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Pseudo-Evaporation of High Specific Activity Alpha-Emitting Materials

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Abstract:

The purpose of this paper is to document a little known physical phenomenon that is unique and particular to high specific-activity actinides. It is part of the folklore among those who work with high alpha-emitting materials that they can self-levitate and migrate as a sort of aerosol, what we call pseudo-evaporation. This is an inquiry into the possible mechanisms of such migration of alpha-emitting species. High specific-activity alpha emitting isotopes that are solids but are somehow, due to the emission of high energy alpha particles, given a recoil kick that can lift them away from a surface into a moving air stream, which would then carry them away.

This paper presents a way to estimate the magnitude of such migration and, perhaps, to achieve enough understanding to allow improving safety procedures when working with high specific-activity actinides such as plutonium-238, polonium-210, and curium-244. An equation is derived giving the rate of pseudo-evaporation of $^{238}PuO_2$. It is concluded to be a function of the material's specific activity to the power of 2. Assuming a cluster of plutonium oxide molecules, the rate of pseudo-evaporation for an isotopically pure $^{238}PuO_2$ cluster would be 1E+03 psuedo-evaporations per second per cm^2, greater than for pure $^{239}PuO_2$ by a factor of about 10^5.

The same phenomena that cause pseudo-evaporation may contribute to the enhanced chemical reactivity of plutonium-238 metal.

Introduction:

The purpose of this paper is to document a little known physical phenomenon that is unique and particular to high specific-activity actinides. There has long been folklore among those who work with high alpha-emitting materials that they can self-levitate and migrate as some sort of aerosol. A particularly difficult material is polonium-210 with its

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half-life of 138 days, plutonium-238 has a half-life of 88 years, and curium-244 with a half-life of 18.1 years. High specific-activity actinides such as ^{210}Po, ^{238}Pu, and ^{244}Cm have the reputation of migrating throughout a glove box as a result of apparent volatility even when the compounds under study would not be expected to be volatile [1]. This phenomenon has been called "fleas" and "ghosting." Similarly, such phenomenon has been observed during shipments, where a clean surface is found to be contaminated upon receipt. This inquiry addresses the issue with the intention of applying its conclusions to the handling of ^{238}Pu, ^{210}Po, and ^{244}Cm. ^{238}Pu has a much lower specific-activity than ^{210}Po and ^{244}Cm, but still sufficiently high in comparison with ^{239}Pu to make its potential for migration to be of interest.

At the outset of this inquiry, it should be made clear that alpha-emitters are susceptible to migration related to daughter product recoil, but beta-emitters are not. This is because the energy imparted to the daughter of an alpha decay may be up to 100 keV whereas the energy imparted by a beta decay can only be in the few eV range.

Alpha recoil migration should also be distinguished from the migration of radon, which is a gas. Polonium is an element, which, although not volatile at room temperature, can form compounds that are volatile in some environments and could migrate as a gas. The focus of this paper, however, is a phenomena of high specific-activity alpha emitting isotopes which remain solids but are somehow, due to the emission of high energy alpha particles, are given a recoil kick that can lift them away from a surface into a moving air stream, which would then carry them away.

Such migration has been part of the folklore, as remarked above. Some have suggested that the observed migration is only a manifestation of poor housekeeping, which is more easily observed when working with high specific-activity isotopes. There are, however, enough controlled observations and anecdotes about migration to cause us to suspect this as the explanation. Earlier presentations by this author at the Actinide Separations Conference and Plutonium Futures conference highlighted this phenomenon. The program and facility management of the LLNL Heavy Element Facility (B251) were well aware of this phenomena prior to embarking on the Risk Reduction Program and planned for it. Extensive training was therefore conducted, as discussed in the other portions of this best practice paper.

Another explanation for this phenomenon is that the high alpha-emitting particles build up a helium concentration in the near-surface region of a solid material and the gas pressure from accumulated helium pops off material. If this were the mechanism, we would expect to see fracturing through the bulk of such materials; but we do not observe this fracturing. The author believes that this is not the case.

Assuming our argument above, that alpha-emission can give rise to migration of high specific-activity actinides, it becomes an important topic for actinide research. Alpha-emission and actinides hold much common ground. Of the naturally occurring alpha-emitters, almost all have an atomic number above 83 (bismuth) and among the actinides themselves, elements 89 – 103, a good two-thirds of their known isotopes are alpha-emitters [2]. The energies of the alpha-decays span from about 4 MeV (4.08 MeV for ^{232}Th) to above 7 MeV for some short-lived isotopes.

Recoil energies

The recoil formulas used in this paper are derived in Appendix A. In the case of ^{238}Pu decay we have the formula,

$$^{238}\mathrm{Pu} \quad \rightarrow \quad ^{234}\mathrm{U} \; + \; ^{4}\mathrm{He} \; + \; 5.50 \text{ MeV}, \tag{1}$$

and the recoil energies of the alpha particle and the uranium daughter product will be

$$E_{\alpha} = 5.41 \text{ MeV, a weighted average} \tag{2}$$

$$E_{U} = 94 \text{ keV, the recoil energy of the } ^{234}\mathrm{U}. \tag{3}$$

It is important to understand the difference between a typical beta-decay and an alpha-decay, which is the subject of our interest in this paper. The mass of the beta-particle is only 1.37 10^{-4} of the alpha-particle and therefore imparts much less recoil energy to the daughter nucleus in a beta decay event. Thus, in the case of ^{90}Sr decay to yttrium,

$$^{90}\mathrm{Sr} \quad \rightarrow \quad ^{90}\mathrm{Y} \; + \; ^{0}\beta \; + \; 0.546 \text{ MeV}, \tag{4}$$

the recoil energy of the daughter atom, ^{90}Y, is 3.3 eV. This is a recoil energy characteristic of the decays of many fission products. Comparing this with alpha decay brings out the important point that the recoil energies for most fission product decays are about four orders of magnitude smaller than for the alpha decays. Thus, pseudo-vaporization of fission products will almost always be negligible while it can be significant for high energy alpha-emitters (i.e., high specific-activity actinides).

This paper argues that the decay-induced ejection of material from a surface (pseudo-evaporation) is related to the energy deposited around the site of the decay and that is related to the recoil energy of the daughter atom. The alpha range in PuO_2 is about 10 μm ($1\text{x}10^{-3}$ cm), but the ^{234}U recoil range in Pu or PuO_2 is about 8 – 10 nm ($8\text{x}10^{-7}$ cm). The recoil creates a cascade of secondary and later knock-on atoms filling a volume of hot material. The heat will diffuse away and while the affected volume increases with time, its average temperature decreases as the energy becomes spread through a larger volume.

Implications:

This paper argues that high energy alpha-emission can give rise to material transport. To the extent that such pseudo-evaporation is true, we need to ask questions about the hazards in handling high alpha specific-activity isotopes. Escape from the surface of a radioactive source can lead to transport to other surfaces. This active material be resuspended and travel further. Questions for consideration include:

- Can high specific-activity isotopes burn through gloves?
    - Answer: This phenomena has been repeatedly observed at different sites in the DOE Complex. Heavy Element Facility management had personally witnessed this phenomena earlier in their career with frustrating needs to change gloves often, e.g., after lunch. The solution was to double glove, i.e., put surgeon gloves on top of glovebox gloves and to change the surgeon gloves frequently.
- Can migration allow high specific-activity isotopes to escape from small leaks in a glove box? For example, could small leaks that would pass unnoticed when handling weapons grade Pu provide a path for significant amounts of ^{238}Pu?
    - Answer: See below.
- Can high specific-activity isotopes pass through HEPA filters by becoming resuspended from surfaces on which they were first captured?
    - Answer: In work at multiple facilities across the DOE Complex with all of these high-specific activity actinides, this has not been observed.
- Can recoiled material become embedded in surfaces and escape at a later date, after the object (e.g., glovebox, shipping container) has been converted to other uses or decommissioned?
    - Answer: This has been observed repeatedly.
- Will recoil fragments adhere to dust already present? Could general dust-suppression inhibit high specific-activity alpha-emitting isotope migration?
    - Answer: It has been observed that dust suppression practices (e.g., stripcoating) inhibit migration. This is discussed in other portions of this best practice paper.

We do not address all of these issues in detail in this paper, but they suggest the relevance of this study to phenomena associated with high specific-activity actinide work.

The "pseudo-evaporation" levitation mechanism:

A first intuitive explanation is often offered for pseudo-evaporation: particles on a surface are driven upward from that surface by the emission of a high energy alpha-particle ejected in a downward direction, like a rocket take-off. Paraphrasing Newton, each force has an equal and opposite force. A few atoms somehow enclose the fast-moving uranium daughter and ride with it as a small particle, a cluster of atoms. Detailed examination, however, shows that is not physically plausible. In oxide materials, the recoil nucleus, ^{234}U produced by the decay of ^{238}Pu, would have to transfer its momentum to the rest of the oxide particle without shattering it. Its initial kinetic energy is 94 keV, which is about 10^4 times greater than crystal lattice binding energies. Thus, there appears to be no mechanism whereby the original recoil nucleus could link its momentum with a cluster of surrounding atoms in the ways that would be required. The rocket “propellant” would blast the rocket apart rather than propelling it.

If we were to go ahead nevertheless and assume that the oxide particle could hold together, the sizes of the ejected particles could be estimated using data in Table 1. This lists what the kinetic energies of several different masses of oxide particles would be in recoiling from an alpha-emission. It assumes that all the molecules in the oxide particle as a whole share the momentum imparted from the alpha-emission. Oxide particles would carry the same momentum as the emitted alpha, but their kinetic energy, which depends on velocity (momentum) squared, decreases for the heavier particles. It can be seen that for a 10 nm particle (which would contain about 1.3×10^4 molecules of PuO_2) the kinetic energy would be about 6 electron volts. This would be enough to break a few chemical bonds and allow the oxide particle to detach from a surface. Larger oxide particles would not have sufficient energy to disconnect themselves. Only smaller ones could theoretically be set free.

Table 1. Recoil oxide particles – kinetic energies and velocities: In this table the recoil material particle kinetic energy, Er, is expressed in two ways, units of eV, electron-volts, and ergs. The velocity of the recoil, Vr, is expressed in cm/s.

Diam. – cm	**1.E-02**	**1.E-03**	**1.E-04**	**1.E-05**	**1.E-06**	**1.E-07**
	100 μm	10 μm	1 μm	100 nm	10 nm	1 nm
Er – ev / particle	6.11E-12	6.11E-09	6.11E-06	6.11E-03	6.11E+00	6.11E+03
Er – ergs / particle	9.73E-24	9.73E-21	9.73E-18	9.73E-15	9.73E-12	9.73E-09
Vr – (recoil velocity) cm/s	1.81E-09	1.81E-06	1.81E-03	1.81E+00	1.81E+03	1.81E+06

Material levitated by effects of a thermal spike:

A more realistic picture is that the alpha recoil creates a cascade of secondary and later knock-on atoms [3]. We assume that they will raise a small volume of material to some elevated temperature, see Fig. 1, which depicts what might happen if they were within a surface region and could eject material away from the surface. If thermal spikes are generated below a surface but near to it, it has been suggested that a shock wave from the event could reach the surface and spall off material. The recoil of the daughter atom will create a region at high temperature. This may boil off material if right at the surface, or if it is within the solid, it may create a shock wave that can spall material from a nearby surface. This is also depicted in Fig. 1.

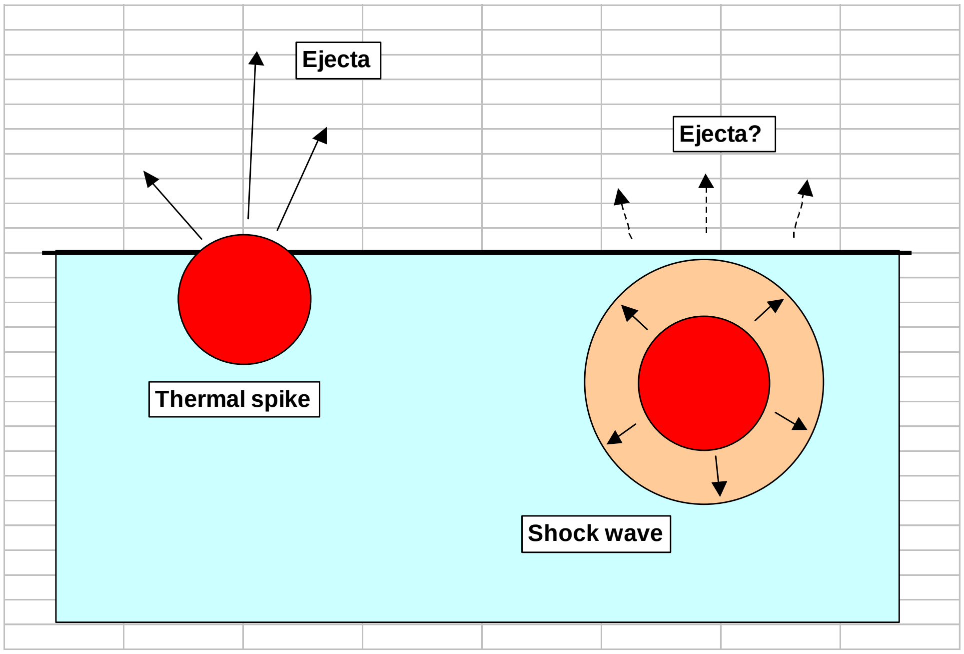

Fig. 1: Consequences of alpha decay within a solid but near its surface.

In any event, the heat will diffuse away from the central region and therefore, while the affected volume increases, its average temperature decreases. If we consider the first knock-on atoms, we can estimate that the 94 keV ^{234}U recoil energy, if dissipated as heat along its path length of about 10 nm, would create a volume of 1E-18 cm^3, and would raise its temperature to around 8580 K, a temperature at which the oxide would vaporize, see Table 2. If this event occurred near the surface so that this material could escape, it appears that it would be in the form of vapor, not droplets or coherent fragments. Larger thermal spike volumes would emit material at lower temperatures. This realization directs us to examine release mechanisms based on a picture of radiation damage in near-surface areas and to try to derive a pseudo-vaporization relationship on that basis.

Table 2. Thermal pikes: Thermal spike temperatures, K, for ^{238}Pu decay where the recoil energy is 94 keV and assuming recoil volumes equal to the cube of the diameters.

Diameter – cm	**1.E-02**	**1.E-03**	**1.E-04**	**1.E-05**	**1.E-06**	**1.E-07**
Diameter – µm/nm	100 µm	10 µm	1 µm	100 nm	10 nm	1 nm
Volume – cm^3	1E-06	1E-09	1E-12	1E-15	1E-18	1E-21
Temp. rise - K	5.12E-07	5.12E-04	8.58E-03	8.58E+00	8.58E+03	est 1E+04

Another ejection mechanism has been proposed to occur in electrically insulating materials. The ionization along both the alpha path and in the recoil region will kick electrons away, see Fig. 2. Before these electrons have a change to migrate back to the core region, a slow process in insulators, the unbalanced positive charge would disassemble itself due to electrostatic repulsion. The resultant forces could drive material away from the ionized region as a shock wave, which on reaching the surface would spall material. It has been suggested that in electrically insulating materials the alpha particle (moving to the lower left in this figure) and the daughter recoil (moving toward the upper right) may create an electrical imbalance. Electrons along the path would be ejected away leaving a core with positively charged ions. These ions would repel one another, explode, and generate a shock wave, which could spall material from the surface.

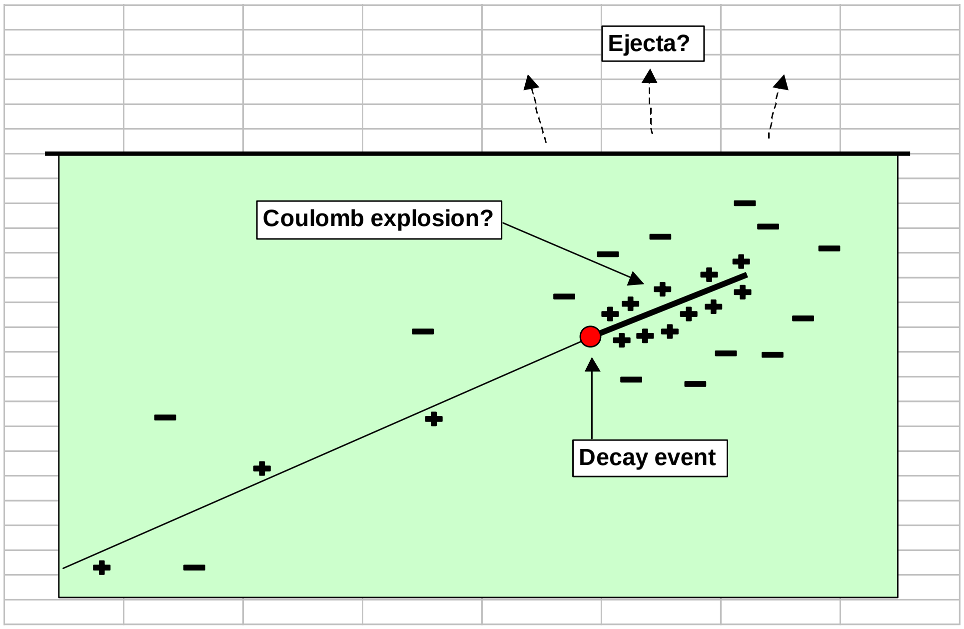

Fig. 2: A coulomb explosion may eject material from a surface.

In summary, there are several possible mechanisms for the ejection of material from surfaces following alpha-decay:

- Material will be ejected from the thermal spike region if it reaches the surface.
- The sudden thermal expansion in a thermal spike below the surface will initiate a shock wave that can spall surface material.
- In oxides or other materials having low electrical conductivity, electrostatic forces in a decay region could initiate shock waves that could spall material.

The theories of damage mechanisms within the interior of materials have been explored [3] but in general, near-surface mechanisms have not been extensively explored, although the consequences of a fission event near a surface have been computed [4]. Nevertheless, we will assume that one or more of such events may serve to eject particles and will calculate what the ejection rates might be.

A pseudo-evaporation formula:

A generic formula can be derived for the rate of ejection of material from the surface of a material. The values for the terms will be discussed. In the following discussion we will picture the volume as a cube for simplicity, although computer calculations show that it will be irregularly shaped [3].

First, we may express the frequency of decay events, dn/dt, as being proportional to the density of activity within the material, its specific activity, S – disintegrations per second (dps)/gram, times the material density, ρ, and the depth within which this activity can yield emitted material from the surface – the thickness, L.

$$dn/dt = S\,L \qquad \text{Ejection events cm}^{-2}\text{sec}^{-1}, \tag{5}$$

Where S is the specific activity, disintegrations cm^{-3} sec-1, and L is the thickness, cm.

It seems reasonable to assume that the volume of material ejected per event, Vr, will be a function of the energy, Er, imparted to the recoil daughter atom. There will be a weighting factor, g. Combined in this term will be the efficiency of the ejection process and an accounting of those decays which occur very close to the surface and those at depths from which the thermal spike region just barely extend to the surface. The weighted volume can be described as

$$Vr = g\,Er \tag{6}$$

$$Vr = L^3$$

or

$$L = (g\,Er)^{1/3} \tag{7}$$

The dimensions of g are $\text{cm}^3\ \text{eV}^{-1}$.

Then, the activity associated with the material ejected in each event will be

S, the activity associated with the material ejected, disintegrations $cm^{-3}sec^{-1}$ (8)

Combining (5), (6), and (7) gives the flux of activity ejected per unit area

$$J = dn/dt * Vr * A \tag{9}$$

$$J = (S L) * (g Er) * (S)$$

or

$$J = (g Er)^{4/3} S^2 \tag{10}$$

Note that the units here are

$$J = (cm^3 eV\text{-}1 * eV)^{4/3} (dis. cm^{-3} sec^{-1})2 = dis. cm^{-2} sec^{-2}, \tag{11}$$

which says that the material is ejected at a rate measured in units of cm^{-2} sec^{-1}has a disintegration rate in units of dis. sec^{-1}.

This equation makes three interesting points:

- The pseudo-evaporation, dps per cm^2 per second, will be proportional to the square of the specific activity.
- The pseudo-evaporation will depend on the recoil energy, Er, to the power of 4/3.
- The factor, g, depends on the assumptions made about the ejection mechanisms and its value will affect the prediction of ejection rates.

As an aside, there is a relationship between the alpha recoil energy, which in turn depends on the decay energy, and the specific activity of alpha-emitters, the Geiger-Nuttall relationship [5]; the former increases as the latter increases. This will be mentioned for the two Pu isotopes in Table 4. For other actinides where the decay energy reaches values near to 7MeV, dependence of psudeo-evaporation on energy may contribute more to an enhanced ejection rate. It might also be the source of variability in the factor, g, with decay energy.

If we want to venture into making estimates of what J might be we need to propose a value for g. To do this, we go back to equation (6), Vr = g Er. Assuming this volume to be that amount of material, which would reach a condition that would cause it to be ejected from the surface. We might, as a first approximation, assume that this effective volume would be that volume that would reach an average temperature approaching the vaporization temperature of the material.

Taking the data from Table 2 suggests that a volume of $(20\ nm)^3$ would reach a temperature of roughly 1000 K. If the material were plutonium oxide with any adsorbed moisture this temperature would cause fragmentation and ejection of material. The associated volume would be 8E-18 cm^3. Then, the derived value of the g-factor for this case would be

$$g = 8E\text{-}18\ cm^3 / 9.38E{+}04\ eV = 1E\text{-}22\ cm^3 / eV \tag{12}$$

And using this in equation (11) for $^{238}PuO_2$ gives

$$J = 1E{+}03\ \text{dps ejected}\ cm^{-2}\ sec^{-1}. \tag{13}$$

On the other hand, for $^{239}PuO_2$

$$J = 8E\text{-}03\ \text{dps ejected}\ cm^{-2}\ sec^{-1}. \tag{14}$$

From inspection of equations (7) and (10), it appears that these conclusions are not strongly dependent on the assumptions of temperatures reached or volumes involved. Thus, these values would appear to be approximately what might really occur.

Character of the "pseudo-evaporated" material and possible resuspension:

The pseudo-evaporation calculation provides little guidance on what particle size distributions or textures should be produced. Table 3 provides some context in which to think about the properties of a range of sizes of particles. We do this because we are interested in the material's transport through the air, adhesion to surfaces, and possible resuspension. The table shows, for example, that in the case of $^{238}PuO_2$ particles, the specific activity is such that a 10 nm diameter particle would experience about 1 atomic disintegration per 2.67E+05 seconds, or every three days. Thus, the frequency of atomic decays within such a particle may have some effect on the stability of the particle, its ability to hold together, migration, adhesion to other airborne particles, and adhesion to surfaces.

We cannot, however, analyze such phenomena one particle at a time. If a cluster of particles are captured by a filter, for example, the alpha decay induced charging of the particle may affect the adhesion of individual particles to capture surfaces.

Another consideration is that the particles are likely to not be spherical or compact material. The effective density of a feathery particle would be much less than for a compact sphere with the consequence that such low density particles can be carried farther in moving air than simple calculations would indicate.

Electrostatic factors need to be taken into account also. A variety of mechanisms might lead to a particle having a net electrostatic charge after an alpha-decay event has occurred within it. Also, the thermal heating of the oxide particle may generate thermionic emission of electrons leaving the particle with a net positive charge. The ionization of the atmosphere will also be important. So, depending on the details of the process, the residual charge might be positive or negative. Such a net charge might hinder particle coalescence or settling under some circumstances. Decay events may also fragment particles.

Table 3: Properties of $^{238}PuO_2$ particles: Particle size, contained activities, and aerodynamic properties.

Diameter – cm	**1.E-02**	**1.E-03**	**1.E-04**	**1.E-05**	**1.E-06**	**1.E-07**
Diameter µm/nm	100 µm	10 µm	1 µm	100 nm	10 nm	1 nm
Mass – grams	5.97E-06	5.97E-09	5.97E-12	5.97E-15	5.97E-18	5.97E-21
Molecules PuO_2 per particle	1.33E+16	1.33E+13	1.33E+10	1.33E+07	1.33E+04	1.33E+01
Activity- dps per particle of material	3.74E+06	3.74E+03	3.74E+00	3.74E-03	3.74E-06	3.74E-09
Can alpha escape? (range is 10 µm)	**no**	**No**	**yes**	**Yes**	**yes**	**yes**
Can U-recoil escape? (range is 10 nm)	**no**	**No**	**no**	**No**	**no**	**yes**
Max energy into oxide particle for each decay – eV	5.59E+06 alpha + recoil	5.59E+06 alpha + recoil	9.38E+04 recoil	9.38E+04 recoil	9.38E+04 recoil	(small) part of recoil
Time between decays – sec	2.67E-07	2.67E-04	2.67E-01	2.67E+02	2.67E+05	2.67E+08
Stokes settling rate in air – cm/sec	1.03E+03	1.37E+01	1.60E-01	4.10E-03	3.42E-04	3.42E-05
Brownian Diffn Coef – cm^2/sec	2.40E-09	2.60E-08	2.90E-07	7.00E-06	5.00E-04	6.00E-02

The effects of high specific-activity alpha-emission on chemical reactivity:

A further effect of high energy alpha-decay and especially the thermal spikes in the near-surface regions of materials is likely to be a chemical modification of those surfaces. For example, PuO_2 is the oxide normally seen on the surface of plutonium metal. It acts as a protective layer on the metal and inhibits further oxidation. If the metal is heated to about 200°C, this oxide reacts with the metal to form the lower valence oxide, Pu_2O_3, which is a catalyst for metal reactions with air and hydrogen [6].

$$3\,PuO_2 + Pu \rightarrow 2\,Pu_2O_3 \qquad (15)$$

It seems probable that this reaction can occur on ^{238}Pu metal, and that would help explain its observed high chemical reactivity. We can estimate the rate of this process as follows. Note that the energy of the ^{234}U recoil is 9.38E+04 eV. The energy from each alpha recoil will spread over a larger and larger volume within a sample of metal with time as the heat diffuses away from the recoil cascade. The average temperature within this expanding volume will decrease. For this calculation we will pick a volume such that the energy has raised the temperature to 200°C. The specific heat of Pu metal is 3.61E-02 cal/deg/gram which translates into C=1.87E+19 eV/deg/cm^3.

If the temperature rises from 20°C (room temperature) to 200°C (at which the reaction, Equation (15), takes place) the volume that includes this elevated temperature Pu metal will be 2.78E-17 cm^3. Now, the area that will be affected by such volumes is 1.88E-06 cm^{2}sec^{-1}. That is, thermal spikes near the surface will heat up the surface and this small fraction of the surface each second. As a year is 3.16E+07 seconds long, it follows that in a year's time, just about all of the surface will be heated, pulse by pulse, to a high enough temperature to initiate the reduction of PuO_2 to the lower oxide and make the surface vulnerable to rapid oxidation.

The enhanced chemical reactivity of high specific-activity actinide metals and some other compounds might be a factor in producing fine grained airborne release fractions. This would be another contributor to the migration of such materials. However, the products of such reactions and plutonium dioxide remaining after complete metal-air reactions would not be capable of further chemical reaction. The further migration of such material would need to be explained as a process of resuspension of the oxide particles.

Discussion:

We can use the equation 10 to derive properties for two plutonium isotopes. Table 4 assumes pure ^{238}Pu and ^{239}Pu. The specific-activity ratios for materials used in practice are usually lower due to the enhancement of ^{239}Pu activity by the addition of the shorter half-life ^{240}Pu and the diminution of ^{238}Pu activity by dilution with other, longer-lived Pu isotopes. The values for R assumed in these calculations is 8 nm.

Table 4: Pseudo-evaporation rates due to alpha-decay: Two isotopes of plutonium are compared.

	^{238}Pu	^{239}Pu	Ratio
Half-life (years)	87.7	24,100	
Specific activities Ci/gram	17.1	.0619	276.3
Energy of alphas, MeV, weighted average	5.486	5.144	1.070
Energy of recoil, MeV, weighted average	9.38E-02	8.76E-02	1.071
Pseudo-evaporation rate dps / cm2 / sec	1.0E+03	8.0E-03	1.3E+05

Since the rate of pseudo-evaporation depends on the square of the specific activity, it appears that the ratio of ^{238}Pu to ^{239}Pu rates is about 10^5. This explains why migration of high specific-activity alpha-emitters can be observed whereas that of low specific-activity emitters is not.

Conclusions:

Pseudo-evaporation, migration, and enhanced chemical activity are phenomena of all high energy alpha-emitters and thus the majority of the actinides. This phenomena is not significant among beta-emitters and thus not important for most of the fission products.

The mechanism of pseudo-evaporation is proposed to be the localized thermal heating of material, thermal spikes, initiated by the recoil daughters of the alpha-decay near the surface of a solid. The temperatures in volumes having edge dimensions of around 10 to 20 nm can reach average temperatures of thousands of degrees K, greater than the melting or even the vaporization temperatures of the material. If these regions are within a distance of the surface less than the dimensions of the high temperature volume, it appears reasonable to assume that they will be ejected from the surface.

The magnitude of this phenomenon will depend on the square of the specific activity of the isotope. For half-lives greater than a few thousand years, it will not normally be an issue. For half-lives shorter than a hundred years (e.g., ^{238}Pu, ^{210}Po, and ^{244}Cm), it can be a significant factor requiring attention in controlling the spread of radioactive contamination.

The release rate from $^{238}PuO_2$ is calculated to be 1.0E+03 dps/cm^2/sec. The rate from $^{239}PuO_2$ it is estimated to be 8E-03 dps/cm^2/sec, smaller by a factor of about 10^5.

It is hard to estimate the particle size distribution of the ejected material. This paper provides tables showing aerodynamic properties of particles likely to lie within the range, but it does not delve into the issues of practical settling rates and resuspension rates.

Another consequence of the thermal spike phenomenon near surfaces is likely to be increased chemical reactivity at the surface. In the case of ^{238}Pu metal this seems likely to reduce PuO_2, a protective oxide layer, to Pu_2O_3, a material that catalyzed the metal's reactions with oxygen and hydrogen.

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Appendix A: Daughter atom recoil equations

The formulas for conservation of momentum when a radioactive disintegration occurs is

$$M_1V_1 + M_2V_2 = 0 \tag{1a}$$

where

M_1 and M_2 are the masses of the two particles which are ejected in opposite directions and V_1 and V_2 are their velocities. Conservation of energy states that the energy of disintegration, E_0, is shared between the two particles.

$$E_o = E_1 + E_2 \tag{2a}$$

$$M_o = M_1 + M_2 \tag{3a}$$

with

$$E_1 = ½\, M_1\, V_1^2 \quad \text{and} \quad E_2 = ½\, M_2\, V_2^2 \tag{4a}$$

Combining these equations gives

$$E_1 = E_0\, M_2 / (M_1 + M_2) \tag{5a}$$

$$V_1 = (2E_1 / M_0)^{½} \tag{6a}$$

In the case of ^{238}Pu decay, the decay energy is 5.498 (71% of the disintegrations) and 5.454 (28% of the disintegrations). For our calculations, we use the weighted average,

$$^{238}\text{Pu} \rightarrow {}^{234}\text{U} + {}^{4}\text{He} + 5.50\text{ MeV}, \tag{7a}$$

and applying equations (4) and (5) gives

$$E_\alpha = 5.41\text{ MeV}, \tag{8a}$$

$$E_U = 94\text{ keV, the recoil energy of the } {}^{234}\text{U}. \tag{9a}$$