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Dose Rate Outside Transport Cask 10-160B from Neutrons Generated in RH TRU Waste from ANL-E

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ABSTRACT

This report summarizes the calculations of the neutron dose rate outside the Transport Cask 10-160B. This cask contains remote-handled transuranic waste from Argonne National Laboratory-East. This waste includes the following radionuclides: Am-241; Pu-238, 239, 240, 241, 242; and U-233, 234, 235, 238. The dose rate from spontaneous fission neutrons is calculated as well as neutrons from induced fission, and the effect of neutron production from (α,n) reaction.

For a typical waste drum content normalized to one gram of Pu-239, the transporter cask surface neutron dose rate is expected to be 4.5E-04 mrem/hr. A few drums may contain up to 16 grams of Pu-239, in which case the cask surface neutron dose rate is expected to be approximately 0.1 mrem/hr. The calculations performed are conservative with respect to dose equivalent rate because it is assumed that the neutrons do not interact with the cask or the waste matrix.

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ACRONYMS

ANL-E	Argonne National Laboratory-East
RH	remote handled
TRU	transuranic waste

Dose Rate Outside Transport Cask 10-160B from Neutrons Generated in RH TRU Waste from ANL-E

1. DOSE RATE FROM SPONTANEOUS FISSION NEUTRONS

A typical (contents in Drums 728-737) isotopic distribution of spontaneous fission isotopes in remote handled (RH) transuranic (TRU) waste from Argonne National Laboratory-East (ANL-E) are in Table 1.

Table 1. Grams of radionuclides present in 10 drum batch (728–737).[1,2]

Radionuclide	Mass (g)
Am-241	5.69E-02
Pu-238	2.85E-03
Pu-239	3.83E+00
Pu-240	6.51E-01
Pu-241	4.27E-02[a]
Pu-242	1.02E-02
U-233	3.39E-06
U-234	7.05E-02
U-235	6.16E+00
U-238	8.19E+00

a. The activity was converted to mass using each radionuclide's specific activity.[3]

The spontaneous fission data for the radionuclides are listed in Table 2.

Table 2. Spontaneous fission data for the radionuclides.[4]

Radionuclides	Protons (Z)	Neutrons (N)	Spontaneous Fission Half-Life (yr)	Spontaneous Fission Yield (n/s-g)
Am-241	95	146	1.05E+14	1.18E+00
Pu-238	94	144	4.77E+10	2.59E+03
Pu-239	94	145	5.48E+15	2.18E-02
Pu-240	94	146	1.16E+11	1.02E+03
Pu-241	94	147	2.50E+15	5.00E-02
Pu-242	94	148	6.84E+10	1.72E+03
U-233	92	141	1.27E+17	8.60E-04
U-234	92	142	2.10E+16	5.02E-02
U-235	92	143	3.50E+17	2.99E-04
U-238	92	146	8.20E+15	1.36E-02

The neutron energy for spontaneous fission of Pu-240 and thermal-neutron induced fission of U-233, U-235, and Pu-239 is 1.3 MeV.[4] Normalizing the mass data in Table 1 to Pu-239 yields the data presented in Table 3.

Table 3. The mass ratio of the radionuclides normalized to Pu-239 mass.

Radionuclide	Mass (g)	Mass Ratio to Pu-239
Am-241	5.69E-02	1.49E-02
Pu-238	2.85E-03	7.44E-04
Pu-239	3.83E+00	1.00E+00
Pu-240	6.51E-01	1.70E-01
Pu-241	4.27E-02	1.11E-02
Pu-242	1.02E-02	2.66E-03
U-233	3.39E-06	8.85E-07
U-234	7.05E-02	1.84E-02
U-235	6.16E+00	1.61E+00
U-238	8.19E+00	2.14E+00

The spontaneous fission yield for the radionuclide is obtained by multiplying mass ratio of individual radionuclide to the spontaneous fission yield (n/s-g). The spontaneous fission yield for the mass ratio of each radionuclide normalized to Pu-239 is shown in Table 4.

Table 4. Spontaneous fission yield for the radionuclide normalized to Pu-239.

Radionuclide	Spontaneous Fission Yield – SP (n/s-g Pu-239)
Am-241	1.75E-02
Pu-238	1.93E+00
Pu-239	2.18E-02
Pu-240	1.73E+02
Pu-241	5.57E-04
Pu-242	4.58E+00
U-233	7.61E-10
U-234	9.24E-04
U-235	4.81E-04
U-238	2.91E-02
Total	179.58

Summing the total number of spontaneous fission (normalized to Pu-239) listed in Table 4 yields 179.58 n/s per 1 g Pu-239.

To obtain the unscattered and unattenuated neutron flux at the outside surface of the cask, the following equation is used:

$$\sum_i SP_i \Big/ 2\pi rh$$

The outside radius of the CNS 10-160B Cask is about 100 cm (3.3 ft) and the height of the cask is assumed 148 cm (4.9 ft) based on 30-gal drums stacked two high.

$$\sum_i SP_i \Big/ 2\pi rh = 179.58/(2 \times 3.1416 \times 100 \times 148) = 1.93\text{E-03 n/cm}^2\text{–s}$$

To obtain the conversion factor for fluence per unit dose equivalent, the energy of the neutron spectra must be known. The average neutron energy is approximately 1.3 MeV.[4] Although the correct procedure would be to obtain the calculated conversion factor at 1.3 Mev, a more conservative approach is to use the conversion factor at 1 MeV. The neutron fluence per unit dose equivalent at 1 Mev[5] is:

27E+06 n/cm^{-2}-rem^{-1}.

Therefore neutron flux to dose equivalent rate conversion factor at 1 MeV is:

(1/27E+06) × 1000 = 3.75E-5 mrem/cm^2-n.

Maximum dose equivalent rate from neutrons at surface of cask is:

1.93E-03 × 3.75E-05 × 3600 = 2.61E-04 mrem/hr.

2. EFFECT OF NEUTRON PRODUCTION FROM (α,n) REACTIONS

There are two (α,n) reactions that can occur with in any matrix; i.e. an oxide reaction and a fluoride reaction. Based on acceptable knowledge, the fluoride (α,n) reaction will not occur because there is no fluorine in the waste. The oxide (α,n) reaction is discussed in this section. Using the data in Table 1, Table 3, and Table 5 a normalized oxide yield can be obtained.

Table 5. Oxide (α,n) reaction yields. [4]

Radionuclide	Mass (g)	Ratio to 1 g Pu-239	Alpha Yield (α/s-g)	Average α Energy (MeV)	(α,n) Yield in Oxide (n/s-g)	(α,n) Yield in Oxide (n/s-1 g Pu-239)
Am-241	5.69E-02	1.49E-02	1.30E+11	5.48E+00	2.69E+03	4.00E+01
Pu-238	2.85E-03	7.44E-04	6.40E+11	5.49E+00	1.34E+04	9.97E+00
Pu-239	3.83E+00	1.00E+00	2.30E+09	5.15E+00	3.81E+01	3.81E+01
Pu-240	6.51E-01	1.70E-01	8.40E+09	5.15E+00	1.41E+02	2.40E+01
Pu-241	4.27E-02	1.11E-02	9.40E+07	4.89E+00	1.30E+00	1.45E-02

Table 5. (continued).

Radionuclide	Mass (g)	Ratio to 1 g Pu-239	Alpha Yield (α/s-g)	Average α Energy (MeV)	(α,n) Yield in Oxide (n/s-g)	(α,n) Yield in Oxide (n/s-1 g Pu-239)
Pu-242	1.02E-02	2.66E-03	1.40E+08	4.90E+00	2.00E+00	5.33E-03
U-233	3.39E-06	8.85E-07	3.50E+08	4.82E+00	4.80E+00	4.25E-06
U-234	7.05E-02	1.84E-02	2.30E+08	4.76E+00	3.00E+00	5.52E-02
U-235	6.16E+00	1.61E+00	7.90E+04	4.40E+00	7.10E-04	1.14E-03
U-238	8.19E+00	2.14E+00	1.20E+04	4.19E+00	8.30E-05	1.77E-04
					Total	112.15

To determine if a reaction is possible the Q value must be determined. The reactions of interest are shown below:

$^{16}O(\alpha,n)^{19}Ne$
$^{17}O(\alpha,n)^{20}Ne$
$^{18}O(\alpha,n)^{21}Ne$.

The Q-value [6] equation is as follows:

$Q = BE_Y + BE_y - BE_X - BE_x$.

Where:

BE = binding energy (MeV)

Y = is the radioisotope produced in the reaction

y = is the neutron being released in the reaction

X = the original radioisotope

x = is the alpha particle.

The Q values for these reactions are listed in Table 6.

Table 6. Q values for oxide (α,n) reaction.

Path	Q (MeV)
16O(α,n)19Ne	-12.1
17O(α,n)20Ne	0.6
18O(α,n)21Ne	-0.7

Because the O-16 reaction has a large negative Q value its reaction is not possible. Once the Q value is obtained it is added to the average alpha energy and multiplied by its natural abundance to get the neutron energy. The abundance of O-17 and O-18 were normalized to 1. The neutron energies determined by each reaction for the radionuclides listed in Table 1 are shown in Table 7. The last column in Table 7 is the summation of the two energies based on their abundance.

Table 7. Neutron energy for oxide (α,n) reaction.

Radionuclide	O-17 Neutron Energy (MeV)	O-18 Neutron Energy (MeV)	Total Neutron Energy (MeV)
U-233	9.01E-01	3.44E+00	4.34E+00
U-234	8.91E-01	3.39E+00	4.28E+00
U-235	8.31E-01	3.09E+00	3.92E+00
U-238	7.96E-01	2.91E+00	3.71E+00
Pu-238	1.01E+00	3.99E+00	5.01E+00
Pu-239	9.56E-01	3.71E+00	4.67E+00
Pu-240	9.56E-01	3.71E+00	4.67E+00
Pu-241	9.13E-01	3.49E+00	4.41E+00
Pu-242	9.14E-01	3.50E+00	4.42E+00
Am-241	1.01E+00	3.99E+00	5.00E+00

A weighted average of the energy will yield approximately 4 MeV neutron energy.

Summing the total number of oxide (α,n) reactions (normalized to Pu-239) listed in Table 5 yields:

112.15 n/s per 1 g Pu-239.

The unscattered and unattenuated neutron flux at the outside surface of the cask is:

$112.15/(2 \times 3.1416 \times 100 \times 148) = 1.21E\text{-}03$ n/cm^2–s.

To obtain the conversion factor for fluence per unit dose equivalent, the energy of the neutron spectra must be known, which is approximately 4 MeV. The conversion factor for 5 MeV was used because it provides a more conservative number. The neutron fluence per unit dose equivalent at 5 Mev is 23E+06 n/cm^{-2}-rem^{-1}.[5]

Therefore neutron flux to dose equivalent rate conversion factor at 5 MeV is:

$(1/23E+06) \times 1000 = 4.35E\text{-}05$ mrem/cm^2-n.

Maximum dose equivalent rate from neutrons at surface of cask is:

$1.21E\text{-}03 \times 4.35E\text{-}05 \times 3600 = 1.89\ E\text{-}04$ mrem/hr.

The total dose equivalent rate from neutrons from spontaneous fission and (α,n) reactions is:

2.61E-04 + 1.89E-04 = 4.5E-04 mrem/hr.

3. NEUTRONS FROM INDUCED FISSION

In determining the induced fission reactions, the fast neutron and thermal neutron components must be considered. The total number of neutrons produced in the 30-gal drum are determined based on the spontaneous fission and the (α,n) contribution. The end product is two dose equivalent rates, one for the fast neutron component and one for the induced thermal component.

Based on calculations, the fast component and induced thermal component do not contribute to the dose equivalent in the third significant digit, therefore it is not included.

4. CONCLUSION

For a typical ANL-E RH TRU waste drum containing 1 g of Pu-239 inside a 10-160B cask, the cask surface neutron dose rate is expected to 4.5E-04 mrem/hr. A few of the ANL-E RH TRU drums may contain up to 16 g of Pu-239. Even for these high plutonium drums, loaded in the 10-160B cask, the neutron dose rate at the cask surface is expected to be approximately 0.1 mrem/hr (4.5E-05 × 10 drums × 16 g/drum). The calculations performed are conservative with respect to dose equivalent rate because it is assumed that the neutrons do not interact with the cask or the waste matrix.

5. REFERENCES

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