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Potential Radon-222 Emissions from the Thorium Nitrate Stockpile

J.W. Terry W.H. Hermes



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Environmental Sciences Division Nuclear Science and Technology Division

POTENTIAL RADON-222 EMISSIONS FROM THE THORIUM NITRATE STOCKPILE

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Potential Radon-222	2 Emissions f	rom the The	orium Nitrate	Stockpile
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ACRONYMS AND ABBREVIATIONS

CFR Code of Federal Regulations

d day

DNSC Defense National Stockpile Center

ft feet g gram h hour

LLW low-level waste

m meter

MeV mega-electron volt mR milliroentgen NTS Nevada Test Site

ORNL Oak Ridge National Laboratory

pCi pico-Curies sec or s second y year

Potential Radon-222	2 Emissions f	rom the The	orium Nitrate	Stockpile
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ABSTRACT

The Defense National Stockpile Center (DNSC), a field level activity of the Defense Logistics Agency, has stewardship of a stockpile of thorium nitrate that has been in storage for decades. The thorium nitrate stockpile was produced from 1959 to 1964 for the Atomic Energy Commission and previously has been under the control of several federal agencies. The stockpile consists of approximately 7 million pounds of thorium nitrate crystals (hydrate form) stored at two depot locations in the United States (75% by weight at Curtis Bay, Maryland, and 25% by weight at Hammond, Indiana). The material is stored in several configurations in over 21,000 drums. The U.S. Congress has declared the entire DNSC thorium nitrate stockpile to be in excess of the needs of the Department of Defense.

Part of DNSC's mission is to safely manage the continued storage, future sales, and/or disposition of the thorium nitrate stockpile. Historically, DNSC has sold surplus thorium nitrate to domestic and foreign companies, but there is no demand currently for this material. Analyses conducted by Oak Ridge National Laboratory (ORNL) in 2001 demonstrated that disposition of the thorium nitrate inventory as a containerized waste, without processing, is the least complex and lowest-cost option for disposition.

A characterization study was conducted in 2002 by ORNL, and it was determined that the thorium nitrate stockpile may be disposed of as low-level waste. The Nevada Test Site (NTS) was used as a case study for the disposal alternative, and special radiological analyses and waste acceptance requirements were documented. Among the special radiological considerations is the emission of ²²⁰Rn and ²²²Rn from buried material. NTS has a performance objective on the emissions of radon: 20 pCi m⁻² sec⁻¹ at the surface of the disposal facility. The radon emissions from the buried thorium nitrate stockpile have been modeled. This paper presents background information and summarizes the results of modeling radon emissions and compares those results with the NTS performance objective.

1. INTRODUCTION

The Defense National Stockpile Center (DNSC), a field level activity of the Defense Logistics Agency, has stewardship of a stockpile of thorium nitrate that has been in storage for decades. The thorium nitrate stockpile was produced from 1959 to 1964 for the Atomic Energy Commission and previously has been under the control of several federal agencies. The stockpile consists of approximately 7 million pounds of thorium nitrate crystals (hydrate form) stored at two depot locations in the United States (75% by weight at Curtis Bay, Maryland, and 25% by weight at Hammond, Indiana). The material is stored in several configurations in over 21,000 drums. The gamma dose at the surface of an isolated drum is 20–30 mR/h; however, the gamma dose within the storage facilities is 60–100 mR/h. The stockpile is classified as source material (10 CFR 20.1003) and is regulated by the U.S. Nuclear Regulatory Commission. The U.S. Congress has declared the entire DNSC thorium nitrate stockpile to be in excess of the needs of the Department of Defense.

Part of DNSC's mission is to safely manage the continued storage, future sales, and/or disposition of the thorium nitrate stockpile. Historically, DNSC has sold surplus thorium nitrate to domestic and foreign companies, but there is no demand currently for this material. Analyses conducted by Oak Ridge National Laboratory (ORNL) in 2001 demonstrated that disposition of the thorium nitrate inventory as a containerized waste, without processing, is the least complex and lowest-cost option for disposition. The viability of this disposition pathway required a determination of which portion, if any, of the inventory exhibits hazardous characteristics (e.g., Resource Conservation and Recovery Act D001 ignitability characteristic—equivalent to Department of Transportation oxidizer and/or D004 toxicity characteristic).

A characterization study was conducted in 2002 by ORNL. One hundred drums were sampled, and the samples were analyzed for chemical and radiological constituents. Salt block monoliths comprise the predominant physical form in the stockpile. Special container inspection and dose management assessments were completed to meet on-site material handling, transportation, and disposal site requirements. The Nevada Test Site (NTS) was used as a case study for the disposal alternative, and special radiological analyses and waste acceptance requirements were documented.

Among the special radiological considerations is the emission of ²²⁰Rn and ²²²Rn from buried material. NTS has a performance objective on the emissions of radon: 20 pCi m⁻² sec⁻¹ at the surface of the disposal facility. The radon emissions from the buried thorium nitrate stockpile have been modeled. This report presents background information and summarizes the results of modeling radon emissions and compares those results with the NTS performance objective.

2. BACKGROUND

Thorium-232 occurs naturally and has a half-life of 1.40×10^{10} years. The 232 Th decay series (see Fig. 1) begins with 232 Th and, after a series of alpha and beta emissions, reaches 208 Pb, a stable end product. In many steps in the series, gamma rays and conversion electrons are emitted. Thorium-232 is not a fissile isotope; but it can be used as a precursor to generate fissionable 233 U, which can sustain a chain reaction. Uranium-233 generates significantly more neutrons in the course of thermal fission than either 235 U or 239 Pu, making it a desirable fuel to sustain criticality, while having excess neutrons to breed more 233 U from thorium.

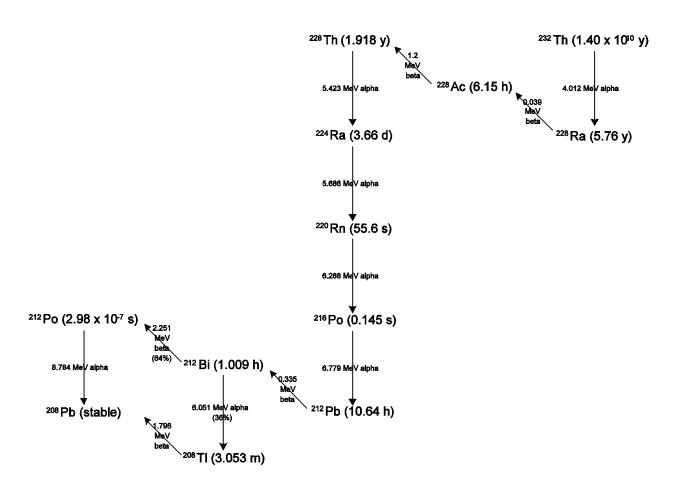


Fig. 1. Thorium-232 decay sequence.

One of the progeny in the decay chain is ²²⁰Rn, a gas, which can escape and spread contamination. The decay chain continues beyond ²²⁰Rn; the highest radiation dose from shielded material comes from ²⁰⁸Tl, which emits a highly penetrating 2.6-MeV gamma ray. Thus ²³²Th progeny present both contamination and exposure risks to personnel and the environment.

Thorium-bearing ores typically contain substantial quantities of uranium. Uranium-238 is the most abundant (99.2745%) of the naturally occurring isotopes of uranium. The half-life of 238 U, 4.47 × 109 years, is about one-third of the half-life of 232 Th. The 238 U decay series (see Fig. 2) begins with 238 U and, after a series of alpha and beta emissions, reaches 206 Pb, a stable end product. As in the 232 Th decay sequence, many steps in the series result in the emission of gamma rays and conversion electrons. Unlike the 232 Th decay series, 238 U does not have a very high energy gamma-emitting progeny. Hence, the primary personnel and environmental radiological concern is contamination spread by 222 Rn, a gas.

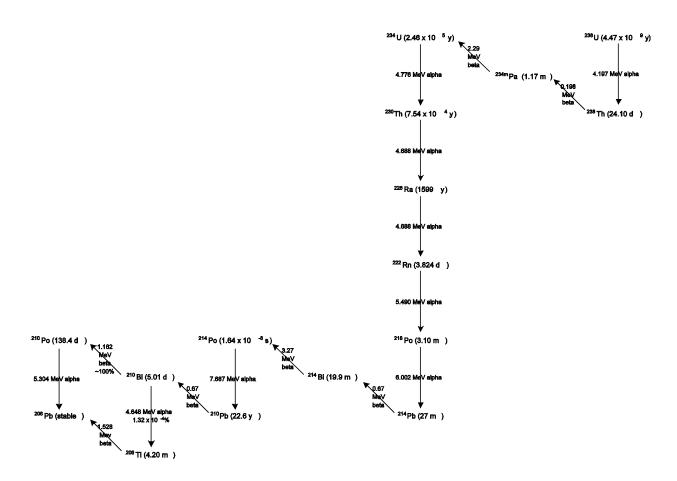


Fig. 2. Uranium-238 decay sequence.

On the normal, macroscopic scale all isotopes of an element have almost identical physical and chemical characteristics. There are isotopic effects, but these effects are quite small for atoms of high atomic mass such as thorium and uranium. Thus, all isotopes of thorium would behave similarly during the physical and chemical processing required to extract thorium from the ore body and convert it to thorium nitrate. These physical and chemical processes would not distinguish between the ²³²Th and ²²⁸Th from the ²³²Th decay sequence and the ²³⁰Th from the ²³⁸U decay sequence.

The thorium and uranium have been in the ore body for millions of years. Over this long time, the ²³⁸U decay sequence has reached secular equilibrium; about 500,000 years is required for the ²³⁸U decay sequence to establish secular equilibrium. In secular equilibrium, there is one radioactive decay of each progeny in the decay sequence for every radioactive decay of the progenitor. Although ²³²Th has a longer half-life than ²³⁸U, secular equilibrium for the ²³²Th decay sequence is established in slightly over 100 years because none of its progeny have a half-life greater than 6 years.

3. DATA

The domestically produced thorium nitrate was chemically processed from monazite sands mined in South Africa; the thorium nitrate produced in France originated from ores mined in the Malagasy Republic; and the thorium nitrate produced in India originated from ores mined in India. The thorium nitrate produced domestically and in France is in the pentahydrate form, and the thorium nitrate produced in India is in the tetrahydrate form. The ²³⁰Th content of the three components of the thorium nitrate stockpile depends upon the amount of ²³⁸U in the respective ore bodies. Preliminary data for the ²³⁸U content of the thorium-containing ore bodies in South African, Malagasy Republic, and India were obtained from a published source (Benedict, Pigford, and Levi 1981). The published ratios of the average ²³⁸U concentrations to the average thorium concentrations are as follows: 0.022—ore from South Africa, 0.050—ore from Malagasy Republic, and 0.042—ore from India.

The ²³⁰Th activity in the ores is assumed to have been equivalent to the ²³⁸U activity because the uranium was present in the ores for geologic eras, providing more than sufficient time for secular equilibrium of the ²³⁸U decay sequence to be established. To perform the calculations required to determine the ²²²Rn emissions, an appropriate collection of constants and general data must be acquired; this information is given in Appendix A. All the thorium in the ore, including all the ²³⁰Th, is assumed to move into the thorium nitrate; this is equivalent to assuming a conversion efficiency of 100%. This assumption would produce an overestimate (conservative estimate) of the amount of ²³⁰Th in the stockpile if the ratios ²³⁸U:Th are representative of the ores from which the thorium nitrate stockpile was produced. These assumptions and detailed calculations are presented in Appendix B. The resulting calculated ²³⁰Th activities per gram of thorium nitrate are as follows:

```
2.685 \times 10^3 pCi/g — ore mined in South Africa and processed in the United States, 6.187 \times 10^3 pCi/g — ore mined in Malagasy Republic and processed in France, and 5.204 \times 10^3 pCi/g — ore mined and processed in India.
```

In the spring and summer of 2002, a sampling and analysis program was conducted for the thorium nitrate stockpile. Representative samples were sent to Southwest Research Institute, a laboratory accredited by NTS, where they were analyzed for chemical and radiological constituents. Detailed assumptions and calculations are presented in Appendix C. The resulting average values of the measured ²³⁰Th activities per gram of thorium nitrate are as follows:

```
3.863 \times 10^3 pCi/g — produced in the United States and stored at Hammond Depot, 4.290 \times 10^3 pCi/g — produced in the United States and stored at Curtis Bay Depot, 39.59 \times 10^3 pCi/g — produced in France and stored at Curtis Bay Depot, and 7.538 \times 10^3 pCi/g — produced in India and stored at Curtis Bay Depot.
```

4. MODELING

NTS has developed a deterministic performance assessment model for disposal of low-level radioactive waste (LLW). The performance assessment included the computation of ²²²Rn emissions from buried LLW. This deterministic performance assessment model is being replaced by a probabilistic model. In the replacement process, a new model for ²²²Rn emissions is being developed and verified.

The newer ²²²Rn emissions model is still under development and is not yet available for use. Hence, the older model was used for illustrative purposes. During discussions with NTS personnel, it was suggested that materials determined to have ²²²Rn emissions substantially below the performance objective of 20 pCi m⁻² sec⁻¹ at the surface of the ground using the older emissions model would likely remain below the performance objective using the newer model.

The NTS radon performance objective applies to any time between the present and 1000 years in the future. Before the NTS model could be applied, the time dependent levels of ²²²Rn in the wastes were determined for both the preliminary ore data (see Appendix D) and the recent measurements (see Appendix E).

Although orders of magnitude more ²²⁰Rn than ²²²Rn will be generated in the thorium nitrate, the emissions of ²²⁰Rn will not be computed. The NTS performance assessment model for deep burial provides a delay of about ½ year between decay of radium into radon and emergence of radon from the ground. Only infinitesimal amounts of ²²⁰Rn could emerge because this delay period is equivalent to roughly 200,000 half-lives for ²²⁰Rn.

Radon-222 is a decay product of both ²³⁰Th and any ²³⁸U contained in the thorium nitrate. Therefore, the activities of both progenitors were considered in the calculations. For the calculations performed using the preliminary ore data, it was assumed that ²³⁸U occurred in the thorium nitrate at a concentration of 1 ppm based upon mass spectroscopic data acquired from a single sample of thorium nitrate from DNSC; at this concentration no appreciable activity levels

of 222 Rn would be generated at any time through 100,000 years in the future. Details of this calculation are presented in Appendix D. The highest measured 238 U activity levels, \sim 8 pCi/g, would produce vanishingly small activity levels of 222 Rn at any time through 100,000 years in the future. Details of this calculation are presented in Appendix E.

The ²³⁰Th in the thorium nitrate from the original ore would create essentially all the ²²²Rn at 1000 years in the future and beyond. The maximum generation of ²²²Rn would occur at slightly less than 10,000 years in the future; details are presented in Appendices D and E. The calculated ²²²Rn activity levels at 1000 years in the future are as follows:

Preliminary ore data

```
0.940 \times 10^3 pCi/g — ore mined in South Africa and processed in the United States, 2.164 \times 10^3 pCi/g — ore mined in Malagasy Republic and processed in France, and 1.821 \times 10^3 pCi/g — ore mined and processed in India.
```

Recent measurements

```
1.564 \times 10^3 pCi/g — produced in the United States and stored at Hammond Depot, 1.744 \times 10^3 pCi/g — produced in the United States and stored at Curtis Bay Depot, 17.47 \times 10^3 pCi/g — produced in France and stored at Curtis Bay Depot, and 2.670 \times 10^3 pCi/g — produced in India and stored at Curtis Bay Depot.
```

Emissions of 222 Rn from the buried thorium nitrate depend upon the physical processes occurring during the movement of the 222 Rn to the ground surface. If the passage of the 222 Rn could be retarded for one or more half-lives ($t_{1/2} = 3.824$ d), much of it would decay into 218 Po and successive 222 Rn progeny which would be trapped because they are not gases. The deeper the 222 Rn is buried, the more opportunity there is for its movement to be retarded or stopped.

At NTS, uranium and uranium-contaminated wastes are typically buried 26 ft below the surface of the ground. Previously, a large quantity of thorium was buried 40 ft below the surface of the ground to ensure that ²²²Rn emissions met the performance objective of 20 pCi m⁻² sec⁻¹ at all times through 1000 years in the future. The deterministic NTS model provides parameters that may be used to determine ²²²Rn emissions from both depths. The ²²²Rn emissions from the shallower depth were found to be unacceptably high for both the preliminary ore and the recently measured data. Details are given in Appendices F and G.

The ²²²Rn emissions from the 40-ft depth were determined to be below the NTS limit through 1000 years in the future. Details of these calculations are presented in Appendices F and G. The calculated values are very low initially and grow to the following levels at 1000 years in the future:

Preliminary ore data

```
0.051 pCi m<sup>-2</sup> sec<sup>-1</sup> — ore mined in South Africa and processed in the United States, 0.118 pCi m<sup>-2</sup> sec<sup>-1</sup> — ore mined in Malagasy Republic and processed in France, 0.099 pCi m<sup>-2</sup> sec<sup>-1</sup> — ore mined and processed in India, and 0.268 pCi m<sup>-2</sup> sec<sup>-1</sup> — total emissions.
```

Recent measurements

```
0.098~pCi~m^{-2}~sec^{-1}— produced in the United States and stored at Hammond Depot, 0.109~pCi~m^{-2}~sec^{-1}— produced in the United States and stored at Curtis Bay Depot, 1.090~pCi~m^{-2}~sec^{-1}— produced in France and stored at Curtis Bay Depot, 0.167~pCi~m^{-2}~sec^{-1}— produced in India and stored at Curtis Bay Depot, and 1.464~pCi~m^{-2}~sec^{-1}— total emissions.
```

5. CONCLUSIONS

The modeled ²²²Rn emissions from the thorium nitrate stockpile buried at NTS meet the NTS emissions limit if the material were buried 40 ft below the surface of the ground. The published values for the average concentrations of uranium in ore bodies are not representative of the concentrations of uranium in the ore bodies from which the DNSC stockpile was produced.

6. REFERENCE

Benedict, M., T.H. Pigford, and H.W. Levi (1981). *Nuclear Chemical Engineering*, 2nd ed., McGraw-Hill, New York.

Potential Radon-222	Fmissions t	from the '	Thorium	Nitrato	Stocknile

Appendix A

APPLICABLE CONSTANTS AND CONVERSION FACTORS

Performing assessments of the potential radon emissions from burial of the thorium nitrate stockpile at Nevada Test Site requires a significant number of physical constants and conversion factors. The constants and conversion factors used in these assessments are presented in this appendix.

Appendix A — Applicable Constants and Conversion Factors

$$\begin{split} N_{A} &= \text{Avogadro's number} & N_{A} = 6.02214199 \times 10^{23} & \text{gm} = 1.000 \times 10^{-3} \text{ kg} \\ \\ m_{Ra226} &= \text{atomic mass of }^{226} \text{Ra} & m_{Ra226} = 226 \text{ gm} \\ \\ m_{Ra222} &= \text{atomic mass of }^{227} \text{Rn} & m_{Ra222} = 222 \text{ gm} \\ \\ m_{Th232} &= \text{atomic mass of }^{229} \text{Th} & m_{Th228} = 228 \text{ gm} \\ \\ m_{Th230} &= \text{atomic mass of }^{230} \text{Th} & m_{Th230} = 230 \text{ gm} \\ \\ m_{Th232} &= \text{atomic mass of }^{232} \text{Th} & m_{Th232} = 232.038051 \text{ gm} \\ \\ m_{Th234} &= \text{atomic mass of }^{234} \text{Th} & m_{Th228} = 234 \text{ gm} \\ \\ m_{U238} &= \text{atomic mass of }^{234} \text{Th} & m_{U235} = 235.043924 \text{ gm} \\ \\ m_{U238} &= \text{atomic mass of }^{238} \text{U} & m_{U238} = 238.050785 \text{ gm} \\ \\ m_{H} &= \text{atomic mass of hydrogen} & m_{H} &= 1.0079 \text{ gm} \\ \\ m_{O} &= \text{atomic mass of oxygen} & m_{O} &= 15.9994 \text{ gm} \\ \\ m_{O} &= \text{atomic mass of hitrogen} & m_{N} &= 14.0067 \text{ gm} \\ \\ \\ m_{ThN} &= \text{mass of thorium nitrate} & m_{ThN} &= m_{Th232} + 4m_{N} + 17m_{O} + 10m_{H} \\ \\ \\ m_{ThN} &= 570.134 \text{ gm} \\ \\ \\ \text{Ci} &= 3.7 \times 10^{10} \text{ Bq} & \text{pCi} &= 1 \times 10^{-12} \text{ Ci} \\ \\ \\ \text{SpA}_{Th228} &= \text{specific activity of }^{228} \text{Th} & \text{SpA}_{Th228} &= \frac{\lambda_{Th238} N_{A}}{m_{Th230}} & \text{SpA}_{Th228} &= 8.202 \times 10^{14} \frac{\text{pCi}}{\text{gm}} \\ \\ \text{SpA}_{Th230} &= \text{specific activity of }^{230} \text{Th} & \text{SpA}_{Th230} &= \frac{\lambda_{Th230} N_{A}}{m_{Th230}} & \text{SpA}_{Th230} &= 2.063 \times 10^{10} \frac{\text{pCi}}{\text{gm}} \\ \\ \end{array}$$

 SpA_{Th232} = specific activity of ²³²Th

 $SpA_{Th232} = \frac{\lambda_{Th232}N_A}{m_{Th232}}$ $SpA_{Th232} = 1.101 \times 10^5 \frac{pCi}{gm}$

SpA_{ThN} = specific activity of ²³² Th in thorium nitrate	$SpA_{ThN} = \frac{\lambda_{Th232} N_A}{m_{ThN}}$	$SpA_{ThN} = 4.482 \times 10^4 \frac{pCi}{gm}$
SpA_{Th234} = specific activity of ²³⁴ Th	$SpA_{Th234} = \frac{\lambda_{Th234} N_A}{m_{Th234}}$	$SpA_{Th234} = 2.315 \times 10^{16} \frac{pCi}{gm}$
SpA_{U235} = specific activity of ²³⁵ U	$SpA_{U235} = \frac{\lambda_{U235} N_A}{m_{U235}}$	$SpA_{U235} = 2.162 \times 10^6 \frac{pCi}{gm}$
SpA_{U238} = specific activity of ²³⁸ U	$SpA_{U238} = \frac{\lambda_{U238} N_A}{m_{U238}}$	$SpA_{U238} = 3.362 \times 10^5 \frac{pCi}{gm}$

Appendix B

THORIUM NITRATE CONSTITUENTS FROM ORE DATA

INTRODUCTION

This appendix presents background information, assigns uranium and thorium activities in the thorium nitrate stockpile based upon published values for the ratios of uranium to thorium in the original ore body, and lists the calculated ²³⁰Th activities per gram of thorium nitrate.

BACKGROUND

Western Australian mineral sands deposits contain up to 10% heavy minerals, of which 1–3% is monazite. This in turn typically contains 5–7% of radioactive thorium and 0.1–0.3% of uranium, which is barely radioactive (UIC 1998).

Thorium-234 and ²³⁰Th are members of the ²³⁸U decay chain. They are found in natural thorium when uranium is present in natural ores. Because the half-life of ²³⁴Th is only 24.1 days, it soon decays in separated thorium, resulting in an inconsequential concentration. Thorium-230, however, has a 75.4-thousand-year half-life and is a significant constituent of thorium from ores containing uranium. The ²³⁰Th/²³²Th atom ratio is given by

$$\frac{^{230}\text{Th}}{^{232}\text{Th}} = r(0,2) = \frac{(0.9927)(7.54 \times 10^4)[r(U,Th)]}{4.47 \times 10^9} = 1.68 \times 10^{-5} r(U,Th) , \qquad [1]$$

where

0.9927 = the atom fraction of ^{238}U in natural uranium,

r(U,Th) = the atomic ratio of U/Th in the ore, and

 4.47×10^9 = the half-life of ²³⁸U.

The ratio of the activity of ²³⁰Th to ²³²Th is given by

$$\frac{^{230}\text{A}}{^{232}\text{A}} = \frac{^{232}\text{H} \times \text{r}(0,2)}{^{230}\text{H}} = \frac{1.40 \times 10^{10}}{(7.54 \times 10^4)(1.68 \times 10^{-5})\,\text{r}(\text{U},\text{Th})} = 3.1\,\,\text{r}(\text{U},\text{Th})\,, \qquad [2]$$

where *H* is the half-life of the designated isotope.

Thorium has been recovered as a by-product of uranium production from ores of the Blind River district in Ontario in which the U/Th ratio is 6. In such thorium, the 230 Th activity is 3.1×6 or 18.6 times the activity of the 232 Th.

There are about 12 minerals that contain thorium in the principal thorium-containing ore. Thorium production was a by-product of monazite processing during the period the stockpile was produced. Most of the world's thorium had come from beach sands where coproduction of rare earths, titanium, and zirconium defrayed much of the cost of extracting thorium. Because monazite is usually a minor constituent of the deposits of other minerals (0.5-1.0 wt %), physical separations are first employed to concentrate the monazite up to 98% purity. The composition of the monazite concentrate, for ThO₂ and U₃O₈, in weight percent, is as follows:

				South African	Malagasay
	India	Brazil	Florida beach sands	monazite rock	Republic
ThO_2	8.88	6.5	3.1	5.9	8.75
U_3O_8	0.35	0.17	0.47	0.12	0.41

From the data above,

				South African	Malagasay
	India	Brazil	Florida beach sands	monazite rock	Republic
Th/U	23.8	13	6.2	46.1	20
r(U/Th)	0.042	0.077	0.161	0.022	0.05

The 230 Th/ 232 Th ratio = r(0,2) and is calculated from Eq. 1:

			South African	Malagasay	
India	Brazil	Florida beach sands	monazite rock	Republic	
7.07×10^{-7}	1.30×10^{-6}	2.71×10^{-6}	3.70×10^{-7}	8.41×10^{-7}	_

The thorium content in thorium nitrate pentahydrate is 40.7% by weight. In 1 MT of the ThN, there is 407 kg of Th, of which 230 Th (in gm/MT ThN) is approximately

0.29 0.53 1.10 0.15 0.34

WEIGHT PERCENT THORIUM-232

Assume that the 232 Th occurs in the concentration given above for South African monazite ore, Malagasy Republic ore, and Indian ore. Benedict, Pigford, and Levi (1981) give weight percent ThO₂. This must be converted to weight percent 232 Th. The mass of ThO₂ is

264 amu and the mass of 232 Th is 232 amu; thus, the weight percent Th is (232/264) times the weight percent ThO₂.

In the following equations, $wt\%_{232}$ is the weight percent of ²³²Th in the ore, and ppm_{232} is the concentration of ²³²Th in the ore.

$$wt\%_{232_0} = 5.9 \left(\frac{232}{264}\right) \qquad wt\%_{232_0} = 5.185$$

$$ppm_{232_0} = wt\%_{232_0} 10^4 \qquad ppm_{232_0} = 5.185 \times 10^4$$

$$wt\%_{232_1} = 8.75 \left(\frac{232}{264}\right) \qquad wt\%_{232_1} = 7.689$$

$$ppm_{232_1} = wt\%_{232_1} 10^4 \qquad ppm_{232_1} = 7.689 \times 10^4$$

$$wt\%_{232_2} = 8.88 \left(\frac{232}{264}\right) \qquad wt\%_{232_2} = 7.804$$

$$ppm_{232_2} = wt\%_{232_2} 10^4 \qquad ppm_{232_2} = 7.804 \times 10^4$$

To establish an upper bound, it is assumed that the partitioning coefficients for ^{230,234}Th equal 1. The Hammond inventory contains 863,685 kg of domestic thorium nitrate, and the Curtis Bay inventory contains 1,507,339 kg of domestic thorium nitrate that originated from South African monazite ore.

$$M_{ThN_0} = (863,685 + 1,507,339) \text{ kg}$$
 $M_{ThN_0} = 2.371 \times 10^6 \text{ kg}$

The Hammond inventory contains no French thorium nitrate. The Curtis Bay inventory contains 627,256 kg French thorium nitrate that originated from Malagasy Republic ore.

$$M_{ThN_1} = 627,256 \text{ kg}$$
 $M_{ThN_1} = 6.273 \times 10^5 \text{ kg}$

The Hammond inventory contains no thorium nitrate from Indian ore, and the Curtis Bay inventory contains 218,556 kg that originated from Indian ore.

$$M_{ThN_2} = 218,556 \text{ kg}$$
 $M_{ThN_2} = 2.186 \times 10^5 \text{ kg}$

WEIGHT PERCENT URANIUM-238

Assume that the 238 U occurs in the ore at the concentrations given above for South African monazite ore, Malagasy Republic ore, and Indian ore. Benedict, Pigford, and Levi (1981) gives weight percent U_3O_8 . This must be converted to weight percent 238 U. The mass of U_3O_8 is 842 amu and the mass of three 238 U atoms is 714 amu. Thus, the weight percent U is (714/842) times the weight percent U_3O_8 .

The weight percent of ²³⁸U in the ore is given by $wt\%_{238}$, and the concentration of ²³⁸U in the ore is given by ppm_{238} .

$$wt\%_{238_0} = 0.12 \left(\frac{714}{842} \right) \qquad wt\%_{238_0} = 0.102$$

$$ppm_{238_0} = wt\%_{2380} 10^4 \qquad ppm_{238_0} = 1.018 \times 10^3$$

$$wt\%_{2381} = 0.41 \left(\frac{714}{842} \right) \qquad wt\%_{2381} = 0.348$$

$$ppm_{238_1} = wt\%_{238_1} 10^4 \qquad ppm_{238_1} = 3.477 \times 10^3$$

$$wt\%_{238_2} = 0.35 \left(\frac{714}{842} \right) \qquad wt\%_{238_2} = 0.297$$

$$ppm_{238_2} = wt\%_{238_2} 10^4 \qquad ppm_{238_2} = 2.968 \times 10^3$$

RATIO OF THORIUM TO URANIUM

The ratio of thorium to uranium in the ore is given by R_{ThU} .

$$j = 0...2$$

$$R_{\text{ThU}_{j}} = \frac{\text{wt\%}_{232_{j}}}{\text{wt\%}_{238_{j}}} \qquad R_{\text{ThU}} = \begin{pmatrix} 50.953 \\ 22.117 \\ 26.293 \end{pmatrix}$$

$$R_{UTh} = R_{ThU}^{-1}$$

$$R_{UTh} = \begin{pmatrix} 0.02 \\ 0.045 \\ 0.038 \end{pmatrix}$$

MASS OF URANIUM IN THE ORE

 M_{NatU} is the mass of natural uranium in the ore. It is assumed to be all ²³⁸U. The mass of ²³²Th in the ore, M_{ThOre} , would be the mass of the inventory times the mass ratio of ²³²Th to thorium nitrate.

$$M_{ThOre} = M_{ThN} \frac{m_{Th232}}{m_{ThN}}$$

$$\mathbf{M}_{\text{ThOre}} = \begin{pmatrix} 9.65 \times 10^{5} \\ 2.553 \times 10^{5} \\ 8.895 \times 10^{4} \end{pmatrix} \text{ kg } \mathbf{M}_{\text{NatU}_{j}} = \frac{\mathbf{M}_{\text{ThOre}_{j}}}{\mathbf{R}_{\text{ThU}_{j}}} \qquad \mathbf{M}_{\text{NatU}} = \begin{pmatrix} 1.894 \times 10^{4} \\ 1.154 \times 10^{4} \\ 3.383 \times 10^{3} \end{pmatrix} \text{ kg}$$

This mass of uranium has an activity, A_{NatU} , determined by its specific activity.

$$A_{NatU} = SpA_{U238}M_{NatU} \qquad \qquad A_{NatU} = \begin{pmatrix} 6.367 \times 10^{12} \\ 3.881 \times 10^{12} \\ 1.137 \times 10^{12} \end{pmatrix} p \text{Ci}$$

THORIUM-230 ACTIVITY IN THE ORE

The 230 Th activity in the ore, $A_{Th230ore}$, is equal to the 238 U activity in the ore, A_{NatU} , because of secular equilibrium.

$$A_{Th230\text{ore}} = A_{NatU}$$

$$A_{Th230\text{ore}} = \begin{pmatrix} 6.367 \times 10^{12} \\ 3.881 \times 10^{12} \\ 1.137 \times 10^{12} \end{pmatrix} pCi$$

The mass of 230 Th carried over to the thorium nitrate may be computed from the activity of 230 Th in the inventory, $A_{Th230ore}$, and the specific activity of 230 Th, SpA_{Th230} .

$$\begin{split} \mathbf{M}_{\text{Ta230are}} &= \frac{\mathbf{A}_{\text{Ta230}}}{\text{SpA}_{\text{Ta230}}} & \mathbf{M}_{\text{Ta230are}} &= \begin{pmatrix} 0.309 \\ 0.188 \\ 0.055 \end{pmatrix} \text{kg} \\ \\ \mathbf{M}_{\text{Th230inv}} &= \mathbf{M}_{\text{Th230inv}} &= \begin{pmatrix} 0.309 \\ 0.188 \\ 0.055 \end{pmatrix} \text{kg} \\ \\ \mathbf{A}_{\text{Th230inv}} &= \begin{pmatrix} 0.309 \\ 0.188 \\ 0.055 \end{pmatrix} \text{kg} \\ \\ \mathbf{A}_{\text{Th230inv}} &= \begin{pmatrix} 6.367 \times 10^{12} \\ 3.881 \times 10^{12} \\ 1.137 \times 10^{12} \end{pmatrix} \text{p Ci} \\ \\ \mathbf{A}_{\text{Th230avg}} &= \frac{\mathbf{A}_{\text{Th230inv}}}{\mathbf{M}_{\text{ThNj}}} & \mathbf{A}_{\text{Th230avg}} &= \begin{pmatrix} 2.685 \times 10^3 \\ 6.187 \times 10^3 \\ 5.204 \times 10^3 \end{pmatrix} \frac{\text{p Ci}}{\text{gm}} \end{split}$$

For an initial 230 Th activity of 1.0000×10^6 Bq, the following table displays the activities in the 230 Th decay sequence at 42 years:

Activities (in Bq)	after 42 years assuming an	Total activity (in Bq) aft	er 42 years assuming an
initial activity of	$1.0000 \times 10^6 \mathrm{Bq}$:	initial activity of 1.0000	\times 10 ⁶ Bq:
²³⁰ Th	999.6×10^{3}	²³⁰ Th series	1.131×10^6
²²⁶ Ra	18.02×10^3		
222 Rn	18.02×10^3		
²¹⁸ Po	18.02×10^3		
²¹⁴ Pb	18.01×10^3		
$^{214}\mathrm{Bi}$	18.01×10^3		
²¹⁴ Po	18.01×10^{3}		
²¹⁰ Pb	7.982×10^{3}		
$^{210}\mathrm{Bi}$	7.976×10^{3}		
²¹⁰ Po	7.806×10^{3}		

Source: Data obtained from the World Information Service on Energy, universal decay calculator, URL: http://www.antenna.nl/wise/uranium/rcc.html, accessed April 10, 2002.

From the data in the table, the activity of 222 Rn, $A_{Rn222Table}$, after 42 years of 230 Th decay, is 18.02×10^{-3} times the original activity of 230 Th, $A_{Th230inv}$.

$$A_{Rn222Table} = 18.02 \times 10^{-3} A_{Th230inv} \qquad A_{Rn222Table} = \begin{pmatrix} 1.147 \times 10^{11} \\ 6.993 \times 10^{10} \\ 2.05 \times 10^{10} \end{pmatrix} pCi$$

CONCLUSIONS

The calculated ²³⁰Th activities per gram of thorium nitrate are as follows:

- 2.685×10^3 pCi/gm for ore mined in South Africa and processed in the United States,
- 6.187×10^3 pCi/gm for ore mined in the Malagasy Republic and processed in France, and
- 5.204×10^3 pCi/gm for ore mined and processed in India.

REFERENCES

Benedict, M., T.H. Pigford, and H.W. Levi (1981). *Nuclear Chemical Engineering*, 2nd ed., McGraw-Hill, New York.

UIC (Uranium Information Centre Ltd.) 1998. "Mineral Sands," Nuclear Issues Briefing Paper 25, Melbourne, Australia, Jan. URL: http://www.uic.com.au/nip25.htm

Appendix C

THORIUM SAMPLE ANALYSIS INFORMATION

INTRODUCTION

In the spring and summer of 2002, a sampling and analysis program was conducted for the thorium nitrate stockpile. Representative samples were sent to Southwest Research Institute (SWRI), a laboratory accredited by Nevada Test Site (NTS), where the samples were analyzed for chemical and radiological constituents. This appendix presents background information, measured radiological characteristics assigned to variables for detailed computations, and the resulting average values for selected characteristics.

BACKGROUND

The Hammond inventory contains 863,685 kg of domestic thorium nitrate, and the Curtis Bay inventory contains 1,507,339 kg of domestic thorium nitrate. Both inventories originated from South African monazite ore.

$$M_{ThN_0}$$
 = 863,685 kg M_{ThN_0} = 8.637 × 10⁵ kg M_{ThN_1} = 1,507,339 kg M_{ThN_1} = 1.507 × 10⁶ kg

The Hammond inventory contains no French thorium nitrate. The Curtis Bay inventory contains 627,256 kg of French thorium nitrate that originated from ore mined in the Malagasy Republic.

$$M_{ThN_2} = 627,256 \, kg$$
 $M_{ThN_2} = 6.273 \times 10^5 \, kg$

The Hammond inventory contains no thorium nitrate from Indian ore, and the Curtis Bay inventory contains 218,556 kg that originated from ore mined in India.

$$M_{ThN_3} = 218,556 \text{ kg}$$
 $M_{ThN_3} = 2.186 \times 10^5 \text{ kg}$

HAMMOND DEPOT DOMESTIC—ICP/AES AND ICP/MS ANALYSES

Among the radiological analyses completed by SWRI, the thorium nitrate from the Hammond Depot domestic (HD) inventory was first. The ratios of thorium and uranium mass to

the mass of thorium nitrate, expressed in parts per million, were obtained using inductively coupled plasma/atomic emission spectroscopy (ICP/AES) and inductively coupled plasma/mass spectroscopy (ICP/MS) analyses, respectively, and are as follows.

$$ppm_{\text{Th232HD}} = \begin{pmatrix} 395,000 \\ 387,000 \\ 394,000 \\ 401,000 \\ 391,000 \\ 397,000 \\ 394,000 \end{pmatrix} \qquad ppm_{\text{Th232HD}} = \begin{pmatrix} 0.42 \\ 0.42 \\ 0.44 \\ 0.42 \\ 0.43 \\ 0.41 \\ 0.45 \end{pmatrix}$$

$$ppm_{\text{Th232HDavg}} = mean(ppm_{\text{Th232HD}}) \qquad ppm_{\text{Th232HDavg}} = 3.938 \times 10^{5}$$

HAMMOND DEPOT DOMESTIC—ALPHA SPECTROSCOPY

 $ppm_{U238HDavg} = mean(ppm_{U238HD})$

The activity per gram of domestic thorium nitrate from Hammond Depot is given below for ²²⁸Th, ²³⁰Th, ²³²Th, ²³⁴U, ²³⁵U, ²³⁶U, ²³⁸U, and total U. This information was obtained by SWRI using alpha spectroscopy.

 $ppm_{U238HDavg} = 4.275 \times 10^{-1}$

$$A_{\text{TM238HD}} = \begin{pmatrix} 54,600 \\ 49,200 \\ 46,800 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TM230HD}} = \begin{pmatrix} 4,470 \\ 3,570 \\ 3,550 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TM232HD}} = \begin{pmatrix} 49,000 \\ 44,500 \\ 44,200 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

$$A_{\text{U233HD}} = \begin{pmatrix} 1.86 \\ 1.32 \\ 0.505 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{U235HD}} = \begin{pmatrix} 0.0769 \\ 0.000 \\ 0.311 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{U236HD}} = \begin{pmatrix} 0.133 \\ -0.064 \\ 0.000 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

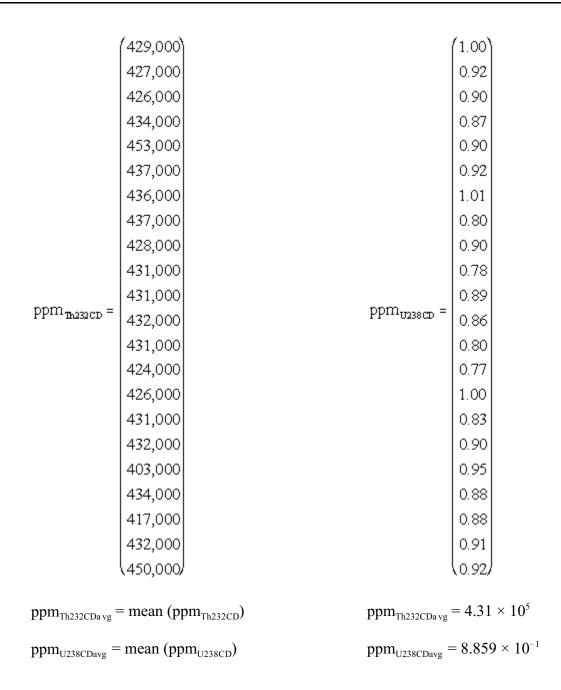
$$A_{\text{U236HD}} = \begin{pmatrix} 1.91 \\ 0.835 \\ 0.568 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TotalUHD}} = \begin{pmatrix} 3.98 \\ 2.09 \\ 1.38 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TotalThHD}} = \begin{pmatrix} 108,070 \\ 97,270 \\ 94,550 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

The ratio of thorium and uranium mass to mass of thorium nitrate from alpha spectroscopy analyses, in parts per million, are as follows:

$$\begin{aligned} & \text{ppm}_{\text{Th232HD1}} = \begin{pmatrix} 446,000 \\ 405,000 \\ 403,000 \end{pmatrix} & \text{ppm}_{\text{TotalUHD}} = \begin{pmatrix} 5.72 \\ 2.48 \\ 1.83 \end{pmatrix} \\ & \text{ppm}_{\text{Th232HD1avg}} = \text{mean(ppm}_{\text{Th232HD1}}) & \text{ppm}_{\text{Th232HD1avg}} = 4.180 \times 10^5 \\ & \text{ppm}_{\text{TotalUHD1avg}} = \text{mean(ppm}_{\text{TotalUHD1}}) & \text{ppm}_{\text{TotalUHD1avg}} = 3.343 \\ & \text{percent}_{\text{Th232HD1avg}} = \text{ppm}_{\text{Th232HD1avg}} 10^{-4} & \text{percent}_{\text{Th232HD1avg}} = 41.8 \\ & A_{\text{Th232HDavg}} = \text{mean(A}_{\text{Th232HD}}) & A_{\text{Th232HDavg}} = 4.59 \times 10^4 \frac{\text{p Ci}}{\text{gm}} \\ & A_{\text{U238HDavg}} = \text{mean(A}_{\text{U238HD}}) & A_{\text{U238HDavg}} = 1.104 \times 10^0 \frac{\text{p Ci}}{\text{gm}} \\ & A_{\text{Th230HDavg}} = \text{mean(A}_{\text{Th230HDavg}} = 3.863 \times 10^3 \frac{\text{p Ci}}{\text{gm}} \end{aligned}$$

CURTIS BAY DEPOT DOMESTIC—ICP/AES AND ICP/MS

The radiological analysis of the Curtis Bay Depot domestic (CD) inventory was completed by SWRI. The ratios of thorium and uranium mass to the mass of thorium nitrate, expressed in parts per million, were obtained using ICP/AES and ICP/MS analyses, respectively, and are as follows:



CURTIS BAY DEPOT DOMESTIC—ALPHA SPECTROSCOPY

Using alpha spectroscopy, SWRI obtained the activity per gram of domestic thorium nitrate from Curtis Bay Depot for ²²⁸Th, ²³⁰Th, ²³²Th, ²³⁴U, ²³⁵U, ²³⁶U, ²³⁸U, and total U:

$$A_{\text{TM238CD}} = \begin{pmatrix} 44,040 \\ 46,190 \\ 46,500 \\ 50,810 \\ 45,910 \\ 45,910 \\ 45,740 \\ 45,640 \end{pmatrix} \xrightarrow{p\text{Ci}} A_{\text{TM239CD}} = \begin{pmatrix} 4,058 \\ 4,436 \\ 4,579 \\ 4,984 \\ 4,099 \\ 4,156 \\ 4,121 \end{pmatrix} \xrightarrow{p\text{Ci}} A_{\text{TM332CD}} = \begin{pmatrix} 43,510 \\ 44,290 \\ 46,670 \\ 49,810 \\ 45,600 \\ 46,460 \\ 46,150 \\ 44,180 \end{pmatrix} \xrightarrow{p\text{Ci}} A_{\text{TM334CD}} = \begin{pmatrix} 0.30 \\ 1.31 \\ 0.71 \\ 0.86 \\ 0.82 \\ 0.78 \\ 0.78 \end{pmatrix}$$

$$A_{\text{TM238CD}} = \begin{pmatrix} 0.30 \\ 1.31 \\ 0.86 \\ 0.82 \\ 0.78 \\ 0.78 \end{pmatrix}$$

$$A_{\text{TM238CD}} = \begin{pmatrix} 0.22 \\ 0.40 \\ 0.00 \\ 0.11 \\ 0.28 \\ 0.00 \end{pmatrix}$$

$$A_{\text{TM335CD}} = \begin{pmatrix} 0.22 \\ 0.40 \\ 0.28 \\ 0.00 \end{pmatrix}$$

$$A_{\text{TM335CD}} = \begin{pmatrix} 0.22 \\ 0.62 \\ 0.25 \end{pmatrix}$$

$$A_{\text{TM335CD}} = \begin{pmatrix} 0.22 \\ 0.30 \\ 0.12 \\ 0.62 \\ 0.25 \end{pmatrix}$$

$$A_{\text{TM335CD}} = \begin{pmatrix} 0.22 \\ 0.33 \\ 0.00 \\ 0.12 \\ 0.62 \\ 0.25 \end{pmatrix}$$

$$A_{\text{TM335CD}} = \begin{pmatrix} 0.22 \\ 0.33 \\ 0.00 \\ 0.12 \\ 0.62 \\ 0.25 \end{pmatrix}$$

$$A_{\text{TM35CD}} = \begin{pmatrix} 0.24 \\ 0.00 \\ 0.11 \\ 0.30 \\ 0.45 \\ 0.22 \\ 0.25 \end{pmatrix}$$

$$A_{\text{TM335CD}} = \begin{pmatrix} 0.22 \\ 0.62 \\ 0.25 \\ 0.12 \\ 0.62 \\ 0.25 \end{pmatrix}$$

$$A_{\text{TM35CD}} = \begin{pmatrix} 91,608 \\ 94,916 \\ 97,749 \\ 105,604 \\ 95,969 \\ 96,046 \\ 93,761 \end{pmatrix}$$

$$A_{\text{TM35CD}} = \begin{pmatrix} 91,608 \\ 94,916 \\ 97,749 \\ 105,604 \\ 95,969 \\ 96,046 \\ 93,761 \end{pmatrix}$$

The ratio of thorium and uranium mass to the mass of thorium nitrate as determined by alpha spectroscopy analyses is as follows, in parts per million:

CURTIS BAY DEPOT FRENCH—ICP/AES AND ICP/MS

The radiological analysis of the Curtis Bay Depot French (CF) inventory was completed by SWRI. The ratios of thorium and uranium mass to the mass of thorium nitrate, expressed in parts per million, were obtained using ICP/AES and ICP/MS analyses, respectively, and are as follows:

$$\begin{array}{c} \left(\begin{array}{c} 411,000\\ 430,000\\ 417,000\\ 425,000\\ 426,000\\ 434,000\\ 426,000\\ 426,000\\ 426,000\\ 426,000\\ 426,000\\ 418,000\\ 418,000\\ 431,000\\ 431,000\\ 434,000\\ 410,000 \end{array}\right) \\ ppm_{\text{Th}232CFavg} = mean(ppm_{\text{Th}232CF}) \\ ppm_{\text{U238CFavg}} = mean(ppm_{\text{U238CF}}) \\ ppm_{\text{U238CFavg}} = 2.185 \times 10^{1} \\ \end{array}$$

The activity per gram of French thorium nitrate from Curtis Bay Depot is given below for ²²⁸Th, ²³⁰Th, ²³²Th, ²³⁴U, ²³⁵U, ²³⁶U, ²³⁶U, and total U. This information was obtained by Southwest Research Institute using alpha spectroscopy.

$$A_{\text{Th228CF}} = \begin{pmatrix} 46,530 \\ 43,080 \\ 44,580 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{Th230CF}} = \begin{pmatrix} 30,070 \\ 49,950 \\ 38,740 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{Th232CF}} = \begin{pmatrix} 41,220 \\ 43,530 \\ 45,440 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

$$A_{\text{U234CF}} = \begin{pmatrix} 7.83 \\ 4.07 \\ 14.48 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{U235CF}} = \begin{pmatrix} 0.73 \\ 0.12 \\ 1.42 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{U236CF}} = \begin{pmatrix} 0.33 \\ 0.52 \\ 0.46 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

$$A_{\text{U236CF}} = \begin{pmatrix} 8.65 \\ 2.45 \\ 14.23 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TotalUCF}} = \begin{pmatrix} 17.54 \\ 7.16 \\ 30.6 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TotalTHCF}} = \begin{pmatrix} 114,370 \\ 138,060 \\ 130,710 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

The ratio of thorium and uranium mass to the mass of thorium nitrate from alpha spectroscopy analyses, in parts per million, are as follows:

$$\begin{split} ppm_{\text{Th232CF}\alpha} &= \begin{pmatrix} 375,000 \\ 396,000 \\ 414,000 \end{pmatrix} \\ ppm_{\text{TotalUCF}\alpha} &= \begin{pmatrix} 26.09 \\ 7.35 \\ 43.02 \end{pmatrix} \\ ppm_{\text{Th232CF}\alpha\text{avg}} &= \text{mean}(\text{ppm}_{\text{Th232CF1}}) \\ ppm_{\text{TotalUCF}\alpha\text{avg}} &= 3.95 \times 10^5 \\ ppm_{\text{TotalUCF}\alpha\text{avg}} &= \text{mean}(\text{ppm}_{\text{TotalUCF1}}) \\ ppm_{\text{TotalUCF}\alpha\text{avg}} &= 25.487 \\ percent_{\text{Th232CF}\alpha\text{avg}} &= ppm_{\text{Th232CF1avg}} & 10^{-4} \\ \end{pmatrix} \\ ppm_{\text{TotalUCF}\alpha\text{avg}} &= 39.5 \\ A_{\text{Th232CFavg}} &= \text{mean}(A_{\text{Th232CF}}) \\ A_{\text{Th232CFavg}} &= \text{mean}(A_{\text{U238CFavg}}) \\ A_{\text{U238CFavg}} &= 8.443 \times 10^6 \frac{\text{p Ci}}{\text{gm}} \\ A_{\text{Th230CFavg}} &= \text{mean}(A_{\text{Th230CFavg}}) \\ A_{\text{Th230CFavg}} &= 3.959 \times 10^4 \frac{\text{p Ci}}{\text{gm}} \end{split}$$

CURTIS BAY DEPOT INDIAN—ICP/AES AND ICP/MS

The radiological analysis of the Curtis Bay Depot Indian (CI) inventory was completed by SWRI. The ratios of thorium and uranium mass to the mass of thorium nitrate, in parts per million, were obtained using ICP/AES and ICP/MS analyses, respectively, and are as follows:

$$ppm_{\text{Th232CI}} = \begin{pmatrix} 447,000 \\ 469,000 \\ 465,000 \\ 445,000 \\ 422,000 \\ 442,000 \\ 451,000 \\ 444,000 \\ 444,000 \\ 447,000 \end{pmatrix} \qquad ppm_{\text{U238CI}} = \begin{pmatrix} 0.94 \\ 0.79 \\ 0.85 \\ 0.93 \\ 0.94 \\ 0.89 \\ 0.91 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.93 \\ 0.94 \\ 0.94 \\ 0.93 \\ 0.94$$

$$\begin{aligned} ppm_{Th232Clavg} &= mean(ppm_{Th232Cl}) \\ ppm_{U238Clavg} &= mean(ppm_{U238Cl}) \end{aligned} \qquad \begin{aligned} ppm_{Th232Clavg} &= 4.476 \times 10^5 \\ ppm_{U238Clavg} &= 9.05 \times 10^{-1} \end{aligned}$$

CURTIS BAY DEPOT INDIAN—ALPHA SPECTROSCOPY

The activity per gram of Indian thorium nitrate from Curtis Bay Depot is given below for ²²⁸Th, ²³⁰Th, ²³²Th, ²³⁴U, ²³⁵U, ²³⁶U, ²³⁸U, and total U. This information was obtained by SWRI using alpha spectroscopy:

$$A_{\text{TM238CI}} = \begin{pmatrix} 47,330 \\ 48,180 \\ 49,880 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TM230CI}} = \begin{pmatrix} 7,532 \\ 7,632 \\ 7,450 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TM232CI}} = \begin{pmatrix} 44,770 \\ 44,580 \\ 48,530 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

$$A_{\text{U234CI}} = \begin{pmatrix} 0.66 \\ 0.35 \\ 0.51 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{U235CI}} = \begin{pmatrix} 0.00 \\ 0.03 \\ 0.13 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{U236CI}} = \begin{pmatrix} 0.37 \\ 0.10 \\ 0.03 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

$$A_{\text{U238CI}} = \begin{pmatrix} 0.22 \\ 0.02 \\ 0.10 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TotalUCI}} = \begin{pmatrix} 1.25 \\ 0.51 \\ 0.77 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{\text{TotalThCI}} = \begin{pmatrix} 99,632 \\ 100,392 \\ 105,860 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

The ratio of thorium and uranium mass to mass of thorium nitrate from alpha spectroscopy analyses, in parts per million, is as follows:

$$ppm_{\text{Th232CL}_{x}} = \begin{pmatrix} 408,000 \\ 406,000 \\ 442,000 \end{pmatrix} \qquad ppm_{\text{TotalUCL}_{x}} = \begin{pmatrix} 0.66 \\ 0.09 \\ 0.36 \end{pmatrix}$$

$$ppm_{\text{Th232CI}_{\alpha}\text{avg}} = mean(ppm_{\text{Th232CI1}}) \qquad ppm_{\text{Th232CI}_{\alpha}\text{avg}} = 4.187 \times 10^{5}$$

$$ppm_{\text{TotalUCI}_{\alpha}\text{avg}} = mean(ppm_{\text{TotalUCI1}}) \qquad ppm_{\text{TotalUCI}_{\alpha}\text{avg}} = 0.37$$

$$ppm_{\text{TotalUCI}_{\alpha}\text{avg}} = ppm_{\text{Th232CII}_{\alpha}\text{vg}} = 10^{-4} \qquad percent_{\text{Th232CI}_{\alpha}\text{avg}} = 41.87$$

$$A_{\text{Th232Clavg}} = \text{mean}(A_{\text{Th232Cl}}) \qquad A_{\text{Th232Clavg}} = 4.596 \times 10^4 \ \frac{\text{p Ci}}{\text{gm}}$$

$$A_{\text{U238Clavg}} = \text{mean}(A_{\text{U238Cl}}) \qquad A_{\text{U238Clavg}} = 1.133 \times 10^{-1} \ \frac{\text{p Ci}}{\text{gm}}$$

$$A_{\text{Th230Clavg}} = \text{mean}(A_{\text{Th230Clavg}}) \qquad A_{\text{Th230Clavg}} = 7.538 \times 10^3 \ \frac{\text{p Ci}}{\text{gm}}$$

ACTIVITY CONCENTRATION

The activity concentration—activity per unit volume—is obtained by multiplying the activity per unit mass by the density. The density measured by SWRI is 1.89 gm/cm^3 . The activity concentration is given by the symbol C with the appropriate identifying subscripts.

$$\begin{split} \rho_{\text{ThN}} &= 1.89 \; \frac{gm}{cm^3} \\ A_{\text{Th230HDavg}} &= 3.863 \times 10^3 \; \frac{p\,\text{Ci}}{gm} \qquad C_{230\text{HD}} = A_{\text{Th230HDavg}\,\rho\,\text{ThN}} \qquad C_{230\text{HD}} = 2.702 \times 10^8 \; \frac{B\,q}{m^3} \\ A_{\text{Th230CDavg}} &= 4.290 \times 10^3 \; \frac{p\,\text{Ci}}{gm} \qquad C_{230\text{CD}} = A_{\text{Th230CDavg}\,\rho\,\text{ThN}} \qquad C_{230\text{CD}} = 3.00 \times 10^8 \; \frac{B\,q}{m^3} \\ A_{\text{Th230CFavg}} &= 3.959 \times 10^4 \; \frac{p\,\text{Ci}}{gm} \qquad C_{230\text{CF}} = A_{\text{Th230CFavg}\,\rho\,\text{ThN}} \qquad C_{230\text{CF}} = 2.768 \times 10^9 \; \frac{B\,q}{m^3} \\ A_{\text{Th230CIavg}} &= 7.538 \times 10^3 \; \frac{p\,\text{Ci}}{gm} \qquad C_{230\text{CI}} = A_{\text{Th230CIavg}\,\rho\,\text{ThN}} \qquad C_{230\text{CI}} = 5.271 \times 10^8 \; \frac{B\,q}{m^3} \end{split}$$

CONCLUSIONS

The average ²³⁰Th activities per gram are as follows:

- 3.863×10^3 pCi/gm for ThN produced in the United States and stored at Hammond Depot,
- 4.290 × 10³ pCi/gm for ThN produced in the United States and stored at Curtis Bay Depot,
- 39.59 × 10³ pCi/gm for ThN produced in France and stored at Curtis Bay Depot, and
- 7.538×10^3 pCi/gm for ThN produced in India and stored at Curtis Bay Depot.

The average ²³⁰Th activity concentrations are as follows:

- 2.70×10^8 Bq/m³ for ThN produced in the United States and stored at Hammond Depot,
- 3.00 × 10⁸ Bq/m³ for ThN produced in the United States and stored at Curtis Bay Depot,
- 27.7×10^8 Bq/m³ for ThN produced in France and stored at Curtis Bay Depot, and
- 5.27×10^8 Bq/m³ for ThN produced in India and stored at Curtis Bay Depot.

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Appendix D

DECAY CALCULATIONS USING ORE DATA

INTRODUCTION

The Nevada Test Site (NTS) radon performance objective, $20 \text{ pCi} \cdot \text{m}^{-2} \cdot \text{sec}^{-1}$, applies to any time between the present and 1000 years in the future. Before the NTS model can be applied, the time-dependent levels of ^{222}Rn must be determined from potential ^{222}Rn sources: ^{230}Th and ^{238}U . The ^{230}Th activities are computed from preliminary ore data, and the ^{238}U activities are computed from previously measured mass spectroscopic data.

This appendix presents background information; evaluates the potential sources of ²²²Rn; determines the time-dependent activities of ²³⁰Th, ²²⁶Ra, and ²²²Rn; and compares the computed ²³²Th, ²³⁰Th and ²²⁶Ra activities to NTS action levels.

The density, ρ_{ThN} , of the thorium nitrate is 1.65 gm/cm³.

$$\rho_{ThN} = 1.65 \frac{gm}{cm^3}$$

$$Vol_{ThN} = \frac{M_{ThN}}{\rho_{ThN}} \qquad Vol_{ThN} = \begin{pmatrix} 1.437 \times 10^3 \\ 3.802 \times 10^2 \\ 1.325 \times 10^2 \end{pmatrix} m^3$$

 Vol_{ThN} is the volume of the thorium nitrate only. It does not include the volume of the drums.

POSSIBLE SOURCES OF RADON-222

There are two possible sources of ²²²Rn in the thorium nitrate inventory: ²³⁰Th and ²³⁸U carried over into the thorium nitrate during chemical processing. All the available ²³⁰Th is assumed to have been processed into the thorium nitrate. The ²³⁸U is an impurity and is assumed to occur at 1 ppm, the level measured earlier by Oak Ridge National Laboratory.

$$\mathrm{ppm}_{238\mathrm{Max}} = 100 \, \frac{\mathrm{mg}}{\mathrm{kg}} \qquad \qquad \mathrm{M}_{\mathrm{ThN}} = \begin{pmatrix} 2.371 \, \times \, 10^6 \\ 6.273 \, \times \, 10^5 \\ 2.186 \, \times \, 10^5 \end{pmatrix} \, \mathrm{kg}$$

$$\mathrm{M}_{238} = \sum_{\mathrm{j}} \mathrm{M}_{\mathrm{ThN}_{\mathrm{j}}} \mathrm{ppm}_{238\mathrm{Max}} \qquad \qquad \mathrm{M}_{238} = 3.217 \, \mathrm{kg}$$

The total ²³⁸U activity is given by $A_{238} = SpA_{1/238}M_{238}$. $A_{238} = 1.081 \times 10^9 \text{ pCi}$

The ²²²Rn activity may be obtained from readily available decay calculators.

For an initial 238 U activity of 1.000×10^6 Bq, the following table displays the activities in the 238 U decay sequence at 1000 years:

Activities (in Bq) after 1000 years assuming		Total activity (in Bq) after 1000 years assuming	
an initial activity of 1.000×10^6 Bq:		an initial activity of 1.000 × 10 ⁶ Bq:	
^{238}U	1.000×10^6	²³⁸ U series	3.003×10^6
²³⁴ Th	1.000×10^6		
^{234m} Pa	1.000×10^6		
^{234}U	2.826×10^{3}		
²³⁰ Th	12.86		
²²⁶ Ra	1.651		
²²² Rn	1.651		
²¹⁸ Po	1.651		
²¹⁴ Pb	1.650		
$^{214}\mathrm{Bi}$	1.650		
²¹⁴ Po	1.650		
²¹⁰ Pb	1.505		
$^{210}\mathrm{Bi}$	1.505		
²¹⁰ Po	1.503		

Source: Data obtained from the World Information Service on Energy, universal decay calculator, URL: http://www.antenna.nl/wise/uranium/rcc.html, accessed April 10, 2002.

From the decay information presented above, the decay of 238 U in the thorium nitrate inventory would produce a 222 Rn activity at 1000 years that is 1.651×10^{-6} the activity of the 238 U.

$$A_{222 from U238} = A_{238} 1.651 \ 10^{-6} \qquad \quad A_{222 from U238} = 1.786 \times 10^{3} \ pCi$$

CURRENT THORIUM-230 ACTIVITIES

The original number of 230 Th atoms in the thorium nitrate inventory is given by $N_{Th230inv}$.

$$N_{\text{Th230inv}} = \frac{A_{\text{Th230inv}}}{\lambda_{\text{Th230}}} \qquad \qquad N_{\text{Th230inv}} = \begin{pmatrix} 8.082 \times 10^{23} \\ 4.926 \times 10^{23} \\ 1.444 \times 10^{23} \end{pmatrix}$$

The mass of 230 Th in the thorium nitrate inventory is given by $M_{\it Th230inv}$.

$$\begin{split} \mathbf{M}_{\text{Th230inw}} &= \frac{\mathbf{N}_{\text{Th230inw}}}{\mathbf{N}_{\text{A}}} \, \mathbf{m}_{\text{Th230}} \\ \mathbf{M}_{\text{Th230inw}} &= \frac{\left(3.087 \times 10^{-1}\right)}{1.881 \times 10^{-1}} \, \text{kg} \\ \\ \mathbf{m}_{\text{totalTh230inw}} &= \sum_{\mathbf{j}} \mathbf{M}_{\text{Th230inw}} \, \mathbf{j} \\ \\ \mathbf{m}_{\text{totalTh230inw}} &= 5.519 \times 10^{-1} \, \text{kg} \\ \\ \mathbf{ppm}_{\text{Th230inw}} &= 1.0 \times 10^{6} \, \frac{\mathbf{M}_{\text{Th230inw}} \, \mathbf{j}}{\mathbf{M}_{\text{ThN}} \, \mathbf{j}} \\ \end{split} \qquad \qquad \mathbf{ppm}_{\text{Th230inw}} &= \begin{pmatrix} 0.130 \\ 0.300 \\ 0.252 \end{pmatrix} \end{split}$$

THORIUM-230 AS A FUNCTION OF TIME

The 230 Th content of the inventory, which varies with time, is given by $A_{Th230}(t)$,

$$A_{Th230}(t,j) = A_{Th230im_j} e^{-\lambda Th230t}$$

Changes in ²³⁰Th activity are evaluated for 100, 1,000, 10,000, and 100,000 years after separation from the ore. The ²³⁰Th produced by each ore type is computed separately.

$$\mathbf{t} = \begin{pmatrix} 100 \\ 1,000 \\ 10,000 \\ 100,000 \end{pmatrix} \quad \mathbf{y} \qquad \mathbf{i} = 0...3 \qquad \mathbf{A}_{\text{Th}230}(\mathbf{t}_{\mathbf{i}},0) = \begin{pmatrix} 6.361 \times 10 \\ 6.309 \times 10^{12} \\ 5.808 \times 10^{12} \\ 2.539 \times 10^{12} \end{pmatrix} \mathbf{pCi}$$

$$\mathbf{A}_{\text{Th}230}(\mathbf{t}_{\mathbf{i}},1) = \begin{pmatrix} 3.877 \times 10^{12} \\ 3.845 \times 10^{12} \\ 3.54 \times 10^{12} \\ 1.548 \times 10^{12} \end{pmatrix} \mathbf{pCi} \qquad \mathbf{A}_{\text{Th}230}(\mathbf{t}_{\mathbf{i}},2) = \begin{pmatrix} 1.136 \times 10^{12} \\ 1.127 \times 10^{12} \\ 1.037 \times 10^{12} \\ 4.536 \times 10^{11} \end{pmatrix} \mathbf{pCi}$$

The ²³⁰Th contained in a gram of the thorium nitrate inventory is given by $SpA_{230ThN}(t,j)$; (j = 0,1,2) refers to South African monazite ore, Malagasy Republic ore, and Indian ore, respectively,

$$SpA_{230 ThN}(t, j) = \frac{A_{Th230}(t, j)}{M_{ThN_j}}$$

The variation with time of the 230 Th activity per gram in the thorium nitrate inventory is given by SpA_{230ThN} . The times are 100, 1,000, 10,000, and 100,000 years after separation.

$$\operatorname{SpA}_{230\,\text{ThN}}(\mathbf{t_i},0) = \begin{pmatrix} 2.683 \times 10^3 \\ 2.661 \times 10^3 \\ 2.45 \times 10^3 \\ 1.071 \times 10^3 \end{pmatrix} \frac{\operatorname{pCi}}{\operatorname{gm}} \qquad \operatorname{SpA}_{230\,\text{ThN}}(\mathbf{t_i},1) = \begin{pmatrix} 6.181 \times 10^3 \\ 6.13 \times 10^3 \\ 5.643 \times 10^3 \\ 2.467 \times 10^3 \end{pmatrix} \frac{\operatorname{pCi}}{\operatorname{gm}}$$

$$SpA_{230 \text{ ThN}} (t_i, 2) = \begin{pmatrix} 5.199 \times 10^3 \\ 5.156 \times 10^3 \\ 4.747 \times 10^3 \\ 2.075 \times 10^3 \end{pmatrix} \frac{p \text{ Ci}}{\text{gm}}$$

RADIUM-226 ACTIVITY AS A FUNCTION OF TIME

The activity of 226 Ra as a function of time is given by $A_{Ra226}(t)$. A Bateman equation for two isotopes is used to compute the activity of 226 Ra assuming an original quantity of 230 Th and specified decay times.

$$A_{Ra226}(t,j) \; = \; \frac{\lambda_{Ra226} A_{Th230 \, \text{inv} \, j}}{\lambda_{Ra226} \; - \; \lambda_{Th230}} \left(e^{-\lambda \; Th230^{t}} \; - \; e^{-\lambda \; Ra226^{t}} \right)$$

The times for evaluation of the 226 Ra activity are 100, 1,000, 10,000, and 100,000 years after separation. The subscript j (j = 0,1,2) refers to South African monazite ore, Malagasy Republic ore, and Indian ore, respectively.

$$A_{\text{Ra226}}(t_{i},0) = \begin{pmatrix} 2.698 \times 10^{11} \\ 2.228 \times 10^{12} \\ 5.848 \times 10^{12} \\ 2.594 \times 10^{12} \end{pmatrix} \text{ pCi} \qquad A_{\text{Ra226}}(t_{i},1) = \begin{pmatrix} 1.664 \times 10^{11} \\ 1.358 \times 10^{11} \\ 3.564 \times 10^{12} \\ 1.581 \times 10^{12} \end{pmatrix} \text{ pCi}$$

$$A_{Ra226}(t_i, 2) = \begin{pmatrix} 4.82 \times 10^{10} \\ 3.979 \times 10^{11} \\ 1.045 \times 10^{12} \\ 4.634 \times 10^{11} \end{pmatrix} p \text{ G}$$

The variation with time of the ²²⁶Ra activity per gram in the thorium nitrate inventory is given by $SpA_{226ThN}(t)$, $SpA_{226ThN}(t,j) = \frac{A_{Ra226}(t,j)}{M_{ThN j}}$ The times are 100, 1,000, 10,000, and 100,000 years after separation.

$$\mathrm{SpA}_{226\,\mathrm{ThN}}(t_{i},0) = \begin{pmatrix} 1.138 \times 10^{2} \\ 9.395 \times 10^{2} \\ 2.467 \times 10^{3} \\ 1.094 \times 10^{3} \end{pmatrix} \frac{\mathrm{pCi}}{\mathrm{gm}} \qquad \mathrm{SpA}_{226\,\mathrm{ThN}}(t_{i},1) = \begin{pmatrix} 2.622 \times 10^{2} \\ 2.164 \times 10^{3} \\ 5.683 \times 10^{3} \\ 2.521 \times 10^{3} \end{pmatrix} \frac{\mathrm{pCi}}{\mathrm{gm}}$$

$$SpA_{226 \text{ThN}}(t_i, 2) = \begin{pmatrix} 2.205 \times 10^2 \\ 1.821 \times 10^3 \\ 4.78 \times 10^3 \\ 2.12 \times 10^3 \end{pmatrix} \frac{p \text{ Ci}}{\text{gm}}$$

RADON-222 ACTIVITY AS A FUNCTION OF TIME

The activity of 222 Rn as a function of time is given by $A_{Rn222}(t)$. A Bateman equation for three isotopes is used to compute the activity of 222 Rn assuming an original quantity of 230 Th and specified decay times. The coefficients of the Bateman equation are C_1 , C_2 , and C_3 .

$$\begin{split} &C_1 = \left(\lambda_{Ra226} - \lambda_{Th230}\right)^{-1} \left(\lambda_{Rn222} - \lambda_{Th230}\right)^{-1} & C_1 = 3.564 \times 10^1 \text{ y}^2 \\ &C_2 = \left(\lambda_{Th230} - \lambda_{Ra226}\right)^{-1} \left(\lambda_{Rn222} - \lambda_{Ra226}\right)^{-1} & C_2 = -3.564 \times 10^1 \text{ y}^2 \\ &C_3 = \left(\lambda_{Th230} - \lambda_{Rn222}\right)^{-1} \left(\lambda_{Ra226} - \lambda_{Rn222}\right)^{-1} & C_3 = 2.284 \times 10^{-4} \text{ y}^2 \\ &A_{Rn222} \left(t, j\right) = \lambda_{Ra226} \lambda_{Rn222} A_{Th230im j} \left(C_1 e^{-\lambda Th230t} + C_2 e^{-\lambda Ra226t} + C_3 e^{-\lambda Rn222t}\right) \end{split}$$

For each ore type [South African monazite (j = 0), Malagasy Republic (j = 1), and Indian (j = 2)], the activities of ²²²Rn at 100, 1,000, 10,000, and 100,000 years after separation from the ore are as follows.

$$A_{\text{Rx}222}(\mathbf{t_i},0) = \begin{pmatrix} 2.698 \times 10^{11} \\ 2.228 \times 10^{12} \\ 5.848 \times 10^{12} \\ 2.594 \times 10^{12} \end{pmatrix} \text{ pCi} \qquad A_{\text{Rx}222}(\mathbf{t_i},1) = \begin{pmatrix} 1.664 \times 10^{11} \\ 1.358 \times 10^{12} \\ 3.564 \times 10^{12} \\ 1.581 \times 10^{12} \end{pmatrix} \text{ p Ci}$$

$$A_{\text{Rx}222}(t_i, 2) = \begin{pmatrix} 4.819 \times 10^{10} \\ 3.979 \times 10^{11} \\ 1.045 \times 10^{12} \\ 4.634 \times 10^{11} \end{pmatrix} \text{ pCi}$$

The time-dependent amount of 222 Rn contained in a gram of the thorium nitrate inventory is given by $A_{222ThN}(t,j)$,

$$A_{222\text{TMM}}(t, j) = \frac{A_{\text{Ph}222}(t, j)}{M_{\text{TMM}_j}}$$

The evaluation times are 100, 1,000, 10,000, and 100,000 years after separation. The computations are performed, respectively, for South African monazite ore (j = 0), Malagasy Republic ore (j = 1), and Indian ore (j = 2).

$$A_{222\text{TMN}}(t_i,0) = \begin{pmatrix} 1.138 \times 10^2 \\ 9.395 \times 10^2 \\ 2.467 \times 10^3 \\ 1.094 \times 10^3 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{222\text{TMN}}(t_i,1) = \begin{pmatrix} 2.621 \times 10^2 \\ 2.164 \times 10^3 \\ 5.683 \times 10^3 \\ 2.521 \times 10^3 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

$$A_{222\text{ThN}}(t_{i},2) = \begin{pmatrix} 2.205 \times 10^{2} \\ 1.821 \times 10^{3} \\ 4.78 \times 10^{3} \\ 2.12 \times 10^{3} \end{pmatrix} \frac{\text{p Ci}}{\text{gm}}$$

$$A_{222Total} = \sum_{j} A_{222ThN} (1000\,y,\,j) \qquad \qquad A_{222Total} = 4.924 \times 10^3 \ \frac{p \, Ci}{gm}$$

The 222 Rn generated by decay of 230 Th, which was processed into the thorium nitrate directly from the ore, is given by $A_{222Th230}$.

$$A_{222 \text{from Th} 230} = A_{222 \text{Total}} \sum_{j} M_{\text{ThN } j} \qquad \qquad A_{222 \text{from Th} 230} = 1.584 \times 10^{13} \text{ pCi}$$

The ratio of 222 Rn activity resulting from decay of 230 Th to 222 Rn activity resulting from decay of 238 U is given by $R_{230to238}$.

$$R_{230\text{to}238} = \frac{A_{222\text{from Th}230}}{A_{232\text{from ID}28}} \qquad R_{230\text{to}238} = 8.872 \times 10^9$$

THORIUM-232 ACTION LEVEL

The 232 Th action level cited in Rev. 4 to the NTS Waste Acceptance Criteria (DOE 2002) is 8.1×10^8 Bq/m³.

The ²³²Th concentration in the thorium nitrate inventory is given by $C_{Th232} = SpA_{ThN}\rho_{ThN}$.

$$C_{Th232} = 2.736 \times 10^9 \frac{Bq}{m^3}$$
 $C_{Action232} = 8.1 \times 10^8 \frac{Bq}{m^3}$

The ratio of the volume activity of the thorium nitrate inventory to the NTS action level is given by

$$R_{CtoA} = \frac{C_{Th232}}{C_{Action232}}$$
 $R_{CtoA} = 3.378$

THORIUM-230 ACTION LEVEL

The 230 Th action level cited in Rev. 4 to the NTS Waste Acceptance Criteria (DOE 2002) is 9.6×10^7 Bq/m³. The concentration of 230 Th in the thorium nitrate inventory varies with time,

$$C_{230}(t,j) = \frac{A_{\text{Th}230}(t,j)\rho_{\text{ThN}}}{M_{\text{ThN}j}} ,$$

where (j = 0,1,2) refers to South African monazite ore, Malagasy Republic ore, and Indian ore, respectively.

$$C_{230}(t_i,0) = \begin{pmatrix} 1.638 \times 10^8 \\ 1.624 \times 10^8 \\ 1.495 \times 10^8 \\ 6.538 \times 10^7 \end{pmatrix} \quad \frac{Bq}{m^3} \qquad C_{230}(t_i,1) = \begin{pmatrix} 3.773 \times 10^8 \\ 3.742 \times 10^8 \\ 3.445 \times 10^8 \\ 1.506 \times 10^8 \end{pmatrix} \quad \frac{Bq}{m^3}$$

$$C_{230}(t_i,2) = \begin{pmatrix} 3.174 \times 10^8 \\ 3.148 \times 10^8 \\ 2.898 \times 10^8 \\ 1.267 \times 10^8 \end{pmatrix} \frac{Bq}{m^3}$$

$$C_{Action230} = 9.6 \times 10^7 \frac{Bq}{m^3}$$

The variation with time of the ratio of the ²³⁰Th concentration in the thorium nitrate inventory to the NTS action level for ²²⁶Ra is given by

$$R_{CTh230}(t,j) = \frac{C_{230}(t,j)}{C_{Action230}}$$

The times are 100, 1,000, 10,000, and 100,000 years after separation. The subscript j (j = 0,1,2) refers to South African monazite ore, Malagasy Republic ore, and Indian ore, respectively.

$$R_{CTM230}(t_i,0) = \begin{pmatrix} 1.706 \\ 1.692 \\ 1.558 \\ 0.681 \end{pmatrix} \qquad R_{CTM230}(t_i,1) = \begin{pmatrix} 3.931 \\ 3.898 \\ 3.589 \\ 1.569 \end{pmatrix} \qquad R_{CTM230}(t_i,2) = \begin{pmatrix} 3.306 \\ 3.279 \\ 3.019 \\ 1.32 \end{pmatrix}$$

RADIUM-226 ACTION LEVEL

The 226 Ra action level cited in Rev. 4 to the NTS Waste Acceptance Criteria (DOE 2002) is 3.6×10^7 Bg/m³.

$$C_{\text{Action226}} = 3.6 \times 10^7 \frac{\text{Bq}}{\text{m}^3}$$

The concentration of ²²⁶Ra in the thorium nitrate inventory varies with time,

$$C_{226}(t,j) = \frac{A_{Ra226}(t,j)\rho_{ThN}}{M_{ThN,j}},$$

where t = 100, 1,000, 10,000, and 100,000 years after separation from the ore.

$$C_{226}(t_i,0) = \begin{pmatrix} 6.9 \times 10^6 \\ 5.7 \times 10^7 \\ 1.5 \times 10^8 \\ 6.7 \times 10^7 \end{pmatrix} \quad \frac{Bq}{m^3} \qquad C_{226}(t_i,1) = \begin{pmatrix} 1.6 \times 10^7 \\ 1.3 \times 10^8 \\ 3.5 \times 10^8 \\ 1.5 \times 10^8 \end{pmatrix} \quad \frac{Bq}{m^3}$$

$$C_{226}(t_i,2) = \begin{pmatrix} 1.3 \times 10^7 \\ 1.1 \times 10^8 \\ 2.9 \times 10^8 \\ 1.3 \times 10^8 \end{pmatrix} \frac{Bq}{m^3}$$

The variation with time of the ratio of the ²²⁶Ra concentration in the thorium nitrate inventory to the NTS action level is given by

$$R_{226}(t,j) = \frac{C_{226}(t,j)}{C_{Action/226}}$$

The times are 100, 1,000, 10,000, and 100,000 years after separation. For each ore (South African monazite, Malagasy Republic, and Indian), between 100 and 1000 years, the increasing ratio reaches and exceeds 1; when this occurs, the ²²⁶Ra concentration is no longer compliant with the NTS action level.

$$R_{226}(t_{i},0) = \begin{pmatrix} 0.193 \\ 1.593 \\ 4.183 \\ 1.855 \end{pmatrix} \qquad R_{226}(t_{i},1) = \begin{pmatrix} 0.445 \\ 3.67 \\ 9.637 \\ 4.275 \end{pmatrix} \qquad R_{226}(t_{i},2) = \begin{pmatrix} 0.374 \\ 3.087 \\ 8.106 \\ 3.596 \end{pmatrix}$$

$$R_{226\text{total}} = \sum_{j} R_{226}(t, j)$$

$$R_{226\text{total}} = \begin{pmatrix} 1.012 \\ 8.351 \\ 21.925 \\ 9.726 \end{pmatrix}$$

The activity concentration of ²²⁶Ra in the thorium nitrate will be slightly in excess of the NTS action level at 100 years. The activity of ²²⁶Ra increases with time until roughly 10,000 years; the current activity concentration of ²²⁶Ra is less than the NTS action level.

CONCLUSIONS

The constituents of the thorium nitrate determined from ore data are near or well above the applicable NTS action levels. The current activity concentration of ²³²Th in the thorium nitrate inventory is about 4.2 times the NTS action level. The current activity concentration of

²³⁰Th in the thorium nitrate inventory is about 10.1 times the NTS action level. The current activity concentration of ²²⁶Ra is less than the NTS action level.

The decay of ²³⁸U is an insignificant source of ²²²Rn in the thorium nitrate inventory. At 1000 years, over 8.9 billion times more ²²²Rn results from the decay of ²³⁰Th that was present in the original ore body than from the decay of ²³⁸U.

REFERENCES

DOE (U.S. Department of Energy) 2002. *Nevada Test Site Waste Acceptance Criteria*, DOE/NV-325-Rev. 4, Nevada Operations Office, Waste Management Division, Feb. URL: http://www.nv.doe.gov/programs/envmgmt/RWAP/pdfs/DOENV325_REV4.pdf

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Appendix E

DECAY CALCULATIONS USING RECENT DATA

INTRODUCTION

The Nevada Test Site (NTS) radon performance objective, $20~pCi \cdot m^{-2} \cdot sec^{-1}$, applies to any time between the present and 1000~years in the future. Before the NTS model can be applied, the time-dependent levels of ^{222}Rn must be determined. These calculations use recent measurements of ^{230}Th and ^{238}U , potential ^{222}Rn sources, made by Southwest Research Institute (SWRI). This appendix presents background information; evaluates the potential sources of ^{222}Rn ; determines the time-dependent activities of ^{230}Th , ^{226}Ra , and ^{222}Rn ; and compares the computed ^{232}Th , ^{230}Th , and ^{226}Ra activities to NTS action levels.

BACKGROUND

The measured density, ρ_{ThN} , of the thorium nitrate is 1.89 gm/cm³. $\rho_{ThN} = 1.89 \frac{gm}{cm^3}$

$$Vol_{\text{ThN}} = \frac{M_{\text{ThN}}}{\rho_{\text{ThN}}} \qquad Vol_{\text{ThN}} = \begin{pmatrix} 4.57 \times 10^2 \\ 7.975 \times 10^2 \\ 3.319 \times 10^2 \\ 1.156 \times 10^2 \end{pmatrix} \text{ m}^3 \qquad j = 0...3$$

$$Vol = \sum_j Vol_{\text{ThN}_j} \qquad Vol = 1.702 \times 10^3 \text{ m}^3$$

 Vol_{ThN} is the volume of the thorium nitrate only. It does not include the volume of the drums.

POSSIBLE SOURCES OF RADON-222

There are two possible sources of ²²²Rn in the thorium nitrate inventory: ²³⁰Th and ²³⁸U carried over into the thorium nitrate during chemical processing. All the available ²³⁰Th is assumed to have been processed into the thorium nitrate. The ²³⁸U is an impurity and is assumed to occur at the highest concentration measured by SWRI for each portion of the inventory.

$$ppm_{238Max} = \begin{pmatrix} 0.45 \\ 1.01 \\ 29.0 \\ 0.94 \end{pmatrix} \frac{mg}{kg}$$

$$M_{ThN} = \begin{pmatrix} 8.637 \times 10^{5} \\ 1.507 \times 10^{6} \\ 6.273 \times 10^{5} \\ 2.186 \times 10^{5} \end{pmatrix} kg$$

$$M_{238} = M_{ThN}ppm_{238Max}$$

$$M_{238} = 20.307 kg$$

The total ²³⁸U activity is given by $A_{238} = SpA_{U238}M_{238}$. $A_{238} = 6.827 \times 10^9 \text{ pCi}$

The ²²²Rn activity may be obtained from readily available decay calculators.

For an initial 238 U activity of 1×10^6 Bq, the following table displays the activities in the 238 U decay sequence at 1000 years:

Activities (in Bq) after 1000 years assuming		Total activity (in Bq) after 1000 years assuming	
an initial activity of 1×10^6 Bq:		an initial activity of 1×10^6 Bq:	
^{238}U	1.000×10^6	238 U series 3.003×10^6	
²³⁴ Th	1.000×10^6		
^{234m} Pa	1.000×10^6		
^{234}U	2.826×10^{3}		
²³⁰ Th	12.86		
²²⁶ Ra	1.651		
222 Rn	1.651		
²¹⁸ Po	1.651		
²¹⁴ Pb	1.650		
$^{214}\mathrm{Bi}$	1.650		
²¹⁴ Po	1.650		
²¹⁰ Pb	1.505		
$^{210}\mathrm{Bi}$	1.505		
²¹⁰ Po	1.503		

Source: Data obtained from the World Information Service on Energy, universal decay calculator, URL: http://www.antenna.nl/wise/uranium/rcc.html, accessed Dec. 6, 2002.

From the decay information presented above, the decay of ^{238}U in the thorium nitrate inventory would produce a ^{222}Rn activity at 1000 years that is 1.651×10^{-6} the activity of the ^{238}U .

$$A_{222 from U238} = 1.651 \times 10^{-6} A_{238}$$
 $A_{222 from U238} = 1.127 \times 10^{4} pCi$

CURRENT THORIUM-230 ACTIVITIES

The ²³⁰Th activities are computed for each distinct portion of the thorium nitrate inventory. The average ²³⁰Th activity per gram of thorium nitrate measured by SWRI is multiplied by the total mass of thorium nitrate for the relevant portion of the inventory.

$$A_{\text{Th230invHD}} = \max(A_{\text{Th230invHD}}) \qquad A_{\text{Th230invHD}} = 4.47 \times 10^{3} \, \frac{\text{pCi}}{\text{gm}}$$

$$A_{\text{Th230invHD}} = A_{\text{Th230invHD}} M_{\text{ThN0}} \qquad A_{\text{Th230invHD}} = 3.861 \times 10^{12} \, \text{pCi}$$

$$A_{\text{Th230invCD}} = \max(A_{\text{Th230invCD}}) \qquad A_{\text{Th230invCD}} = 4.984 \times 10^{3} \, \frac{\text{pCi}}{\text{gm}}$$

$$A_{\text{Th230invCD}} = A_{\text{Th230invCD}} M_{\text{ThN1}} \qquad A_{\text{Th230invCD}} = 7.513 \times 10^{12} \, \text{pCi}$$

$$A_{\text{Th230invCF}} = \max(A_{\text{Th230invCF}}) \qquad A_{\text{Th230invCF}} = 4.995 \times 10^{4} \, \frac{\text{pCi}}{\text{gm}}$$

$$A_{\text{Th230invCF}} = A_{\text{Th230invCF}} M_{\text{ThN2}} \qquad A_{\text{Th230invCI}} = 3.133 \times 10^{13} \, \text{pCi}$$

$$A_{\text{Th230invCI}} = \max(A_{\text{Th230invCI}}) \qquad A_{\text{Th230invCI}} = 7.632 \times 10^{3} \, \frac{\text{pCi}}{\text{gm}}$$

$$A_{\text{Th230invCI}} = 7.632 \times 10^{3} \, \frac{\text{pCi}}{\text{gm}}$$

The ²³⁰Th activities are computed for the Hammond Depot domestic inventory, Curtis Bay Depot domestic inventory, Curtis Bay Depot French inventory, and Curtis Bay Depot Indian inventory, respectively. These computations are based on measurements made by SWRI.

$$A_{\text{Th/230 inw}} = \begin{pmatrix} 3.861 \times 10^{12} \\ 7.513 \times 10^{12} \\ 3.133 \times 10^{13} \\ 1.668 \times 10^{12} \end{pmatrix} \text{pCi}$$

THORIUM-230 AS A FUNCTION OF TIME

The ²³⁰Th content of the inventory, which varies with time, is given by $A_{Th230}(t,j)$, j=0...3), where the subscripts j=0,1,2,3 refer to Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot Indian inventories, respectively,

$$A_{Th230}(t,j) = A_{Th230im_j} e^{-\lambda Th230t}$$

$$t = \begin{pmatrix} 100 \\ 1,000 \\ 10,000 \\ 100,000 \end{pmatrix}$$
 y $i = 0...3$

The change in 230 Th activity with time is given by $A_{Th230}(t)$. The times are 100, 1,000, 10,000, and 100,000 years after analysis by SWRI. The 230 Th generated by each component of the inventory is computed separately.

$$A_{\text{Th}230}(t_{i},0) = \begin{pmatrix} 3.857 \times 10^{12} \\ 3.825 \times 10^{12} \\ 3.522 \times 10^{12} \\ 1.540 \times 10^{12} \end{pmatrix} pCi \qquad A_{\text{Th}230}(t_{i},1) = \begin{pmatrix} 7.506 \times 10^{12} \\ 7.444 \times 10^{12} \\ 6.853 \times 10^{12} \\ 2.996 \times 10^{12} \end{pmatrix} pCi$$

$$A_{\text{Th}230}(t_{i},2) = \begin{pmatrix} 3.130 \times 10^{13} \\ 3.104 \times 10^{13} \\ 2.858 \times 10^{13} \\ 1.250 \times 10^{13} \end{pmatrix} pCi \qquad A_{\text{Th}230}(t_{i},3) = \begin{pmatrix} 1.666 \times 10^{12} \\ 1.653 \times 10^{12} \\ 1.522 \times 10^{12} \\ 6.652 \times 10^{11} \end{pmatrix} pCi$$

The ²³⁰Th contained in a gram of the thorium nitrate inventory is given by $A_{230ThN}(t,j)$, where (j = 0,1,2,3) refers to Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot French, and Curtis Bay Depot Indian inventories, respectively,

$$A_{230 \text{ TeN}}(t, j) = \frac{A_{1230}(t, j)}{M_{1200}}.$$

The times for evaluation of the ²³⁰Th activity in the inventory are 100, 1,000, 10,000, and 100,000 years after analysis by SWRI.

$$A_{230\text{TeN}}(t_{i},0) = \begin{pmatrix} 4.466 \times 10^{3} \\ 4.429 \times 10^{3} \\ 4.077 \times 10^{3} \\ 1.783 \times 10^{3} \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{230\text{TeN}}(t_{i},1) = \begin{pmatrix} 4.979 \times 10^{3} \\ 4.938 \times 10^{3} \\ 4.546 \times 10^{3} \\ 1.988 \times 10^{3} \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

$$A_{230\,\text{ThN}}(t_i,2) = \begin{pmatrix} 4.99 \times 10^4 \\ 4.949 \times 10^4 \\ 4.556 \times 10^4 \\ 1.992 \times 10^4 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad A_{230\,\text{ThN}}(t_i,3) = \begin{pmatrix} 7.625 \times 10^3 \\ 7.562 \times 10^3 \\ 6.962 \times 10^3 \\ 3.044 \times 10^3 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

RADIUM-226 ACTIVITY AS A FUNCTION OF TIME

The activity of 226 Ra as a function of time is given by $A_{Ra226}(t,j)$. A Bateman equation for two isotopes is used to compute the activity of 226 Ra assuming an original quantity of 230 Th and specified decay times. The times are 100, 1,000, 10,000, and 100,000 years after analysis by SWRI. The subscript j (j = 0,1,2,3) refers to Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot French, and Curtis Bay Depot Indian inventories, respectively.

$$A_{Ra226}(t,j) \; = \; \frac{\lambda_{Ra226} A_{Th230 inv \; j}}{\lambda_{Ra226} \; - \; \lambda_{Th230}} \left(e^{-\lambda \; Th230^t} \; - \; e^{-\lambda \; Ra226^t} \right)$$

$$A_{Ra226}(t_{i},0) = \begin{pmatrix} 1.636 \times 10^{11} \\ 1.351 \times 10^{12} \\ 3.546 \times 10^{12} \\ 1.573 \times 10^{12} \end{pmatrix} pCi$$

$$A_{Ra226}(t_{i},1) = \begin{pmatrix} 3.184 \times 10^{11} \\ 2.628 \times 10^{12} \\ 6.9 \times 10^{12} \\ 3.061 \times 10^{12} \end{pmatrix} pCi$$

$$A_{Ra226}(t_{i},2) = \begin{pmatrix} 1.328 \times 10^{12} \\ 1.096 \times 10^{13} \\ 2.878 \times 10^{13} \\ 1.277 \times 10^{13} \end{pmatrix} pCi \qquad A_{Ra226}(t_{i},3) = \begin{pmatrix} 7.069 \times 10^{10} \\ 5.836 \times 10^{11} \\ 1.532 \times 10^{12} \\ 6.796 \times 10^{11} \end{pmatrix} pCi$$

The ²²⁶Ra contained in a gram of the thorium nitrate inventory is given by $A_{226ThN}(t)$.

$$A_{226 \text{ ThN}}(t, j) = \frac{A_{Ra226}(t, j)}{M_{ThN_j}}$$

The variation of ²²⁶Ra activity per gram is evaluated at 100, 1,000, 10,000, and 100,000 years after analysis by SWRI.

$$A_{226\,\text{TeN}}(t_i,0) = \begin{pmatrix} 1.894 \times 10^2 \\ 1.564 \times 10^3 \\ 4.106 \times 10^3 \\ 1.821 \times 10^3 \end{pmatrix} \quad \frac{p\,\mathrm{Ci}}{\mathrm{gm}} \qquad A_{226\,\text{TeN}}(t_i,1) = \begin{pmatrix} 2.112 \times 10^2 \\ 1.744 \times 10^3 \\ 4.578 \times 10^3 \\ 2.031 \times 10^3 \end{pmatrix} \quad \frac{p\,\mathrm{Ci}}{\mathrm{gm}}$$

$$A_{226\,\text{ThN}}\left(t_{i},2\right) = \begin{pmatrix} 2.117 \times 10^{3} \\ 1.748 \times 10^{4} \\ 4.588 \times 10^{4} \\ 2.035 \times 10^{4} \end{pmatrix} \quad \frac{p\text{Ci}}{\text{gm}} \qquad A_{226\,\text{ThN}}\left(t_{i},3\right) = \begin{pmatrix} 3.234 \times 10^{2} \\ 2.67 \times 10^{3} \\ 7.01 \times 10^{3} \\ 3.11 \times 10^{3} \end{pmatrix} \quad \frac{p\text{Ci}}{\text{gm}}$$

VARIATION OF RADON-222 ACTIVITY WITH TIME

The activity of 222 Rn as a function of time is given by $A_{Rn222}(t)$. A Bateman equation for three isotopes is used to compute the activity of 222 Rn assuming an original quantity of 230 Th and specified decay times. The coefficients of the Bateman equation are C_1 , C_2 , and C_3 .

$$C_1 = (\lambda_{Ra226} - \lambda_{Th230})^{-1} (\lambda_{Rn222} - \lambda_{Th230})^{-1}$$

$$C_1 = 3.564 \times 10^1 \text{ y}^2$$

$$C_2 = (\lambda_{Th230} - \lambda_{Ra226})^{-1} (\lambda_{Rn222} - \lambda_{Ra226})^{-1}$$

$$C_2 = -3.564 \times 10^1 \text{ y}^2$$

$$\begin{split} &C_3 = \left(\lambda_{\text{Th}230} \ - \ \lambda_{\text{Rn}222}\right)^{-1} \left(\lambda_{\text{Ra}226} \ - \ \lambda_{\text{Rn}222}\right)^{-1} \\ &A_{\text{Rn}222} \left(t,\,j\right) \ = \ \lambda_{\text{Ra}226} \lambda_{\text{Rn}222} A_{\text{Th}230\text{km}\,j} \left(C_1 e^{-\lambda \text{Th}230t} \ + \ C_2 e^{-\lambda \text{Ra}226t} \ + \ C_3 e^{-\lambda \text{Rn}222t}\right) \end{split}$$

For each portion of the thorium nitrate inventory (Hammond Depot domestic, Curtis Bay domestic, Curtis Bay French, and Curtis Bay Indian), the activities of ²²²Rn are computed at 100, 1,000, 10,000, and 100,000 years after analysis by SWRI.

$$A_{\text{Rn222}}(t_{i},0) = \begin{pmatrix} 1.636 \times 10^{11} \\ 1.351 \times 10^{12} \\ 3.546 \times 10^{12} \\ 1.573 \times 10^{12} \end{pmatrix} \text{pCi} \qquad A_{\text{Rn222}}(t_{i},1) = \begin{pmatrix} 3.183 \times 10^{11} \\ 2.628 \times 10^{12} \\ 6.900 \times 10^{12} \\ 3.061 \times 10^{12} \end{pmatrix} \text{pCi}$$

$$A_{\text{Rn222}}(t_{i},2) = \begin{pmatrix} 1.328 \times 10^{12} \\ 1.096 \times 10^{13} \\ 2.878 \times 10^{13} \\ 1.277 \times 10^{13} \end{pmatrix} \text{pCi} \qquad A_{\text{Rn222}}(t_{i},3) = \begin{pmatrix} 7.068 \times 10^{10} \\ 5.836 \times 10^{11} \\ 1.532 \times 10^{12} \\ 6.796 \times 10^{11} \end{pmatrix} \text{pCi}$$

The variation with time of the ²²²Rn activity per gram is given by

$$A_{222ThN}(t,j) = \frac{A_{Rn222}(t,j)}{M_{ThN_j}}$$
.

The times are 100, 1,000, 10,000, and 100,000 years after analysis by SWRI. The computations are performed for the following inventories, respectively, Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot Indian.

$$\mathbb{A}_{222\text{ThN}}(\mathbf{t_i},0) = \begin{pmatrix} 1.894 \times 10^2 \\ 1.564 \times 10^3 \\ 4.106 \times 10^3 \\ 1.821 \times 10^3 \end{pmatrix} \frac{\text{pCi}}{\text{gm}} \qquad \mathbb{A}_{222\text{ThN}}(\mathbf{t_i},1) = \begin{pmatrix} 2.112 \times 10^2 \\ 1.744 \times 10^3 \\ 4.578 \times 10^3 \\ 2.031 \times 10^3 \end{pmatrix} \frac{\text{pCi}}{\text{gm}}$$

$$A_{222\text{TMN}}(t_i,2) = \begin{pmatrix} 2.116 \times 10^3 \\ 1.747 \times 10^4 \\ 4.588 \times 10^4 \\ 2.035 \times 10^4 \end{pmatrix} \quad \frac{p\text{Ci}}{\text{gm}} \qquad A_{222\text{TMN}}(t_i,3) = \begin{pmatrix} 3.234 \times 10^2 \\ 2.670 \times 10^3 \\ 7.010 \times 10^3 \\ 3.110 \times 10^3 \end{pmatrix} \quad \frac{p\text{Ci}}{\text{gm}}$$

The total ²²²Rn activity at 1000 years is given by $A_{222Total}$.

$$A_{222Total} = \sum_{j} A_{222ThN} (1000\,\text{y},\,j) \qquad \qquad A_{222Total} = \ 2.345 \times 10^4 \ \frac{p\,\text{Ci}}{gm}$$

The 222 Rn generated by decay of 230 Th, which was processed into the thorium nitrate directly from the ore, is given by $A_{222Th230}$.

$$A_{222 \text{from Th} 230} = A_{222 \text{Total}} \sum_{j} M_{\text{ThN } j} \qquad \qquad A_{222 \text{from Th} 230} = 7.544 \times 10^{13} \text{ pCi}$$

The ratio of 222 Rn activity resulting from decay of 230 Th to 222 Rn activity resulting from decay of 238 U is given by $r_{230to238}$.

$$r_{230\text{to}238} = \frac{A_{222\text{from Th}230}}{A_{222\text{from II}28}}$$
 $r_{230\text{to}238} = 6.693 \times 10^9$

THORIUM-232 ACTION LEVEL

The 232 Th action level cited in Rev. 4 to the NTS Waste Acceptance Criteria (DOE 2002) is 8.1×10^8 Bq/m³. The 232 Th activity concentration (activity per unit volume) in the thorium nitrate inventory is calculated from the maximum concentrations measured by SWRI.

$$ppm_{232\text{Max}} = \begin{pmatrix} 401,000 \\ 453,000 \\ 453,000 \\ 465,000 \end{pmatrix} \frac{mg}{kg} \qquad \qquad M_{\text{ThN}} = \begin{pmatrix} 8.637 \times 10^5 \\ 1.507 \times 10^6 \\ 6.273 \times 10^5 \\ 2.186 \times 10^5 \end{pmatrix} kg$$

$$M_{232} = ppm_{232\text{Max}} M_{\text{ThN}} \qquad \qquad M_{232} = 1.415 \times 10^6 \text{ kg}$$

$$C_{232} = \frac{\text{SpA}_{\text{Th}232}\text{M}_{232}}{\text{Vol}} \qquad C_{232} = 3.387 \times 10^9 \ \frac{\text{Bq}}{\text{m}^3} \qquad C_{\text{action}} = 8.1 \times 10^8 \ \frac{\text{Bq}}{\text{m}^3}$$

$$r_{\text{CtoA}} = \frac{C_{232}}{C_{\text{action}}} \qquad \qquad r_{\text{CtoA}} = 4.182$$

THORIUM-230 ACTION LEVEL

The 230 Th action level cited in Rev. 4 to the NTS Waste Acceptance Criteria (DOE 2002) is 9.6×10^7 Bq/m³. The concentration (activity per unit volume) of 230 Th in the thorium nitrate inventory varies with time,

$$C_{230}(t,j) = \frac{A_{Th230}(t,j)\rho_{ThN}}{M_{ThN,j}} ,$$

where (j = 0,1,2,3) refers to Hammond Depot domestic inventory, Curtis Bay Depot domestic inventory, Curtis Bay Depot French inventory, and Curtis Bay Depot Indian inventory, respectively.

$$C_{230}(t_{i},0) = \begin{pmatrix} 3.123 \times 10^{8} \\ 3.097 \times 10^{8} \\ 2.851 \times 10^{8} \\ 1.247 \times 10^{8} \end{pmatrix} \frac{Bq}{m^{3}} \qquad C_{230}(t_{i},1) = \begin{pmatrix} 3.482 \times 10^{8} \\ 3.453 \times 10^{8} \\ 3.179 \times 10^{8} \\ 1.39 \times 10^{8} \end{pmatrix} \frac{Bq}{m^{3}}$$

$$C_{230}(t_{i},2) = \begin{pmatrix} 3.49 \times 10^{9} \\ 3.461 \times 10^{9} \\ 3.186 \times 10^{9} \\ 1.393 \times 10^{9} \end{pmatrix} \frac{Bq}{m^{3}} \qquad C_{230}(t_{i},3) = \begin{pmatrix} 5.332 \times 10^{8} \\ 5.288 \times 10^{8} \\ 4.868 \times 10^{8} \\ 2.128 \times 10^{8} \end{pmatrix} \frac{Bq}{m^{3}}$$

 $C_{Action230} = 9.6 \times 10^7 \frac{Bq}{m^3}$

For t = 0, the date of measurement,

$$A_{\text{OTh}230}(t,0) = 3.862 \times 10^{12} \text{ pCi}$$
 $A_{\text{OTh}230}(t,1) = 7.515 \times 10^{12} \text{ pCi}$

$$A_{0Th230}(t,2) = 3.134 \times 10^{13} \text{ pCi}$$
 $A_{0Th230}(t,3) = 1.669 \times 10^{12} \text{ pCi}$

The total current ²³⁰Th activity is given by $A_{0Th230Total}$.

$$A_{0Th230Total} = \sum_{i} A_{0Th230}(t, j)$$

$$A_{0Th230Total} = 4.439 \times 10^{13} \text{ pCi}$$

The current activity concentration of ²³⁰Th is given by $C_{0Th230Total}$.

$$C_{0 \text{Th} 230 \text{Total}} = \frac{A_{0 \text{Th} 230 \text{Total}} \rho_{\text{ThN}}}{\sum_{j} M_{\text{ThN} j}} \qquad C_{0 \text{Th} 230 \text{Total}} = 9.65 \times 10^{8} \frac{B \, q}{m^{3}}$$

The current activity concentration of 230 Th in the thorium nitrate exceeds the 230 Th action limit by the factor r_{230} .

$$r_{230} = \frac{C_{0Th230Total}}{C_{\text{Action 230}}} \qquad r_{230} = 10.1$$

RADIUM-226 ACTION LEVEL

The 226 Ra action level cited in Rev. 4 to the NTS Waste Acceptance Criteria (DOE 2002) is 3.6×10^7 Bq/m³.

$$C_{Action226} = 3.6 \times 10^7 \frac{Bq}{m^3}$$

The concentration of ²²⁶Ra in the thorium nitrate inventory varies with time,

$$C_{Ra226}(t,j) = \frac{A_{Ra226}(t,j)\rho_{ThN}}{M_{ThN}}$$
.

$$C_{\text{Ra226}}(t_{i},0) = \begin{pmatrix} 1.325 \times 10^{7} \\ 1.094 \times 10^{8} \\ 2.871 \times 10^{8} \\ 1.274 \times 10^{8} \end{pmatrix} \frac{Bq}{m^{3}} \qquad C_{\text{Ra226}}(t_{i},1) = \begin{pmatrix} 1.477 \times 10^{7} \\ 1.219 \times 10^{8} \\ 3.201 \times 10^{8} \\ 1.42 \times 10^{8} \end{pmatrix} \frac{Bq}{m^{3}}$$

$$C_{\text{Ra226}}(t_{i},2) = \begin{pmatrix} 1.48 \times 10^{8} \\ 1.222 \times 10^{9} \\ 3.208 \times 10^{9} \\ 1.423 \times 10^{9} \end{pmatrix} \frac{Bq}{m^{3}} \qquad C_{\text{Ra226}}(t_{i},3) = \begin{pmatrix} 2.262 \times 10^{7} \\ 1.867 \times 10^{8} \\ 4.902 \times 10^{8} \\ 2.175 \times 10^{8} \end{pmatrix} \frac{Bq}{m^{3}}$$

The variation with time of the ratio of the ²²⁶Ra concentration in the thorium nitrate inventory to the NTS action level is given by

$$r_{\text{CRa226}}(t,j) = \frac{C_{\text{Ra226}}(t,j)}{C_{\text{Action226}}} \ . \label{eq:cra226}$$

The times are 100, 1,000, 10,000, and 100,000 years after measurement by SWRI.

$$r_{\text{CRa226}}(t_i,0) = \begin{pmatrix} 0.368 \\ 3.038 \\ 7.976 \\ 3.538 \end{pmatrix} \qquad r_{\text{CRa226}}(t_i,1) = \begin{pmatrix} 0.41 \\ 3.387 \\ 8.893 \\ 3.945 \end{pmatrix}$$

$$r_{\text{CRa226}}(t_i,2) = \begin{pmatrix} 4.112 \\ 33.946 \\ 89.122 \\ 39.534 \end{pmatrix} \qquad r_{\text{CRa226}}(t_i,3) = \begin{pmatrix} 0.628 \\ 5.187 \\ 13.617 \\ 6.04 \end{pmatrix}$$

$$r_{\text{CRa226total}} = \sum_{j} r_{\text{CRa226}}(t, j)$$

$$r_{\text{CRa226total}} = \begin{bmatrix} 5.518 \\ 45.557 \\ 119.608 \\ 53.057 \end{bmatrix}$$

The activity concentration of ²²⁶Ra in the thorium nitrate will be slightly in excess of the NTS action level at 100 years. The activity of ²²⁶Ra increases with time until roughly 10,000 years; hence, the current activity concentration of ²²⁶Ra is near the NTS action level.

CONCLUSIONS

The constituents of the thorium nitrate are near or well above the applicable NTS action levels. The present activity concentration of ²³²Th in the thorium nitrate inventory is about 4.2 times the NTS action level. The current activity concentration of ²³⁰Th in the thorium nitrate inventory is about 10.1 times the NTS action level. The current activity concentration of ²²⁶Ra is near the NTS action level.

The decay of 238 U is an insignificant source of 222 Rn in the thorium nitrate inventory. At 1000 years, over 6.6 billion times more 222 Rn results from the decay of 230 Th than from the decay of 238 U.

REFERENCE

DOE (U.S. Department of Energy) 2002. *Nevada Test Site Waste Acceptance Criteria*, DOE/NV-325-Rev. 4, Nevada Operations Office, Waste Management Division, Feb. URL: http://www.nv.doe.gov/programs/envmgmt/RWAP/pdfs/DOENV325 REV4.pdf

Appendix F

RADON-222 EMISSION CALCULATIONS USING PRELIMINARY ORE DATA

INTRODUCTION

The Nevada Test Site (NTS) is required to limit the emissions of radon from buried materials. Radon emissions from uranium and thorium decay sequences increase with time. The performance objective for NTS burial sites is $20 \text{ pCi} \cdot \text{m}^2 \cdot \text{sec}^{-1}$ at 1000 years in the future.

The calculation of radon emissions has been performed for two cells at NTS: an upper cell, 26-ft depth, and a lower cell, 40-ft depth. The upper cell depth is typically used for burial of uranium and uranium-containing materials. The lower cell depth has been used for burial of thorium and thorium-containing materials. The data used for these calculations are derived from published estimates of the uranium content of thorium-containing ores.

This appendix presents the model used to perform the calculations, the computation of radon emissions from an upper cell, and the computation of radon emissions from a lower cell.

BACKGROUND

The February 1995 draft performance assessment (DOE 1995) provides the following information that may be used to determine the emission of ²²²Rn from a burial cell. The parameters are specific to the geometry of Pit 6 (the one used for thorium from Fernald).

$$J = 1 \times 10^{12} [K_{up} C_{up}(t) + K_{low} C_{low}(t)] \; (pCi \cdot m^{-2} \cdot sec^{-1}) \; , \label{eq:J}$$

where

J = surface 222 Rn flux from burial pit, pCi · m $^{-2}$ · sec $^{-1}$,

 K_{low} = ratio of surface flux to ²²⁶Ra activity concentration in the lower cell, m/sec, K_{up} = ratio of surface flux to ²²⁶Ra activity concentration in the upper cell, m/sec,

 C_{low}^{up} = activity concentration for ²²⁶Ra in the lower cell, Ci/m³, and C_{up} = activity concentration for ²²⁶Ra in the upper cell, Ci/m³.

$$K_{low} = 3.3 \times 10^{-11} \ \frac{m}{sec}$$
 $K_{up} = 5.3 \times 10^{-8} \ \frac{m}{sec}$

$$Vol_{ThN} = \begin{pmatrix} 1.437 \times 10^{3} \\ 3.802 \times 10^{2} \\ 1.325 \times 10^{2} \end{pmatrix} m^{3} \qquad t = \begin{pmatrix} 100 \\ 1,000 \\ 10,000 \\ 100,000 \end{pmatrix} y \qquad i = 0...3$$

EMISSIONS FROM THE UPPER CELL

The first evaluation of emissions is done assuming that the thorium nitrate inventory is placed in the upper cell.

$$C_{\mathbf{up}}(t,j) = \frac{A_{Ra226}(t,j)}{Vol_{ThN_j}}$$

The activity concentration equals the ²²⁶Ra activity divided by the volume of the thorium nitrate inventory produced from a specific ore: South African monazite, Malagasy Republic, or Indian.

 $C_{low}(t,j) = 0$ Calculations are performed below to determine the emissions from thorium nitrate buried in the lower cell.

$$J(t) = \left(K_{low} \cdot C_{low}(t) + K_{up} \cdot C_{up}(t)\right)$$

The conversion to picoCuries given in DOE (1995) has been ignored because the activity of ²²⁶Ra is already computed in picoCuries.

The values of $C_{up}(t)$ are computed at 100, 1,000, 10,000, and 100,000 years after separation from the ores: South African monazite, Malagasy Republic, and Indian. There is a peak in the 226 Ra concentration near 10,000 years.

$$\begin{split} \mathbf{C}_{\mathbf{up}}\left(\mathbf{t_{i}},0\right) &= \begin{pmatrix} 1.878 \times 10^{8} \\ 1.55 \times 10^{9} \\ 4.07 \times 10^{9} \\ 1.805 \times 10^{9} \end{pmatrix} \frac{\mathbf{pCi}}{\mathbf{m}^{3}} \qquad \mathbf{C}_{\mathbf{up}}\left(\mathbf{t_{i}},1\right) = \begin{pmatrix} 4.326 \times 10^{8} \\ 3.571 \times 10^{9} \\ 9.376 \times 10^{9} \\ 4.159 \times 10^{9} \end{pmatrix} \frac{\mathbf{pCi}}{\mathbf{m}^{3}} \\ \mathbf{C}_{\mathbf{up}}\left(\mathbf{t_{i}},2\right) &= \begin{pmatrix} 3.639 \times 10^{8} \\ 3.004 \times 10^{9} \\ 7.887 \times 10^{9} \\ 3.499 \times 10^{9} \end{pmatrix} \frac{\mathbf{pCi}}{\mathbf{m}^{3}} \end{split}$$

$$C_{low}(t_i,0) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad C_{low}(t_i,1) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad C_{low}(t_i,2) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix}$$

The 222 Rn emissions from the ground above the disposal cell, J(t,j), are computed at 100, 1,000, 10,000, and 100,000 years after separation from each of the ores: South African monazite, Malagasy Republic, and Indian. The subscript j (j = 0,1,2) refers to the ores in the sequence given in the previous sentence.

The NTS performance objective for radon is an emission of less than 20 pCi \cdot m⁻² \cdot sec⁻¹. $J_{obj} < 20 \frac{pCi}{m^2 \cdot sec}$

For the case of the upper cell only, $J(t,j) = K_{up} \cdot C_{up}(t,j)$.

$$\begin{split} J(t_i,0) &= \begin{pmatrix} 9.952 \times 10^0 \\ 8.216 \times 10^1 \\ 2.157 \times 10^2 \\ 9.568 \times 10^1 \end{pmatrix} \frac{p\text{Ci}}{\text{m}^2 \cdot \text{sec}} \qquad J(t_i,1) = \begin{pmatrix} 2.293 \times 10^1 \\ 1.893 \times 10^2 \\ 4.969 \times 10^2 \\ 2.204 \times 10^2 \end{pmatrix} \frac{p\text{Ci}}{\text{m}^2 \cdot \text{sec}} \\ J(t_i,2) &= \begin{pmatrix} 1.929 \times 10^1 \\ 1.592 \times 10^2 \\ 4.18 \times 10^2 \\ 1.854 \times 10^2 \end{pmatrix} \frac{p\text{Ci}}{\text{m}^2 \cdot \text{sec}} \end{split}$$

$$J_{\text{total}}(t) = \sum_{j} J(t, j) \qquad \qquad J_{\text{total}}(t) = \begin{pmatrix} 52 \\ 431 \\ 1131 \\ 502 \end{pmatrix} \; \frac{p \, \text{Ci}}{m^2 \cdot \text{sec}} \qquad J_{\text{total}}(1000 \; \text{y}) = 431 \; \frac{p \, \text{Ci}}{m^2 \cdot \text{sec}}$$

If the thorium nitrate inventory were buried in the upper cell, the ²²²Rn emissions would exceed the performance objective for all times examined.

EMISSIONS FROM THE LOWER CELL

The activity concentration equals the ²²⁶Ra activity divided by the volume of the thorium nitrate inventory produced from a specific ore: South African monazite, Malagasy Republic, or Indian.

$$C_{low}(t, j) = \frac{A_{Ra226}(t, j)}{Vol_{ThN_j}}$$
 $C_{up}(t, j) = 0$

Calculations are performed in the previous section to determine the impact if thorium nitrate were stored in the upper cell.

The conversion to picoCuries given in DOE (1995) has been ignored because the activity of ²²⁶Ra is already computed in picoCuries.

$$J(t) = \left(K_{low} \cdot C_{low}(t) + K_{up} \cdot C_{up}(t)\right)$$

$$C_{\mathbf{w}}(\mathbf{t_{i}},0) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad C_{\mathbf{w}}(\mathbf{t_{i}},1) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad C_{\mathbf{w}}(\mathbf{t_{i}},2) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix}$$

The values of $C_{uv}(t)$ are all zero because there is no upper disposal cell.

The values of $C_{low}(t)$ are computed at 100, 1,000, 10,000, and 100,000 years after separation from the ores: South African monazite, Malagasy Republic, and Indian. There is a peak in the 226 Ra concentration near 10,000 years.

$$C_{low}(t_i,0) = \begin{pmatrix} 1.878 \times 10^8 \\ 1.550 \times 10^9 \\ 4.070 \times 10^9 \\ 1805 \times 10^9 \end{pmatrix} \frac{p \, C_i}{m^3} \qquad C_{low}(t_i,1) = \begin{pmatrix} 4.326 \times 10^8 \\ 3.571 \times 10^9 \\ 9.376 \times 10^9 \\ 4.159 \times 10^9 \end{pmatrix} \frac{p \, C_i}{m^3}$$

$$C_{low}(t_i, 2) = \begin{pmatrix} 3.639 \times 10^8 \\ 3.004 \times 10^9 \\ 7.887 \times 10^9 \\ 3.499 \times 10^9 \end{pmatrix} \frac{p \, Ci}{m^3}$$

For the case of a lower cell only, the upper cell term is dropped, giving $J(t,j) = K_{low} \cdot C_{low}(t,j)$.

The 222 Rn emissions from the ground above the disposal cell, J(t,j), are computed at 100, 1,000, 10,000, and 100,000 years after separation from each of the ores: South African monazite, Malagasy Republic, and Indian. The subscript j (j = 0,1,2) refers to the ores in the sequence given in the previous sentence.

The NTS performance objective for radon is an emission of less than 20 pCi \cdot m⁻² \cdot sec⁻¹.

$$J_{obj} < 20 \frac{pCi}{m^2 \cdot sec}$$

$$J(t_i,0) = \begin{pmatrix} 0.0062 \\ 0.0512 \\ 0.1343 \\ 0.0596 \end{pmatrix} \frac{pCi}{m^2 \cdot sec} \quad J(t_i,1) = \begin{pmatrix} 0.0143 \\ 0.1179 \\ 0.3094 \\ 0.1373 \end{pmatrix} \frac{pCi}{m^2 \cdot sec} \quad J(t_i,2) = \begin{pmatrix} 0.012 \\ 0.0991 \\ 0.2603 \\ 0.1155 \end{pmatrix} \frac{pCi}{m^2 \cdot sec}$$

$$J_{\text{total}}(t) = \sum_{j} J(t, j) \qquad \qquad J_{\text{total}}(t) = \begin{pmatrix} 0.032 \\ 0.268 \\ 0.704 \\ 0.312 \end{pmatrix} \frac{pCi}{m^2 \cdot \text{sec}} \qquad J_{\text{total}}(1,000 \, \text{y}) = 0.268 \, \frac{pCi}{m^2 \cdot \text{sec}}$$

The ratio of the calculated 222 Rn emissions from the 40-ft-deep trench to the performance objective is given by r_{calc^2PO} . The ratios are calculated at 100, 1,000, 10,000, and 100,000 years after analysis of the thorium nitrate by Southwest Research Institute.

$$r_{calc2P0}\left(t\right) = \frac{J_{total}(t)}{J_{obj}} \qquad \qquad r_{calc2P0}\left(t\right) = \begin{pmatrix} 0.16\\1.34\\3.52\\1.56 \end{pmatrix} \% \qquad \qquad r_{calc2obj}\left(1000 \text{ y}\right) = 1.34\%$$

The total computed 222 Rn emissions from the thorium nitrate inventory, when buried in a 40-ft-deep trench, would be well below the NTS performance objective, 20 pCi · m⁻² · sec⁻¹, for all times considered.

CONCLUSIONS

The thorium nitrate inventory must be buried below the standard 26-ft depth for uranium. If the thorium nitrate were buried at the 40-ft depth used for the Fernald thorium, the modeled radon emissions from the thorium nitrate would be substantially below the NTS performance objective.

REFERENCE

DOE (U.S. Department of Energy) 1995. *Performance Assessment for the Area 5 Radioactive Waste Management Site at the Nevada Test Site*, Draft, PE-NTS-18, prepared by Reynolds Electrical and Engineering Co., Inc., Las Vegas, Nev., for the Waste Management Division, DOE Nevada Operations Office.

Appendix G

RADON-222 EMISSION CALCULATIONS USING MAXIMUM VALUES

INTRODUCTION

The Nevada Test Site (NTS) is required to limit the emissions of radon from buried materials. Radon emissions from uranium and thorium decay sequences increase with time. The performance objective for NTS burial sites is $20 \text{ pCi} \cdot \text{m}^{-2} \cdot \text{sec}^{-1}$ at 1000 years in the future.

The calculation of radon emissions has been performed for two cells at NTS: an upper cell, 26-ft depth, and a lower cell, 40-ft depth. The upper cell depth is typically used for burial of uranium and uranium-containing materials. The lower cell depth has been used for burial of thorium and thorium-containing materials. The data used for these calculations are derived from measurements made by the Southwest Research Institute (SWRI) on samples from the thorium nitrate stockpile.

This appendix presents the model used to perform the calculations, the computation of radon emissions from an upper cell, and the computation of radon emissions from a lower cell.

BACKGROUND

The February 1995 draft performance assessment (DOE 1995) provides the following information that may be used to determine the emission of ²²²Rn from a burial cell. The parameters are specific to the geometry of Pit 6 (the one used for thorium from Fernald).

$$J = 1 \times 10^{12} [K_{up} C_{up}(t) + K_{low} C_{low}(t)] (pCi \cdot m^{-2} \cdot sec^{-1}),$$

where

J = surface 222 Rn flux from burial pit, pCi · m $^{-2}$ · sec $^{-1}$,

 K_{low} = ratio of surface flux to ²²⁶Ra activity concentration in the lower cell, m/sec, K_{un} = ratio of surface flux to ²²⁶Ra activity concentration in the upper cell, m/sec,

 C_{low} = activity concentration for ²²⁶Ra in the lower cell, Ci/m³, and

 C_{up} = activity concentration for ²²⁶Ra in the upper cell, Ci/m³.

$$K_{low} = 3.3 \times 10^{-11} \frac{m}{sec}$$
 $K_{up} = 5.3 \times 10^{-8} \frac{m}{sec}$

$$Vol_{TMN} = \begin{pmatrix} 4.57 \times 10^{2} \\ 7.975 \times 10^{2} \\ 3.319 \times 10^{2} \\ 1156 \times 10^{2} \end{pmatrix} m^{3} \qquad t = \begin{pmatrix} 100 \\ 1,000 \\ 10,000 \\ 100,000 \end{pmatrix} y$$

EMISSIONS FROM THE UPPER CELL

The first evaluation of emissions is done assuming that the thorium nitrate inventory is placed in the upper cell.

$$C_{\mathbf{up}}\left(t,j\right) = \frac{A_{Ra226}(t,j)}{Vol_{ThN_{j}}}$$

The activity concentration equals the ²²⁶Ra activity divided by the volume of a specific portion of the thorium nitrate: Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot Indian inventories.

 $C_{low}(t,j) = 0$ Calculations are performed below to determine the emissions from thorium nitrate buried in the lower cell.

$$J(t) = \left(K_{low} \cdot C_{low}(t) + K_{up} \cdot C_{up}(t)\right)$$

The conversion to picoCuries given in DOE (1995) has been ignored because the activity of ²²⁶Ra is already computed in picoCuries.

The values of $C_{up}(t)$ are computed at 100, 1,000, 10,000, and 100,000 years after analysis by SWRI from the Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot French, and Curtis Bay Depot Indian inventories.

$$C_{\mathbf{up}}(t_{i},0) = \begin{pmatrix} 3.58 \times 10^{8} \\ 2.956 \times 10^{9} \\ 7.76 \times 10^{9} \\ 3.442 \times 10^{9} \end{pmatrix} \frac{p \, Ci}{m^{3}} \qquad C_{\mathbf{up}}(t_{i},1) = \begin{pmatrix} 3.992 \times 10^{8} \\ 3.296 \times 10^{9} \\ 8.652 \times 10^{9} \\ 3.838 \times 10^{9} \end{pmatrix} \frac{p \, Ci}{m^{3}}$$

$$C_{\mathbf{up}}(t_{i},2) = \begin{pmatrix} 4.001 \times 10^{9} \\ 3.303 \times 10^{10} \\ 8.671 \times 10^{10} \\ 3.847 \times 10^{10} \end{pmatrix} \frac{p \, Ci}{m^{3}} \qquad C_{\mathbf{up}}(t_{i},3) = \begin{pmatrix} 6.113 \times 10^{8} \\ 5.046 \times 10^{9} \\ 1.325 \times 10^{10} \\ 5.877 \times 10^{9} \end{pmatrix} \frac{p \, Ci}{m^{3}}$$

$$\mathbf{C}_{\text{low}}(\mathbf{t_i},0) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad \mathbf{C}_{\text{low}}(\mathbf{t_i},1) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad \mathbf{C}_{\text{low}}(\mathbf{t_i},2) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad \mathbf{C}_{\text{low}}(\mathbf{t_i},3) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix}$$

The ²²²Rn emissions from the ground above the disposal cell, J(t,j), are computed at 100, 1,000, 10,000, and 100,000 years after analysis by SWRI from the Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot Indian inventories. The subscript j (j = 0,1,2,3) refers to the analyzed materials in the sequence given in the previous sentence.

The NTS performance objective for radon is an emission of less than 20 pCi \cdot m⁻² \cdot sec⁻¹. $J_{obj} \le 20$

$$J_{obj} < 20 \frac{pCi}{m^2 \cdot sec}$$

For the case of the upper cell only, $J(t,j) = K_{up} \cdot C_{up}(t,j)$.

$$J(t_i,0) = \begin{pmatrix} 1.897 \times 10^1 \\ 1.567 \times 10^2 \\ 4.113 \times 10^2 \\ 1.824 \times 10^2 \end{pmatrix} \frac{p\text{Ci}}{m^2 \cdot \text{sec}} \qquad \qquad J(t_i,1) = \begin{pmatrix} 2.116 \times 10^1 \\ 1.747 \times 10^2 \\ 4.586 \times 10^2 \\ 2.034 \times 10^2 \end{pmatrix} \frac{p\text{Ci}}{m^2 \cdot \text{sec}}$$

$$J(t_i,2) = \begin{pmatrix} 2.12 \times 10^2 \\ 1.75 \times 10^3 \\ 4.596 \times 10^3 \\ 2.039 \times 10^3 \end{pmatrix} \frac{p\,\mathrm{Ci}}{m^2 \cdot \mathrm{sec}} \qquad \qquad J(t_i,3) = \begin{pmatrix} 3.24 \times 10^1 \\ 2.675 \times 10^2 \\ 7.022 \times 10^2 \\ 3.115 \times 10^2 \end{pmatrix} \frac{p\,\mathrm{Ci}}{m^2 \cdot \mathrm{sec}}$$

$$J_{total}(t) = \sum_{j} J(t, j) \qquad J_{total}(t) = \begin{pmatrix} 285 \\ 2349 \\ 6168 \\ 2736 \end{pmatrix} \frac{pCi}{m^2 \cdot sec} \qquad J_{total}(1000 \text{ y}) = 2349 \frac{pCi}{m^2 \cdot sec}$$

The ratio of the calculated 222 Rn emissions from the 40-ft-deep trench to the performance objective is given by $r_{calc2PO}$. The ratios are calculated at 100, 1,000, 10,000, and 100,000 years after analysis of the thorium nitrate by SWRI.

$$r_{\text{cak2PO}}(t) = \frac{J_{\text{total}}(t)}{J_{\text{obj}}}$$
 $r_{\text{cak2PO}}(t) = \begin{pmatrix} 1,423\\11,746\\30,839\\13,680 \end{pmatrix} \%$ $r_{\text{calc2PO}}(1000 \text{ y}) = 11,746\%$

If the thorium nitrate inventory were buried in the upper cell, the ²²²Rn emissions would exceed the performance objective for all times examined.

EMISSIONS FROM THE LOWER CELL

The second evaluation of emissions is done assuming that the thorium nitrate inventory is placed in the lower cell.

$$C_{low}(t, j) = \frac{A_{Ra226}(t, j)}{Vol_{ThN_j}}$$

The activity concentration equals the ²²⁶Ra activity divided by the volume of a specific portion of the thorium nitrate: Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot Indian inventories.

 $C_{up}(t,j) = 0$ It is assumed that there is no material in the upper cell.

$$J(t) = \left(K_{low} \cdot C_{low}(t) + K_{up} \cdot C_{up}(t)\right)$$

The conversion to picoCuries given in DOE (1995) has been ignored because the activity of ²²⁶Ra is already computed in picoCuries.

$$C_{\mathbf{w}}(\mathbf{t}_{\mathbf{i}},0) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad C_{\mathbf{w}}(\mathbf{t}_{\mathbf{i}},1) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad C_{\mathbf{w}}(\mathbf{t}_{\mathbf{i}},2) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \qquad C_{\mathbf{w}}(\mathbf{t}_{\mathbf{i}},3) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix}$$

The values of $C_{uv}(t)$ are all zero because there is nothing in the upper disposal cell.

The values of $C_{low}(t)$ are computed at 100, 1,000, 10,000, and 100,000 years after analysis by SWRI from the Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot French, and Curtis Bay Depot Indian inventories. There is a peak in the 226 Ra concentration near 10,000 years.

$$\begin{split} C_{low}(t_i,0) &= \begin{pmatrix} 3.580 \times 10^8 \\ 2.956 \times 10^9 \\ 7.760 \times 10^9 \\ 3.442 \times 10^9 \end{pmatrix} \frac{pCi}{m^3} \\ C_{low}(t_i,1) &= \begin{pmatrix} 3.992 \times 10^8 \\ 3.296 \times 10^9 \\ 8.652 \times 10^9 \\ 3.838 \times 10^9 \end{pmatrix} \frac{pCi}{m^3} \\ C_{low}(t_i,2) &= \begin{pmatrix} 4.001 \times 10^9 \\ 3.303 \times 10^{10} \\ 8.671 \times 10^{10} \\ 3.847 \times 10^{10} \end{pmatrix} \frac{pCi}{m^3} \\ C_{low}(t_i,3) &= \begin{pmatrix} 6.113 \times 10^8 \\ 5.046 \times 10^9 \\ 1.325 \times 10^{10} \\ 5.877 \times 10^9 \end{pmatrix} \frac{pCi}{m^3} \end{split}$$

For the case of material in the lower cell only, $J(t,j) = K_{low} \cdot C_{low}(t,j)$.

The ²²²Rn emissions from the ground above the disposal cell, J(t,j), are computed at 100, 1,000, 10,000, and 100,000 years analysis by SWRI from the Hammond Depot domestic, Curtis Bay Depot domestic, Curtis Bay Depot Indian inventories. The subscript j (j = 0,1,2,3) refers to the analyzed materials in the sequence given in the previous sentence.

The NTS performance objective for
222
Rn is an emission of less than $20 \text{ pCi} \cdot \text{m}^{-2} \cdot \text{sec}^{-1}$.
$$J_{obj} < 20 \frac{\text{pCi}}{\text{m}^2 \cdot \text{sec}}$$

$$J(t_i,0) = \begin{pmatrix} 0.0118 \\ 0.0975 \\ 0.2561 \\ 0.1136 \end{pmatrix} \frac{p\,\mathrm{Ci}}{m^2 \cdot \mathrm{sec}} \qquad \qquad J(t_i,1) = \begin{pmatrix} 0.0132 \\ 0.1088 \\ 0.2855 \\ 0.1267 \end{pmatrix} \frac{p\,\mathrm{Ci}}{m^2 \cdot \mathrm{sec}}$$

$$J(t_i, 2) = \begin{pmatrix} 0.132 \\ 1.0899 \\ 2.8615 \\ 1.2694 \end{pmatrix} \frac{pCi}{m^2 \cdot sec} \qquad \qquad J(t_i, 3) = \begin{pmatrix} 0.020 \\ 0.167 \\ 0.437 \\ 0.194 \end{pmatrix} \frac{pCi}{m^2 \cdot sec}$$

$$J_{\text{total}}(t) = \sum_{j} J(t, j) \qquad \qquad J_{\text{total}}(t) = \begin{pmatrix} 0.177 \\ 1.463 \\ 3.840 \\ 1.704 \end{pmatrix} \frac{pCi}{m^2 \cdot \text{sec}} \qquad J_{\text{total}}(1,000\,\text{y}) = 1.463 \ \frac{pCi}{m^2 \cdot \text{sec}}$$

The ratio of the calculated 222 Rn emissions from the 40-ft-deep trench to the performance objective is given by $r_{calc2PO}$. The ratios are calculated at 100, 1,000, 10,000, and 100,000 years after analysis of the thorium nitrate by SWRI.

$$r_{\text{cak}2P0}(t) = \frac{J_{\text{total}}(t)}{J_{\text{obj}}}$$
 $r_{\text{cak}2P0}(t) = \begin{pmatrix} 0.9\\7.3\\19.2\\8.5 \end{pmatrix}\%$ $r_{\text{calc}2PO}(1000 \text{ y}) = 7.3\%$

The total computed 222 Rn emissions from the thorium nitrate inventory, when buried in a 40-ft-deep trench, would be well below the NTS performance objective, 20 pCi · m⁻² · sec⁻¹, at the surface of the ground, for all times considered. These computations were performed using the maximum 230 Th values reported by SWRI.

CONCLUSIONS

The thorium nitrate inventory must be buried below the 26-ft depth that is standard for uranium. If the thorium nitrate were buried at the 40-ft depth used for the Femald thorium, the modeled radon emissions from the thorium nitrate would be substantially below the NTS performance objective.

REFERENCE

DOE (U.S. Department of Energy) 1995. Performance Assessment for the Area 5 Radioactive Waste Management Site at the Nevada Test Site, Draft, PE-NTS-18, prepared by Reynolds Electrical and Engineering Co., Inc., Las Vegas, Nev., for the Waste Management Division, DOE Nevada Operations Office.

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