reports from PuFF operations and previous studies on radioactive contamination. In particular, the spread of $^{238}\text{PuO}_2$ contamination was presented as difficult to contain and adversely impacted operations. Previously, this was attributed to several factors characteristic of $^{238}\text{PuO}_2$:

- the extremely fine particle size distribution,
- high specific activity of Pu-238,
- the inability of fines to agglomerate (and thus settle out of air),
- alpha recoil driven decrepitation,
- Pu-238 penetrating into substrates causing internal contamination
- the self-heating nature of the oxide particles.

These attributes of $^{238}\text{PuO}_2$ are briefly discussed and their relevance to D&D activities is summarized. The extremely fine particle size of ball milled powders is expected to be a legacy of operations and a high contamination level is expected. However, the decay heat exhibited by these fines is insufficient to heat them up more than a few degrees above ambient temperatures. In addition, the dry environment used during operations no longer exists. Therefore, it is anticipated that these powders would have some adsorbed water vapor on their surfaces and would likely agglomerate and settle out over time. Alpha recoil driven decrepitation may occur and hinder the containment of these particulates or their capture on filter media as has been shown in previous studies.

The Decon Gel strippable coating was suggested as a possible alternative to other decontamination techniques because it may offer significant advantages in its application to conventional liquid decontamination solutions. However, it is recommended that testing of this product in a small contaminated area, such as a glovebox, be performed to assess its performance before large-scale use in building 235-F.

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