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The Radiological Hazard of Plutonium Isotopes and Specific Plutonium Mixtures

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Edited by Ruth Barks, Group CIC-1

Figures by James M. Mahan, Group CIC-1

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George Heindel
James Clow
William Inkret
Guthrie Miller

Los Alamos
NATIONAL LABORATORY

Los Alamos, New Mexico 87545

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THE RADIOLOGICAL HAZARD OF PLUTONIUM ISOTOPES AND SPECIFIC PLUTONIUM MIXTURES

George Heindel, James Clow, William Inkret, and Guthrie Miller

Abstract

The US Department of Energy defines the hazard categories of its nuclear facilities based upon the potential for accidents to have significant effects on specific populations and the environment. In this report, the authors consider the time dependence of hazard category 2 (significant on-site effects) for facilities with inventories of plutonium isotopes and specific weapons-grade and heat-source mixtures of plutonium isotopes. The authors also define relative hazard as the reciprocal of the hazard category 2 threshold value and determine its time dependence. The time dependence of both hazard category 2 thresholds and relative hazards are determined and plotted for 10,000 years to provide useful information for planning long-term storage or disposal facilities.

BACKGROUND

A 1992 US Department of Energy (DOE) standard[1] defines hazard categorization for nuclear facilities. According to this standard, a DOE nuclear facility is designated as either a hazard category 1, 2, or 3 facility depending on the facility's potential for significant accidental consequences to workers, the public, or the environment. By categorizing a facility, DOE determines the level of sophistication required in the safety analysis report and in the level of review and approval by DOE.

The three hazard categories are as follows:

- Hazard category 1 facilities are those with "potential for significant off-site consequences." [1,3]
- Hazard category 2 facilities are those with "potential for significant on-site consequences." [1,3]
- Hazard category 3 facilities are those with "potential for only significant localized consequences [1,3]

DOE contractors compare the facility inventory to a threshold table to determine a preliminary hazard category for the facility; final hazard category determination occurs after more complete facility-specific hazard analysis. The DOE standard[1] provides threshold values for hazard categories both in mass units (grams) and activity units (curies). The specific activity (curies/gram) for each isotope is what relates the two values of mass units (grams) and activity units (curies).

The standard[1] lists hazard category 2 and 3 threshold values for a large number of radioisotopes. If the facility exceeds the hazard category 2 threshold and has not been designated a hazard category 1 facility by the program secretarial officer (PSO), then the facility is considered to be a hazard category 2 facility. A facility is hazard category 3 if its inventory exceeds the hazard category 3 threshold but does not exceed the hazard category 2 threshold. If a facility inventory is less than the hazard category 3 threshold, the facility may not be considered a nuclear facility. Hazard category 2 threshold values represent quantities of radionuclides that could result in a dose of 10 mSv (1 rem) 300 meters away if one uses standard air dispersion and dose calculations. Hazard category 3 values are derived from the Environmental Protection Agency definitions of reportable quantities for radionuclides published in 40 CFR 302.4, Appendix B. These values represent levels of materials which, if released, would produce 0.10-Sv (10-rem) dose, 30 meters away, based on a 24-hour exposure.

In previous work[2], the authors duplicated DOE methodology, found some errors in the DOE standard, determined hazard category 2 threshold values for additional isotopes, and published results in a LANL fact sheet.

For the purposes of this paper, we have defined the concept of relative hazard as the reciprocal of the hazard category 2 threshold value for that isotope or mixture.

METHOD

Plutonium Isotopes

The plutonium isotopes of interest are ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{244}Pu, each of which is the parent of relatively long series decay chains (Appendix A). Standard references provided the authors with the identity of the decay series and their half-lives. We calculated specific activities from the half-lives and took the available hazard category 2 threshold values from DOE-STD-1027-92. If the threshold values were not available, we calculated threshold activities using the methodology of the DOE standard (Appendix C). References 4 and 5 provided

committed effective dose equivalents (CEDE) and cloud shine dose equivalents (CSDE) for this calculation.

Using Bateman equations[6] (Appendix B), we calculated, as a function of time, the amount of each daughter present in each decay series. We then determined the hazard category 2 threshold (grams) as a function of time for a mass of material that was a pure plutonium isotope at time = 0, using the sum of fractions method described in Appendix B. This is a DOE-specified technique to account for the dose contribution of each member of a mixture of radionuclides.

By taking the reciprocal of this value, we determined the relative hazard of that material as a function of time (Figures 1, 2). All abscissa are time in years. Because spontaneous fission occurs a very small fraction of the time for ^{238}Pu, ^{240}Pu, ^{242}Pu, and ^{244}Pu, we considered neither the in-growth of fission fragments nor criticality in this analysis.

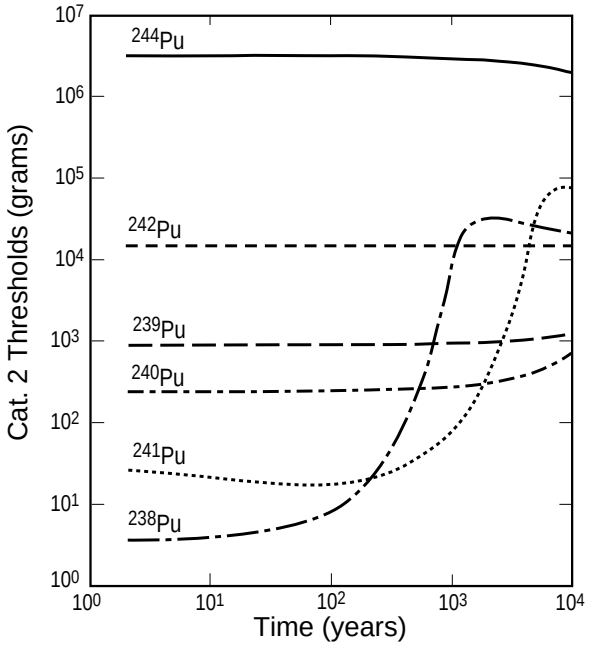

Figure 1. Category 2 threshold as a function of time starting with pure isotopes

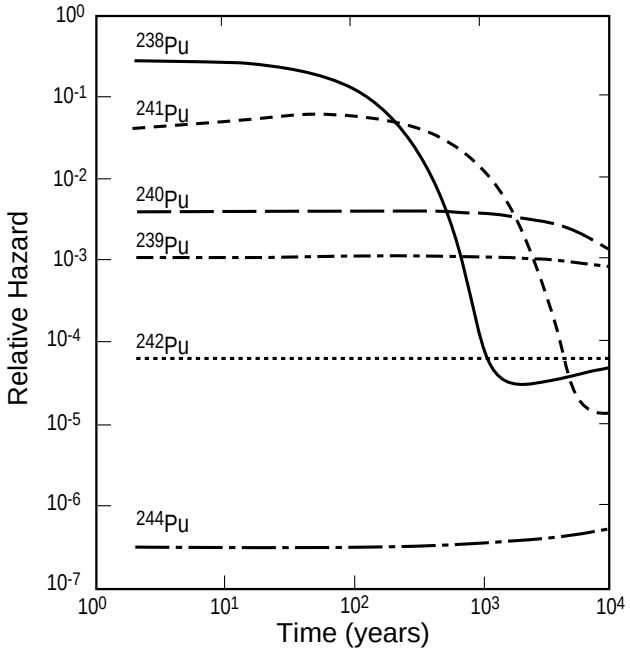

Figure 2. Relative hazard as a function of time starting with pure isotopes

Figure 2 does not make readily apparent the increase in hazard of ^{241}Pu during the first few decades.

Figure 3 better illustrates this, being the same graph of ^{241}Pu but with a linear instead of logarithmic relative hazard scale.

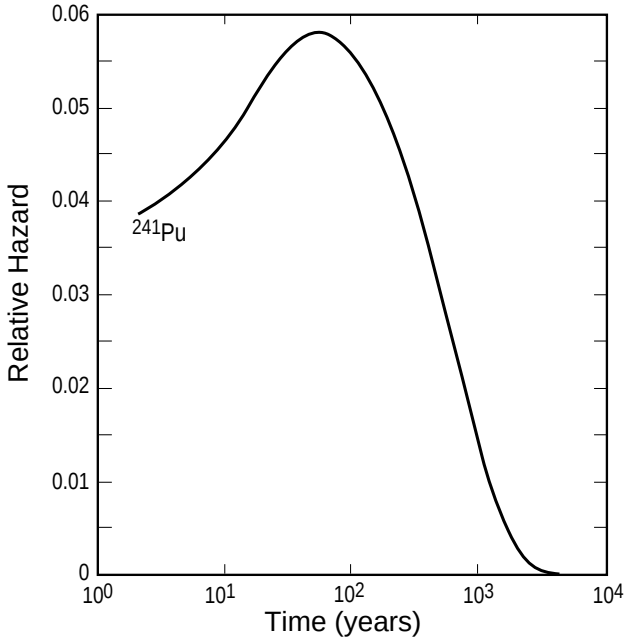

Figure 3. ^{241}Pu relative hazard as a function of time starting with pure isotope

Specific Plutonium Mixtures

Table 1 is a list of subject plutonium mixtures with their respective compositions. The Nuclear Materials Measurement and Accountability Group (NMT-4) of the Nuclear Materials Technology Division provided this data.

Table 1. List of subject plutonium mixtures with their respective compositions

Material Type	^{238}Pu (weight %)	^{239}Pu (weight %)	^{240}Pu (weight %)	^{241}Pu (weight %)	^{242}Pu (weight %)	^{244}Pu (weight %)
MT42 84%	1.02	1.37	10.32	3.13	84.14	0.02
MT42 90%	0.72	1.26	6.4	1.86	89.77	
MT42 95%	0.45	0.56	2.47	0.906	95.58	0.029
MT 51	0.006	96.77	3.13	0.076	0.018	
MT 52	0.01	93.78	6	0.2	0.02	
MT 53	0.03	91.08	8.45	0.366	0.071	
MT 54	0.046	87.42	11.5	0.81	0.22	
MT 55	0.06	83.88	14.73	1.03	0.304	
MT 56	0.061	81.9	16.51	1.18	0.355	
MT 57	0.433	74.63	20.7	2.55	1.69	
MT 83 83%	83.89	13.8	1.9	0.32	0.09	
MT 83 89%	89.26	10.07	0.633	0.021	0.015	

The DOE standard suggests that a facility processing weapons-grade plutonium should be classified based on the amount of dominant isotope, without specifying the amount of other trace isotopes involved. We thought it would be interesting to compare the category 2 thresholds using this "dominant isotope method" as contrasted with the calculated "sum-of-fractions method" described in Appendix B. Table 2 displays the results of this comparison.

Table 2. Comparison of category 2 thresholds using the "dominant isotope method"

Material Type	Cat. 2 Threshold by Sum of Fractions Method (grams)	Cat. 2 Threshold by Dominant Isotope Method (grams)	Ratio: Sum of Fraction to Dominant Isotope
MT 51	813	913	0.89
MT 52	730	913	0.80
MT 53	644	913	0.71
MT 54	542	913	0.59
MT 55	487	913	0.53
MT 56	463	913	0.51
MT 57	265	913	0.29
MT 42 84%	225	1.51E+04	0.015
MT 42 90%	333	1.51E+04	0.022
MT 42 95%	573	1.51E+04	0.038
MT 83 83%	4.29	3.63	1.18
MT 83 89%	4.03	3.63	1.11

Knowing the in-growth of daughter isotopes starting with pure plutonium isotopes and their respective category 2 thresholds, we applied these to the above mixtures to determine their category 2 thresholds and relative hazards as functions of time. (Figures 4–9).

RESULTS

Plutonium Isotopes (Figures 1–3)

- The relative hazard of ^{238}Pu decreases exponentially for approximately 2000 years. This exponential decrease is followed by a turnaround resulting from the in-growth of ^{234}U, ^{230}Th, ^{218}Po, ^{214}Po, and ^{210}Po. Three of these species, ^{218}Po, ^{214}Po, and ^{210}Po comprise ~32% of the increased hazard at 10,000 years and follow ^{222}Rn in the decay series. Their presence depends on whether radon stays with the material or outgasses. The results presented here assume the radon remains with the original material.
- The relative hazard of ^{239}Pu is constant over 10,000 years. With a half-life of 24000 years and a first daughter , ^{235}U, with a half-life of 7.04E+08 years, changes in the first 10,000 years are minimal.

- The relative hazard of ^{240}Pu decreases noticeably during the last several-thousand years of the 10,000-year study period due exclusively to its 6540-year half-life. Early daughters have such long half-lives (10^7 and 10^{10} years) that they and later daughters do not contribute.
- The relative hazard of ^{241}Pu is a bit more interesting. It peaks at about 60 years with an increased hazard of about 60% due to the in-growth of ^{241}Am. It then decreases with the decay of the ^{241}Am to a minimum at about 8000 years, when it begins to increase again mostly due to the in-growth of ^{237}Np.
- The relative hazard of ^{242}Pu is constant over 10,000 years. With a half-life of 376,000 years, changes in the first 10,000 years are minimal.
- The relative hazard of ^{244}Pu is constant for the first few-thousand years but then begins to increase (69% in the last few-thousand years), due to the in-growth of ^{240}Pu. ^{240}U and ^{240}Np are in secular equilibrium (equal activities) with ^{244}Pu very shortly after the creation of the ^{244}Pu; however, their presence does not contribute significantly to the relative hazard because they are beta/gamma emitters and as such have very high category 2 thresholds.

Specific Plutonium Mixtures (Figures 4–9)

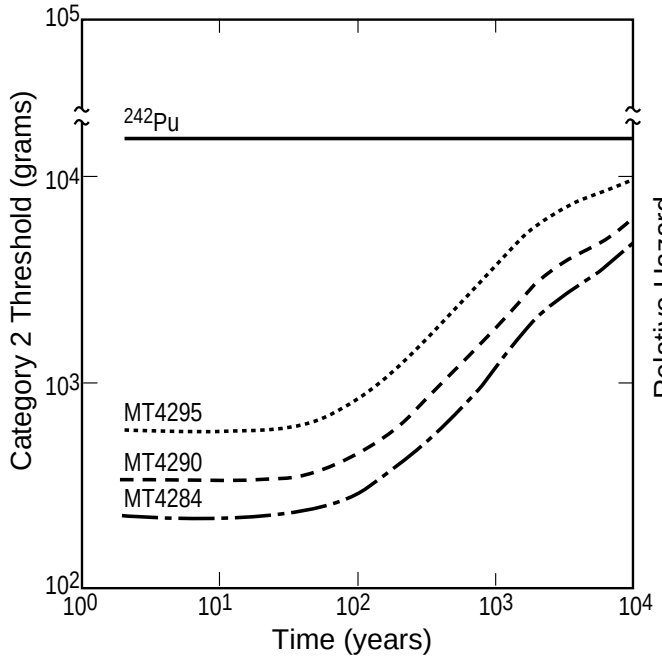

Figure 4. Time dependence of category 2 thresholds for the MT42 mixtures

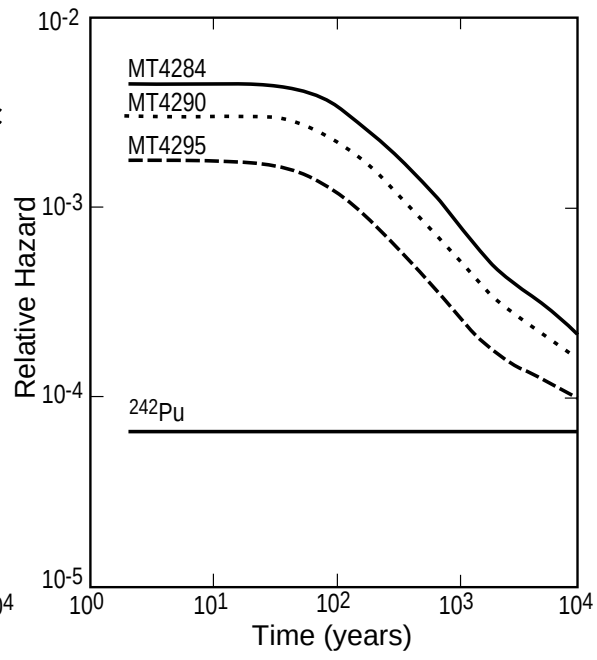

Figure 5. Time dependence of the relative hazard for the MT42 mixtures

Figures 4 and 5 illustrate the time dependence of the category 2 thresholds and relative hazard for the MT42 mixtures. The ^{238}Pu and ^{241}Pu components of these mixtures have a large effect on the

values for the mixtures. The slowly decreasing relative hazard with time can be seen to follow closely the curves for ^{238}Pu and the ^{241}Pu/^{241}Am isotopes.

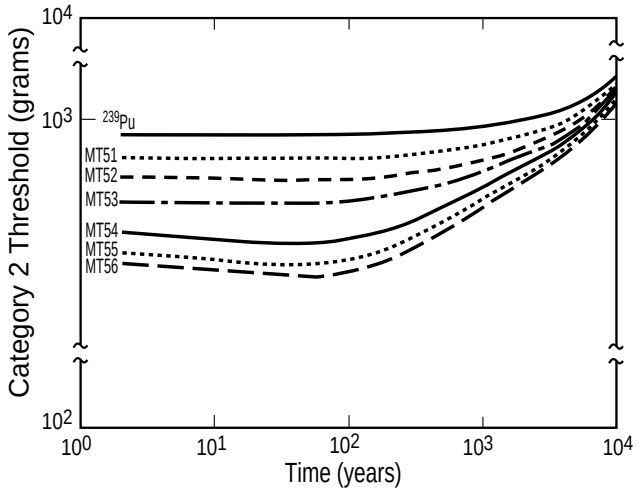

Figure 6. Time dependence of category 2 thresholds for the MT5x mixtures

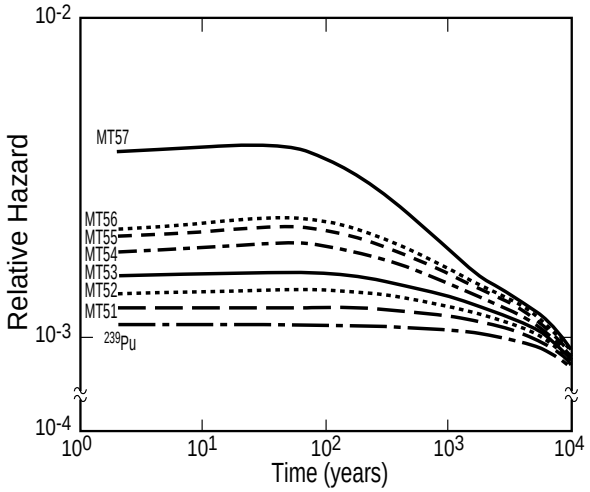

Figure 7. Time dependence of the relative hazard for the MT5x mixtures

Figures 6 and 7 illustrate the time dependence of the category 2 thresholds and relative hazard for the MT5x mixtures. The relative hazard for the first 10,000 years is dominated by the ^{240}Pu and ^{241}Pu/^{241}Am components. After this much time, the curves are converging to the pure ^{239}Pu curve.

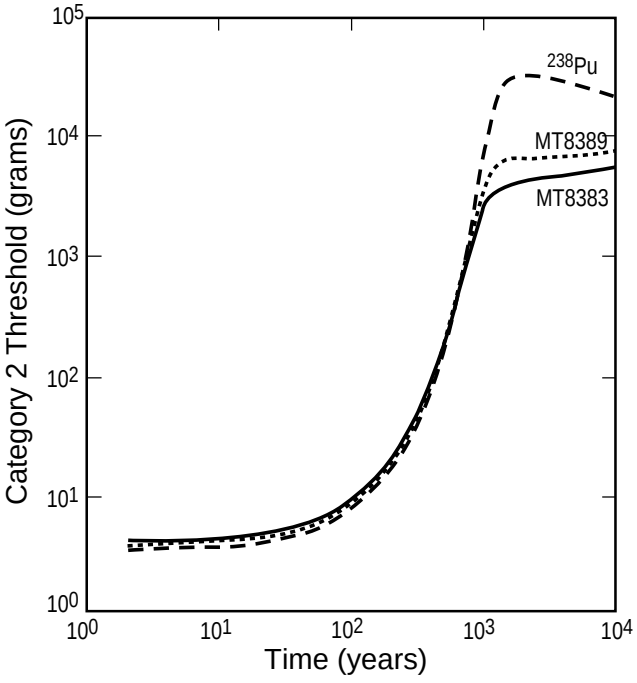

Figure 8. Time dependence of the category 2 thresholds for the MT83 mixtures

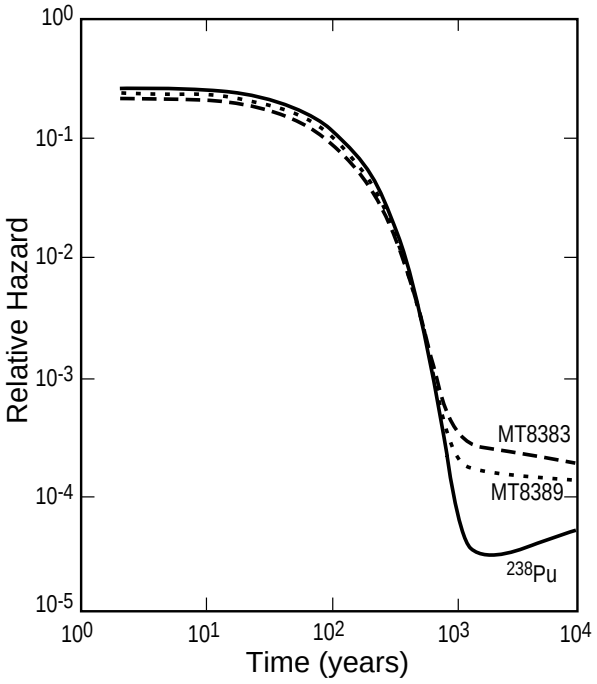

Figure 9. Time dependence of the relative hazard for the MT83 mixtures

Figures 8 and 9 illustrate the time dependence of the category 2 thresholds and relative hazard for the MT83 mixtures. The relative hazard for the first thousand years is dominated by the ^{238}Pu component. However, since the half-life of ^{238}Pu is 87.7 years, the relative hazard is dominated by the ^{239}Pu component (~10%) after a thousand years.

CONCLUSIONS

To determine a preliminary hazard category for a nuclear facility with a lifetime on the order of 100 years or less, one should pay particular attention to inventories rich in ^{241}Pu and the MT42 series of specific mixtures.

- The relative hazard of ^{241}Pu increases 60% during the first 60 years of its life.
- The relative hazard of the MT42 series is dominated by isotopes other than ^{242}Pu rendering it 26–66 times more hazardous than ^{242}Pu alone.

To evaluate the hazard of a long-term (10,000-year) storage facility, one should consult the time dependence of the relative hazard of plutonium isotopes and specific mixtures presented in this report, since time dependence is not readily apparent from a consideration of the initially dominant isotope alone.

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6. Bateman, H., "Solution of a System of Differential Equations Occurring in the Theory of Radio-active Transformations," *Proc. Cambridge Phil. Soc.* **15**, 428, (1910).

APPENDIX A. Charts of Decay Chains for Subject Isotopes

^{238}Pu

Isotope	Half-Life (years)	Specific Act. (Ci/g)	Cat.2 Threshold (grams)
^{238}Pu	87.7	17.1	3.63
^{234}U	2.45E+05	6.24E-03	3.53E+04
^{230}Th	7.7E+04	2.02E-02	4.40E+03
^{226}Ra	1.6E+03	0.989	2.62E+09
^{222}Rn	1.05E-02 (3.82 d)	1.53E+05	1.10E+03
^{218}Po	5.80E-06 (3 m)	2.83E+08	1.94E-07*
^{214}Pb	5.09E-05 (27 m)	3.28E+07	0.128
^{214}Bi	3.74E-05 (20 m)	4.47E+07	9.09E-02
^{214}Po	5.20E-12 (164 usec)	3.21E+14	1.71E-13*
^{210}Pb	22.3	76.3	29.0
^{210}Bi	1.37E-02 (5 d)	1.24E+05	1.20
^{210}Po	0.379	4.49E+03	7.8E-02
^{206}Pb	stable		

* No dose conversions were available. Threshold was taken to be 55 Ci for alpha emitters and 4.30E+05 Ci for beta-gamma emitters as provided by DOE-STD-1027-92.

^{239}Pu

Isotope	Half-Life (years)	Specific Act. (Ci/g)	Cat. 2 Threshold (grams)
^{239}Pu	2.4E+04	6.23E-02	9.13E+02
^{235}U	7.04E+08	2.16E-06	1.10E+08
^{231}Th	2.91E-03 (25.5 h)	5.32E+05	65.9
^{231}Pa	3.28E+04	4.72E-02	4.66E+02
^{227}Ac	21.8	72.2	5.89E-02
^{223}Fr	4.15E-05 (21.8 m)	3.86E+07	0.12
^{223}Ra	0.0312 (11.4 d)	5.14E+04	7.44E-02
^{219}Rn	1.3E-07 (4 sec)	1.26E+10	8.45E-05
^{215}Po	5.64E-11 (1.8 msec)	2.95E+13	1.86E-12*
^{211}Pb	6.86E-05 (36 m)	2.47E+07	0.144
^{211}Bi	4.07E-06 (2 m)	4.16E+08	1.33E-07*
^{207}Tl	9.08E-06 (4.8 m)	1.9E+08	2.26E-03*
^{207}Pb	stable		

* No dose conversions were available. Threshold was taken to be 55 Ci for alpha emitters and 4.30E+05 Ci for beta-gamma emitters as provided by DOE-STD-1027-92.

^{240}Pu

Isotope	Half-Life (years)	Specific Act. (Ci/g)	Cat. 2 Threshold (grams)
^{240}Pu	6.54E+03	2.28E-01	2.47E+02
^{236}U	2.34E+07	6.47E-05	8.50E+05*
^{232}Th	1.40E+10	1.10E-07	1.63E+08
^{228}Ra	5.75	2.73E+02	25.0
^{228}Ac	6.99E-04 (6.13 h)	2.24E+06	4.38E-02
^{228}Th	1.913	8.20E+02	0.112
^{224}Ra	1.00E-02 (3.66 d)	1.60E+05	6.19E-02
^{220}Rn	1.76E-06 (55.6 s)	9.23E+08	0.130
^{216}Po	4.76E-09 (0.15 s)	3.48E+11	1.58E-10*
^{212}Pb	1.21E-03 (10.6 h)	1.39E+06	0.128
^{212}Bi	1.15E-04 (60.6 m)	1.47E+07	9.24E-02
^{208}Tl	5.83E-06 (3.07 m)	2.94E+08	1.46E-03*
^{208}Pb	stable		

* No dose conversions were available. Threshold was taken to be 55 Ci for alpha emitters and 4.30E+05 Ci for beta-gamma emitters as provided by DOE-STD-1027-92.

^{241}Pu

Isotope	Half-Life (years)	Specific Act. (Ci/g)	Cat. 2 Threshold (grams)
^{241}Pu	14.4	103	28.0
^{241}Am	432	3.43	16.0
^{237}Np	2.14E+06	7.05E-04	8.30E+04
^{233}Pa	7.40E-02 (27 d)	2.07E+04	1.58E+02
^{233}U	1.59E+05	9.65E-03	2.30E+04
^{229}Th	7.34E+03	0.213	67.1
^{225}Ra	4.05E-02 (14.8 d)	3.92E+04	9.60E-02
^{225}Ac	2.74E-02 (10 d)	5.80E+04	4.90E-02
^{221}Fr	9.13E-06 (4.8 m)	1.77E+08	3.10E-07*
^{217}At	1.02E-09 (32.2 msec)	1.61E+12	3.42E-11*
^{213}Bi	8.68E-05 (45.6 m)	1.93E+07	8.64E-02
^{213}Po	1.33E-13 (4.2 usec)	1.26E+16	4.44E-15*
^{209}Pb	3.71E-04 (3.25 h)	4.61E+06	69.0
^{209}Bi	stable		

* No dose conversions were available. Threshold was taken to be 55 Ci for alpha emitters and 4.30E+05 Ci for beta-gamma emitters as provided by DOE-STD-1027-92.

^{242}Pu

Isotope	Half-Life (years)	Specific Act. (Ci/g)	Cat. 2 Threshold (grams)
^{242}Pu	3.76E+05	3.93E-03	1.51E+04
^{238}U	4.47E+09	3.36E-07	7.08E+08
^{234}Th	6.60E-02	2.31E+04	37.4
^{234m}Pa	2.23E-06 (1.17min)	6.86E+08	6.27E-04*
^{234}U	2.45E+05	6.24E-03	3.52E+04
^{230}Th	8.00E+04	1.94E-02	4.59E+03
^{226}Ra	1.60E+03	0.989	2.62E+09
^{222}Rn	1.05E-02	1.54E+05	1.05E+03
^{218}Po	5.80E-06 (3.05 min)	2.83E+08	1.94E-07*
^{214}Pb	5.10E-05 (26.8 min)	3.28E+07	0.128
^{214}Bi	3.79E-05 (19.9 min)	4.41E+07	9.09E-02
^{214}Po	5.21E-12 (164 usec)	3.12E+14	1.76E-13*
^{210}Pb	22.3	76.4	28.8
^{210}Bi	1.37E-02	1.24E+05	1.21
^{210}Po	0.379	4.49E+03	7.85E-02
^{206}Pb	stable		

* No dose conversions were available. Threshold was taken to be 55 Ci for alpha emitters and 4.30E+05 Ci for beta-gamma emitters as provided by DOE-STD-1027-92.

^{244}Pu

Isotope	Half-Life (years)	Specific Act. (Ci/g)	Cat. 2 Threshold (grams)
^{244}Pu	8.26E+07	1.77E-05	3.35E+06
^{240}U	1.61E-03 (14.1 h)	9.25E+05	0.465
^{240}Np	1.24E-04 (65 m)	1.20E+07	3.58E-02
^{240} Pu	6.54E+03	2.28E-01	247
^{236}U	2.34E+07	6.47E-05	8.50E+05*
^{232}Th	1.40E+10	1.10E-07	1.63E+08
^{228}Ra	5.75	273	25
^{228}Ac	6.99E-04 (6.13 h)	2.24E+05	4.38E-02
^{228}Th	1.913	820	0.112
^{224}Ra	1.00E-02 (3.66 d)	1.60E+05	6.19E-02
^{220}Rn	1.76E-06 (55.6 s)	9.23E+08	0.13
^{216}Po	4.76E-09 (0.15 s)	3.48E+11	1.58E-10*
^{212}Pb	1.21E-03 (10.6 h)	1.39E+06	0.128
^{212}Bi	1.15E-04 (60.6 m)	1.47E+07	9.24E-02
^{208}Tl	5.83E-06 (3.07 m)	2.94E+08	1.46E-03*
^{208}Pb	stable		

* No dose conversions were available. Threshold was taken to be 55 Ci for alpha emitters and 4.30E+05 Ci for beta-gamma emitters as provided by DOE-STD-1027-92.

APPENDIX B

Bateman Equations:

In radioactive decay series (such as listed in Appendix A), the time rate of change in activity of any member of the series depends on the activity of the member before it (source of supply) and its own activity, or decay rate (sink). Expressed mathematically:

$$\frac{dA_i}{dt} = \lambda_{i-1}\frac{dA_{i-1}}{dt} - \lambda_i A_i$$

where A_i= the activity of the i^{th} member of the series,

$\lambda_i = \frac{ln2}{T_{1/2}}$ = the decay constant of the i^{th} member, and

$T_{1/2}$ = the half life of the i^{th} member

Bateman provided the following solution to find the activity of any member of the decay series as a function of time:

$$A_n(t) = A_1^0 \sum_{i=1}^{n} C_i e^{-\lambda_i t}$$

where A_1^0 = the initial activity of the parent,
$A_n(t)$ = the activity of the n^{th} member at time t, and

$$C_i = \frac{\prod_{k=2}^{n}\lambda_k}{\prod_{j=1}^{n}(\lambda_j - \lambda_i)}: \quad j \neq i$$

C_i = 1 by definition

This solution assumes the initial condition (at t=0) that: $A_1(0) = A_1^0$ and all other $A_n(0) = 0$; that is that there are no daughter isotopes.

Sum of Fractions Method:

If the facility inventory includes more than one radioactive isotope, the inventory limit for a hazard category 2 is such that:

$$\sum_{i=1}^{n} \left(\frac{m_i}{Th_i} \right) = 1$$

where m_i = the mass inventory of the i^{th} isotope, and
Th_i = the category 2 mass threshold limit for that isotope.

Likewise, then, for a mixture of total mass, M, comprising n different isotopes, each represented by a fraction, f_i, of the total,

$$M * \sum_{i=1}^{n} \left(\frac{f_i}{Th_i} \right) = 1$$

It follows, then, that the category 2 threshold for this mixture is:

$$M = \frac{1}{\sum_{i=1}^{n} \left(\frac{f_i}{Th_i} \right)}$$

APPENDIX C. Category 2 Threshold Quantities:

The category 2 threshold is the amount of material that, if released, could result in a dose of 10mSv (1 rem) to a person 300 meters away. This calculation is as follows:

$$\text{Threshold (grams)} = \frac{1}{RF * SA * \frac{X}{Q} * (CEDE * RR + CSDE)}$$

where:

RF = airborne release fraction (see reference 1)
SA = specific activity (Ci/gm)
X/Q = atmospheric dispersion (sec/m^3)
CEDE = committed effective does equivalent (rem/Ci)
RR = respiration rate = 3.50E-04 m^3/sec
CSDE* = cloud shine dose equivalent (rem-m^3/Ci-sec)

*The external dose to a person immersed in a cloud of radioactive material.

Los Alamos
NATIONAL LABORATORY

Los Alamos, New Mexico 87545