ORAU Team **Dose Reconstruction Project for NIOSH** Estimation of Neutron Dose Rates from Alpha-Neutron Reactions in Uranium and Thorium Compounds	Document Number: ORAUT-OTIB-0024 Effective Date: 04/07/2005 Revision No.: 00 Controlled Copy No.:_____ Page 1 of 16
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RECORD OF ISSUE/REVISIONS

ISSUE AUTHORIZATION DATE	EFFECTIVE DATE	REV. NO.	DESCRIPTION
Draft	12/06/2004	00-A	New technical information bulletin to provide estimation of neutron doses from uranium and thorium compounds that contain low atomic number components. Initiated by Cindy W. Bloom.
Draft	01/13/2004	00-B	Updated document based on ORAU Team comments. Clarified intent of OTIB, added additional clarifying information, and a comparison of the measured to calculated dose rate. Initiated by Robert Hysong.
Draft	03/11/2005	00-C	Updated document based on OCAS comments. Corrected formatting errors and added generic words regarding OTIBs to the purpose. Initiated by Cindy W. Bloom.
Draft	03/11/2005	00-D	Corrected references to tables. Initiated by Robert Hysong.
04/07/2005	04/07/2005	00	First approved issue. Initiated by Cindy W. Bloom.

1.0 PURPOSE

Technical Information Bulletins (TIBs) are general working documents that provide guidance concerning the preparation of dose reconstructions at particular sites or categories of sites. TIBs will be revised in the event additional relevant information is obtained. TIBs may be used to assist NIOSH in the completion of individual dose reconstructions.

The purpose of this document is to provide a quick estimate of neutron doses from alpha particle collisions with low atomic number materials. This document provides an estimate of neutron doses at sites that processed thorium and uranium compounds with low atomic number components, but did not perform neutron measurements.

2.0 SCOPE

This TIB estimates neutron production and dose rates from the alpha-neutron reaction in uranium and thorium compounds. The chemical forms of uranium considered were UF_4 and UF_6 as well as the uranium oxide forms (UO_2, UO_3, U_3O_8) and soda salt ($Na_2U_2O_7$). Little information could be found about the chloride form (UCl_4); as a consequence, UCl_4 is considered by identifying another target material (chemical form) that bounds its neutron emission rate. Based on neutron yield data for ^{210}Po and ^{234}U sources, the neutron emission rate of the chloride form would fall below the oxide form (see Tables 2-1 and 2-2). Consequently, it is reasonable to assume that the neutron emission rate from UCl_4 is bounded by UF_4. Thorium in the form of a fluoride (ThF_4) and a nitrate ($Th(NO_3)_4$) were also analyzed. Little information could be found on the neutron yield for nitrogen. Based on neutron yield data for a ^{210}Po source and a nitrogen target shown in Table 2-2 below, the neutron emission rate from the nitrogen form is seven times lower than the oxygen yield. As a consequence, the use of the oxygen yield alone for thorium nitrate compounds, $Th(NO_3)_4$, will produce a slight overestimate of neutron yield.

Table 2-1. Neutron yields for trace impurities in uranium –234.[a]

Target Element	Neutron Yield per 10^6 Alphas (4.7 MeV Alpha)	Neutron Yield (n/s-Ci)	Neutron Yield (n/s-g)
Li	1.60E-01	5.92E+03	3.70E+01
Be	4.40E+01	1.63E+06	1.02E+04
B	1.24E+01	4.59E+05	2.87E+03
C	5.10E-02	1.89E+03	1.18E+01
O	4.00E-02	1.48E+03	9.25E+00
F	3.10E+00	1.15E+05	7.17E+02
Na	5.00E-01	1.85E+04	1.16E+02
Mg	4.20E-01	1.55E+04	9.71E+01
Cl	1.00E-02	3.70E+02	2.31E+00

a. Adapted from DOE 2000.

Table 2-2. Neutron yields for various elements and a polonium–210 source.[a]

Target Element	Neutron Yield per 10^6 alphas (5.3 MeV Alpha)	Neutron Yield (n/s-Ci)	Neutron Yield (n/s-g)
Li	2.60E+00	9.62E+04	4.32E+08
Be	8.00E+01	2.96E+06	1.33E+10
B	2.40E+01	8.88E+05	3.99E+09
C	1.10E-01	4.07E+03	1.83E+07
O	7.00E-02	2.59E+03	1.16E+07
F	1.20E+01	4.44E+05	1.99E+09
Na	1.50E+00	5.55E+04	2.49E+08
Mg	1.40E+00	5.18E+04	2.33E+08
N	1.00E-02	3.70E+02	1.66E+06
Cl	1.10E-01	4.07E+03	1.83E+07

a. Adapted from Auguston and Reilly 1974.

As long as there is an adequate amount of available target material (fluorine, oxygen, sodium, or chlorine) and that material is intimately mixed with the source material (uranium or thorium), neutron production essentially depends on the amount of the source material present. This TIB analysis made the bounding assumption that there is an adequate amount of target material in all the chemical forms analyzed which results in the maximum neutron emission rate.

Neutron dose rates have been calculated for sources of natural uranium, natural thorium, and a mixture of thorium isotopes with and without alpha-emitting progeny contribution. Dose rates have been calculated for natural uranium (^{238}U, ^{234}U, and ^{235}U) assuming the following: (1) no alpha-emitting progeny are present; (2) alpha-emitting progeny are present in secular equilibrium through ^{226}Ra and ^{223}Ra; and (3) alpha-emitting progeny are present in secular equilibrium through ^{210}Po and ^{211}Po. Dose rates have been calculated for natural thorium (^{232}Th and ^{228}Th) assuming the presence of alpha-emitting progeny in secular equilibrium through ^{224}Ra and assuming the presence of alpha-emitting progeny in secular equilibrium through ^{212}Po. Finally, the neutron dose rate for a mixture of thorium isotopes containing natural thorium (^{232}Th and ^{228}Th) and ^{230}Th has been evaluated assuming the presence of alpha-emitting progeny in secular equilibrium through ^{224}Ra and assuming the presence of alpha-emitting progeny in secular equilibrium through ^{212}Po. Radon (^{222}Rn and ^{223}Rn) is present in the decay chains of natural uranium (^{238}U and ^{235}U), and ^{220}Rn is present in the natural thorium decay chain (^{232}Th). As an inert gas, radon would not be chemically bound with the target material; as a consequence, actual neutron yield contribution might be lower from radon and alpha-emitting radon progeny than calculated here. The presence or absence of progeny and their degree of equilibrium will vary depending on site-specific processes and practices. As a consequence, the presence of progeny and degree of equilibrium should be determined on a site-specific basis.

3.0 NEUTRON ENERGY SPECTRUM

Neutron production increases with the available alpha particle energy as indicated by comparing the neutron yields in Table 2-1 (4.7 MeV alpha) with the yields in Table 2-2 (5.3 MeV alpha) for the same target element. Likewise, the average and maximum neutron energies resulting from an alpha neutron reaction in a target element increase with increasing alpha energy. The resulting neutron energies also vary with the angle of neutron emission resulting in a fairly continuous spectrum of neutron energies from very low energies to the maximum neutron energy. Table 3-1 presents experimental results of the average and maximum neutron energies resulting from 4.0, 4.5, 5.0 and 5.5 MeV alpha particles bombarding oxygen and fluorine targets. Table 3-2 presents the average and maximum neutron energies from several alpha-neutron sources and various target elements. The data in Tables 3-1 and 3-2 are in fair agreement; however, the highest alpha energy considered in this TIB is 8.78 MeV from ^{212}Po in the ^{232}Th decay chain. Consequently, the average and maximum neutron energies shown in Table 3-1 below are expected to increase by a factor determined by linear interpolation of the data in Table 3-2 resulting in the average and maximum neutron energies shown in Table 3-3. Even though higher energy neutrons can be produced by higher energy alpha particles, the fluence per unit dose equivalent is very insensitive to the neutron energies considered in the ^{232}Th decay chain (Mlekodaj 2002). The neutron fluence per unit dose equivalent conversion factor varies by only 14% in the 1.0- to 10.0-MeV neutron energy range as indicated in Table 3-4 below (Shleien, Slaback, and Birky 1998).

Table 3-1. Average and maximum (α,n) neutron energies (MeV) for various alpha particle energies and oxygen and fluorine targets.[a]

Source Alpha Energy (MeV)	Oxygen Target Element (MeV)		Fluorine Target Element (MeV)	
	Average	Maximum	Average	Maximum
4.0	1.71	3.10	0.73	1.80
4.5	1.89	3.60	1.00	2.30
5.0	1.86	4.00	1.12	2.70
5.5	2.02	4.50	1.21	3.20

a. Jacobs and Liskien 1983

Table 3-2. Average and maximum (α,n) neutron energies (MeV) for ^{238}Pu, ^{239}Pu and ^{241}Am alpha particles and oxygen, fluorine and sodium targets.[a]

Source Alpha Energy (MeV)	Oxygen Target (MeV)		Fluorine Target (MeV)		Sodium Target (MeV)	
	Average	Maximum	Average	Maximum	Average	Maximum
^{238}Pu (5.5 MeV)	2.0	5.8	1.3	3.2	--	--
^{239}Pu (5.2 MeV)	1.9	5.5	1.4	2.8	--	1.8
^{241}Am (5.48 MeV)	--	--	1.3	2.5	--	--

a. NRC 1991
-- No Data

Table 3-3. Average and maximum (α,n) neutron energies (MeV) for 4.0 to 8.8 MeV alpha sources and oxygen and fluorine targets.[a]

Source Alpha Energy (MeV)	Oxygen Target Element (MeV)		Fluorine Target Element (MeV)	
	Average	Maximum	Average	Maximum
4.0	1.71	3.10	0.73	1.80
4.5	1.89	3.60	1.00	2.30
5.0	1.86	4.00	1.12	2.70
5.5	2.02	4.50	1.21	3.20
6.0	2.10	4.95	1.41	3.65
6.5	2.19	5.41	1.56	4.11
7.0	2.28	5.90	1.72	4.57
7.5	2.37	6.33	1.87	5.03
8.0	2.46	6.79	2.03	5.49
8.5	2.55	7.25	2.19	5.95
8.8	2.60	7.53	2.28	6.23

a. Based on Scaling Data from Table 2-3 from Jacobs and Liskien 1983

Table 3-4 Mean quality factors, Q, and fluence per unit dose equivalent for monoenergetic neutrons.

Neutron Energy (MeV)	Quality Factor (Q)	Fluence per Unit Dose Equivalent (neutron cm^{-2} rem^{-1})	Dose Equivalent per Unit Fluence (rem per neutron cm^{-2})	Dose Equivalent Rate per Unit Neutron Flux (rem h^{-1} per neutron s^{-1} cm^{-2})
2.5E-08	2.0	9.8E+08	1.0E-09	3.7E-06
1.0E-07	2.0	9.8E+08	1.0E-09	3.7E-06
1.0E-06	2.0	8.1E+08	1.2E-09	4.4E-06
1.0E-05	2.0	8.1E+08	1.2E-09	4.4E-06
1.0E-04	2.0	8.4E+08	1.2E-09	4.3E-06
1.0E-03	2.0	9.8E+08	1.0E-09	3.7E-06
1.0E-02	2.5	1.0E+09	9.9E-10	3.6E-06
1.0E-01	7.5	1.7E+08	5.9E-09	2.1E-05
5.0E-01	11	3.9E+07	2.6E-08	9.2E-05
1.0E+00	11	2.7E+07	3.7E-08	1.3E-04
2.5E+00	9	2.9E+07	3.4E-08	1.3E-04
5.0E+00	8	2.3E+07	4.3E-08	1.6E-04

Neutron Energy (MeV)	Quality Factor (Q)	Fluence per Unit Dose Equivalent (neutron cm^{-2} rem^{-1})	Dose Equivalent per Unit Fluence (rem per neutron cm^{-2})	Dose Equivalent Rate per Unit Neutron Flux (rem h^{-1} per neutron s^{-1} cm^{-2})
7.0E+00	7	2.4E+07	4.2E-08	1.5E-04
1.0E+01	6.5	2.4E+07	4.2E-08	1.5E-04
1.4E+01	7.5	1.7E+07	5.9E-08	2.1E-04
2.0E+01	8	1.6E+07	6.3E-08	2.3E-04

a. Adapted from Shleien, Slaback, and B. K. Birky 1998 and NCRP 1971.

The following assumptions were made to convert from neutron production rate per gram of source material to neutron dose rate at 1- and 3-ft distances.

1. Point source geometry has been assumed to calculate the neutron flux at distances of 1 and 3 feet from the source.

2. The average neutron energy from alpha-neutron reactions is 2.0 MeV; consequently a dose equivalent rate –to– fluence rate conversion factor for 2.0 MeV neutrons of 1.3×10^{-4} rem/hr per neutron/cm^2-s (Shleien, Slaback, and Birky 1998) was used to determine dose rates. Self-shielding (attenuation and scatter) within the source-target compound matrix is assumed to be negligible. This is a very conservative assumption, because containers exceeding a few kilograms of material will result in a lower neutron energy spectrum and lower neutron flux outside the container due to neutron scatter and attenuation within the source.

3. Dose rate calculations are based on a source of one gram of natural uranium or thorium (source material). The entire mass of uranium compounds can be assumed conservatively to be attributable to the uranium present in the compounds considered here. Site-specific adjustments for mass might be necessary for some thorium compounds because the mass of thorium in a solution of thorium nitrate ($Th(NO_3)_4$) represents only 48% of a gram of $Th(NO_3)_4$ and 75% of a gram of ThF_4. Similarly, the mass of uranium in a solution of uranyl nitrate [$UO_2(NO_3)_2$] represents only 60% of the total mass of the solution.

A Microsoft® Excel spreadsheet, Alpha-NforTIB12-01-04Draft2.xls, was created to perform the calculations in this TIB. The spreadsheet is available to calculate neutron production rates and doses from modified source compositions and is useful for inputting known masses of material to obtain an estimate of neutron dose rate.

4.0 NEUTRON YIELDS

Only a limited amount of thick target (α,n) yield data is available in the literature. Table 4-1 below presents neutron yield data from Table 6-5 of DOE Standard –1136, 2000, Salmon and Hermann, 1992, and Auguston and Reilly, 1974. The neutron yield data presented in DOE 2000 were originally published in Table 11-3 of *Passive Nondestructive Assay of Nuclear Materials*, NUREG/CR-5550 (NRC 1991). The neutron yields in the DOE 2000 are based on pure oxides and fluorides and do not account for either the neutron contribution from spontaneous fission neutrons from ^{238}U, ^{235}U and ^{234}U, or the presence of other low-atomic-number impurities (DOE 2000). The ratio of neutron yield data for oxygen in Salmon and Hermann (1992) to the yield data reported by DOE (2000) ranges from 2.8 to 2.9. The ratio of neutron yield data for oxygen in the Auguston and Reilly (1974) to the yield data in Salmon and Hermann (1992) are in good agreement ranging from 0.9 to 0.93.

The ratio of neutron yield data for fluorine in Salmon and Hermann (1992) to the yield data in DOE (2000) ranges from 0.6 to 1.9. The ratio of the neutron yield data for fluorine in Salmon and Hermann (1992) to the yield data in Augustin and Reilly (1974) are in better agreement ranging from 0.4 to 1.1.

Table 4-2 below presents neutron yield data for a ^{234}U source and several target elements from Table 6-6 of DOE 2000, Salmon and Hermann 1992, and Auguston and Reilly 1974. The yield data from DOE 2000, Table 6-6 are in very good agreement with the Salmon and Hermann (1992) and Auguston and Reilly (1974) data. The yields from Salmon and Hermann (1992) are greater than the DOE 2000, Table 6-6 data by a factor of only 0.89 to 1.11.

The neutron yield data from Salmon and Hermann is used in the ALPHN computer code to calculate the neutron production rate in canisters of vitrified high level waste. Some degree of code verification was obtained from the ORIGEN-S code on the same example problem. The difference in results was 0.5% (Salmon and Hermann 1992). Validation of the ORIGEN-S code by comparing its results to the results of experiments conducted at Savannah River Laboratory has also been performed. The experiments used glass composition containing a uniformly distributed known amount of ^{238}Pu. The total (α,n) neutron source predicted by ORIGEN-S was 2% greater than the average of three samples measured at Savannah River Laboratory. These results can be taken as some degree of validation of ALPHN (Salmon and Hermann 1992). Consequently, the neutron yield data from Salmon and Hermann (1992) has been chosen to calculate neutron dose rates in this TIB. In addition, the Salmon and Hermann (1992) yield data appears to be in good agreement with Table 6-6 of DOE 2000, and with Auguston and Reilly 1974. Lastly, the Salmon and Hermann (1992) reference provides a method to calculate the neutron yields for all of the decay progeny in this TIB based on the average alpha energy of the source radionuclide.

Table 4-1. Neutron yields in neutrons per second per gram of source isotope (n/s-g)[a].

Source Isotope/Ave. Alpha Energy (MeV)	DOE Standard Table 6-5 Target		Salmon and Hermann Table 3 Interpolation Target		Auguston and Reilly Tables 7.1 and 7.2 Target	
	Oxygen	Fluoride	Oxygen	Fluoride	Oxygen	Fluoride
^{234}U 4.77	3.00E+00	5.80E+02	8.62E+00	7.81E+02	9.15E+00	1.56E+03
^{238}U 4.19	8.30E-05	2.80E-02	2.42E-04	1.71E-02	2.64E-04	4.50E-02
^{235}U 4.40	7.10E-04	8.00E-02	1.97E-03	1.48E-01	2.11E-03	3.60E-01
^{232}Th 4.00	2.20E-05	--	5.68E-05	3.57E-03	--	--

a. DOE 2000, Salmon and Hermann 1992, Auguston and Reilly 1974
-- No data

Table 4-2 Neutron yields (n/s-g) for ^{234}U with trace Impurities[a].

Target Element	DOE Standard Table 6-5	DOE Standard Table 6-6	Salmon & Hermann Table 3 Interpolation	Auguston & Reilly Tables 7.1 & 7.2
Fluoride	5.80E+02	7.17E+02	7.81E+02	1.56E+03
Oxide	3.00E+00	9.25E+00	8.26E+00	9.15E+00

Target Element	DOE Standard Table 6-5	DOE Standard Table 6-6	Salmon & Hermann Table 3 Interpolation	Auguston & Reilly Tables 7.1 & 7.2
Sodium	--	1.16E+02	1.29E+02	1.98E+02

a. DOE 2000, Salmon and Hermann 1992, Auguston and Reilly 1974
-- No Data

Tables 4-3 through 4-6 list neutron yields for fluorine, oxygen, and sodium targets for sources of ^{238}U, ^{235}U, ^{232}Th, ^{230}Th, and associated progeny. Soda salt ($Na_2U_2O_7$) yields have been calculated for uranium isotopes on an atom percent weighted basis (22.22% Na and 77.77% O). Target yields have been derived based on the data and methodology presented in Salmon and Hermann (1992).

The neutron yields for fluoride, sodium, oxygen, and soda salt listed in Tables 4-2 through 4-5 were used to derive a neutron multiplication factor. This factor accounts for the production of neutrons from alpha-emitting progeny in the ^{235}U, ^{238}U and ^{232}Th decay chains. For example, the neutron multiplication factor for the ^{238}U decay chain has been calculated by dividing the sum of the alpha abundance weighted neutron yields through ^{226}Ra by the yield for ^{238}U plus ^{234}U. The neutron multiplication factors for the ^{235}U and ^{232}Th chains were calculated in the same manner. The resulting neutron multiplication factors are listed in Tables 4-7 and 4-8 below.

To account for the radioactivity distribution of isotopes in natural uranium, activity-weighted multiplication factors (see Tables 4-3 and 4-4) have been calculated as the product of the neutron multiplication factor and the fraction of alpha-emitting activity in each chain. For natural uranium in equilibrium through ^{226}Ra and ^{223}Ra, the ^{238}U chain accounts for 95.1% of the alpha-emitting activity while ^{235}U accounts for 4.85% of the alpha-emitting activity. For natural uranium in equilibrium through ^{210}Po and ^{211}Po, the ^{238}U chain accounts for 97% of the alpha-emitting activity while ^{235}U accounts for 3% of the alpha-emitting activity.

Table 4-3. Neutron yields for ^{238}U chain isotopes.[a]

Isotope	Average alpha energy (MeV)	Alpha emission % abundance	Alpha abundance weighted O yield (n/s-Ci)	Alpha abundance weighted Na yield (n/s-Ci)	Alpha abundance weighted soda salt yield (n/s-Ci)	Alpha abundance weighted F yield (n/s-Ci)
U-238	4.194	1.00E+02	7.19E+02	6.56E+03	2.02E+03	5.09E+04
U-234	4.773	1.00E+02	1.38E+03	2.07E+04	5.68E+03	1.25E+05
Th-230	4.665	1.00E+02	1.24E+03	1.71E+04	4.77E+03	1.07E+05
Ra-226	4.774	1.00E+02	1.38E+03	2.07E+04	5.69E+03	1.25E+05
Rn-222	5.489	1.00E+02	2.48E+03	5.49E+04	1.41E+04	2.84E+05
Po-218	6.001	1.00E+02	3.35E+03	9.43E+04	2.35E+04	4.54E+05
At-218	6.693	1.00E+02	5.00E+03	1.62E+03	4.25E+03	7.63E+03
Bi-214	5.461	2.10E-02	5.11E-01	1.12E+01	2.89E+00	5.82E+01
Po-214	7.686	1.00E+02	7.86E+03	2.59E+05	6.36E+04	1.35E+06
Pb-210	3.720	1.90E-06	6.31E-06	3.62E-05	1.29E-05	3.94E-04
Bi-210	4.625	1.30E-04	1.55E-03	2.05E-02	5.76E-03	1.31E-01
Po-210	5.304	1.00E+02	2.17E+03	4.48E+04	1.17E+04	2.38E+05
Total U-238 to Ra-226			4.72E+03	6.51E+04	1.81E+04	4.08E+05
Activity-weighted neutron multiplication factor U-238 to Ra-226			2.14E+00	2.27E+00	2.24E+00	2.21E+00
Total U-238 to Po-210			2.56E+04	5.19E+05	1.35E+05	2.74E+06
Activity-weighted neutron multiplication factor U-238 to Po-210			1.18E+01	1.85E+01	1.71E+01	1.51E+01

a. Adapted from Salmon and Hermann (1992).

Table 4-4. Neutron yields for ^{235}U chain isotopes.[a]

Isotope	Average alpha energy (MeV)	Alpha emission % abundance	Alpha abundance weighted O yield (n/s-Ci)	Alpha abundance weighted Na yield (n/s-Ci)	Alpha abundance weighted soda salt yield (n/s-Ci)	Alpha abundance weighted F yield (n/s-Ci)
U-235	4.378	1.00E+02	9.10E+02	9.57E+03	2.83E+03	6.83E+04
Pa-231	4.923	1.00E+02	1.57E+03	3.20E+04	8.33E+03	1.50E+05
Ac-227	4.931	1.38E+00	2.18E+01	3.59E+02	9.67E+01	2.09E+03
Th-227	5.902	1.00E+02	3.18E+03	8.66E+04	2.17E+04	4.21E+05
Ra-223	5.697	1.00E+02	2.83E+03	7.07E+04	1.79E+04	3.52E+05
Rn-219	6.812	1.00E+02	5.34E+03	1.73E+05	4.27E+04	8.23E+05
Po-215	7.386	1.00E+02	7.00E+03	2.29E+05	5.64E+04	1.15E+06
At-215	8.026	1.00E+02	8.86E+03	2.92E+05	7.17E+04	1.58E+06
Bi-211	6.550	1.00E+02	4.58E+03	1.48E+05	3.64E+04	6.90E+05
Po-211	7.420	9.89E+01	7.02E+03	2.30E+05	5.66E+04	1.16E+06
Total U-235 to Ra-223			8.51E+03	1.99E+05	5.09E+04	9.94E+05
Alpha activity-weighted neutron multiplication factor U-235 to Ra-223			4.54E-01	1.01E+00	8.71E-01	7.05E-01
Total U-235 to Po-211			4.13E+04	1.27E+06	3.15E+05	6.39E+06
Alpha activity-weighted neutron multiplication factor U-235 to Po-211			1.36E+00	3.99E+00	3.33E+00	2.81E+00

a. Adapted from Salmon and Hermann (1992).

Table 4-5. Neutron yields for ^{232}Th chain isotopes.[a]

Isotope	Average alpha energy (MeV)	Alpha emission % abundance	Alpha abundance weighted O yield (n/s-Ci)	Alpha abundance weighted Na yield (n/s-Ci)	Alpha abundance weighted soda salt yield (n/s-Ci)	Alpha abundance weighted F yield (n/s-Ci)
Th-232	4.000	1.00E+02	5.18E+02	3.39E+03	1.16E+03	3.25E+04
Th-228	5.399	1.00E+02	2.33E+03	5.00E+04	1.29E+04	2.62E+05
Ra-224	5.675	1.00E+02	2.79E+03	6.90E+04	1.75E+04	3.45E+05
Rn-220	6.288	1.00E+02	3.98E+03	1.22E+05	3.03E+04	5.75E+05
Po-216	6.779	1.00E+02	5.25E+03	1.70E+05	4.19E+04	8.06E+05
Bi-212	6.051	3.59E+01	1.24E+03	5.31E+04	1.28E+04	2.48E+05
Po-212	8.784	1.00E+02	7.41E+03	2.34E+05	5.79E+04	1.34E+06
Total Th-232 to Ra-224			5.64E+03	N/A	N/A	6.39E+05
Neutron multiplication factor Th-232 to Ra-224			1.09E+01	N/A	N/A	1.97E+01
Total Th-232 to Po-212			2.35E+04			3.61E+06
Neutron multiplication factor Th-232 to Po-212			4.54E+01			1.11E+02

a. Adapted from Salmon and Hermann (1992).

Table 4-6. Neutron yields for ^{230}Th.[a]

Isotope	Average alpha energy (MeV)	Alpha emission % abundance	Alpha abundance weighted O yield (n/s-Ci)	Alpha abundance weighted Na yield (n/s-Ci)	Alpha abundance weighted soda salt yield (n/s-Ci)	Alpha abundance weighted F yield (n/s-Ci)
Th-230	4.665	1.00E+02	1.24E+03	1.71E+04	4.77E+03	1.07E+05
Neutron multiplication factor			1.00E+00	N/A	N/A	1.00E+00

a. Adapted from Salmon and Hermann (1992).

Table 4-7. Multiplication factors for decay chains in equilibrium with ^{223}Ra, ^{226}Ra, and ^{224}Ra.

Low Z element	U-235	U-238	Th-232
Oxygen	9.35	2.25	10.9
Soda salt (Na_2O_7)	18.0	2.36	N/A
Fluorine	14.5	2.32	19.7

Table 4-8. Multiplication factors for decay chains in equilibrium with ^{211}Po, ^{210}Po, and ^{212}Po.

Low Z element	U-235	U-238	Th-232
Oxygen	45.1	12.2	45.4
Soda salt (Na_2O_7)	111.0	17.6	N/A
Fluorine	93.6	15.6	111.0

5.0 URANIUM COMPOUND DOSE RATES

Following the chemical separation of uranium from other elements, naturally occurring uranium consists of ^{238}U, ^{235}U, and ^{234}U. Table 5-1 summarizes natural uranium properties and isotopic composition. The neutron flux from a gram of natural uranium (0.99284 g ^{238}U, 0.000053 g ^{234}U, and 0.00711 g ^{235}U) following chemical separation is calculated as the sum of the products of each isotope fraction per gram, isotope specific activity, and isotope neutron yield. The neutron flux for each chemical compound of interest [UO_2, UO_3, U_3O_8, soda salt ($Na_2U_2O_7$), UF_4, and UF_6] has been calculated. The neutron flux at 1 and 3 ft. was calculated as the quotient of the neutron production rate per gram of natural uranium and the surface area of a sphere with a radius of 1 and 3 ft (surface areas of 1.167×10^4 cm^2 and 1.051×10^5 cm^2, respectively). The neutron flux was converted to dose rate using a conversion factor of 1.3×10^{-4} rem/hr per neutron/cm^2-s. The neutron dose rates at 1 and 3 ft from 1 gram of natural uranium with no alpha-emitting progeny present are listed in Table 5-2 below.

Table 5-1. Natural uranium isotopes and properties.

Property	U-234	U-235	U-238
Weight percent	5.30E-03	7.11E-01	9.9284E+01
Activity percent	4.89E+01	2.2E+00	4.89E+01
Half-life (Y)	2.45E+05	7.04E+8	4.47E+09
Specific activity (Ci/g)	6.25E-03	2.16E-6	3.36E-07

Table 5-2. Natural uranium-per-gram dose rates without alpha-emitting progeny.[a]

	Dose rate (no progeny) rem/hr-gram	
Chemical form	1 ft	3 ft
UO_2/UO_3/ U_3O_8 (ore)	7.91E-12	8.79E-13
$Na_2U_2O_7$ (soda salt)	2.89E-11	3.21E-12
UF_4/UF_6	6.62E-10	7.36E-11

a. Uranium isotopes following chemical separation

According to DOE (2000), neutrons of approximately 2 MeV energy are generated by the interaction of alpha particles from uranium with the nuclei of fluoride and other low atomic number atoms. The magnitude of neutron flux will vary based on the total activity of uranium (which is a function of enrichment) and the chemical compound. In the case of UF_6, the typically measured neutron dose rates for large cooled storage cylinders are as follows:

Natural-5% enrichment: 0.01-0.2 mrem/h (no distance specified in DOE 2000)

Very high enrichment (97+%): 2-4 mrem/h (contact) and 1-2 mrem/h (3 ft)

Assuming that a “large” storage container is 100 pounds and assuming that no progeny are present, the Microsoft® Excel spreadsheet, Alpha-NforTIB12-01-04Draft2.xls, created to perform the calculations in this TIB yields the following results:

Natural-5% enrichment: 0.03-0.14 mrem/h (1 ft) and 0.003-0.016 mrem/h (3 ft)

Very high enrichment (97+%): 4.0 mrem/h (1 ft) and 0.45 mrem/h (3 ft)

The neutron dose rates calculated by the Microsoft® Excel spreadsheet for the case of natural-5% enrichment are in the same range as those in DOE 2000. For the very HEU case, neutron dose rates calculated by the Microsoft® Excel spreadsheet are underestimated by a factor of 2 – 4 at 3 ft due to the point source geometry assumed.

For calculation of the neutron dose rates from uranium with progeny present, an effective neutron multiplication factor for natural uranium in secular equilibrium with radium and polonium is calculated for each compound as the sum of the activity-weighted multiplication factors (see Tables 4-3 and 4-4) for ^{238}U and ^{235}U. The effective neutron multiplication factors are presented in Tables 5-3 and 5-4 below. When alpha-emitting progeny are present in natural uranium, the effective neutron multiplication factors are multiplied by the neutron dose rate resulting from natural uranium (^{238}U, ^{234}U, ^{235}U) when no other alpha-emitting progeny are present.

Table 5-3. Effective neutron multiplication factors for natural uranium in equilibrium with radium.

Low Z element	Natural uranium
Oxygen	2.59
Soda salt (Na_2O_7)	3.11
Fluorine	2.91

Table 5-4. Effective neutron multiplication factors for natural uranium in equilibrium with polonium.

Low Z element	Natural uranium
Oxygen	13.2
Soda salt (Na_2O_7)	20.4
Fluorine	17.9

Table 5-5 lists neutron dose rates from naturally occurring uranium assuming the presence of alpha-emitting progeny in secular equilibrium through ^{226}Ra in the ^{238}U chain and through ^{223}Ra in the ^{235}U chain. Table 5-5 dose rates are calculated as the product of the dose rates listed in Table 5-2 and the effective neutron multiplication factors listed in Table 5-3. The presence or absence of progeny and their degree of equilibrium will vary depending on site-specific processes and practices. As a consequence, the presence of progeny and degree of equilibrium should be determined on a site-specific basis.

Table 5-5. Natural uranium-per-gram dose rates with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.

Chemical form	Dose rate rem/h-gram	
	1 ft	3 ft
$UO_2/UO_3/U_3O_8$ (ore)	2.05E-11	2.28E-12
$Na_2U_2O_7$ (soda salt)	9.00E-11	1.00E-11
UF_4/UF_6	1.93E-09	2.14E-10

Table 5-6 lists neutron dose rates from naturally occurring uranium assuming the presence of alpha-emitting progeny in secular equilibrium through ^{210}Po in the ^{238}U chain and through ^{211}Po in the ^{235}U chain. Table 5-6 dose rates are calculated as the product of the dose rates listed in Table 5-2 and the effective neutron multiplication factors listed in Table 5-4.

Table 5-6. Natural uranium-per-gram dose rates with alpha-emitting progeny in secular equilibrium with ^{210}Po in the ^{238}U chain and ^{211}Po in the ^{235}U chain

	Dose rate rem/h-gram	
Chemical form	**1 ft.**	**3 ft.**
$UO_2/UO_3/$ U_3O_8 (ore)	1.04E-10	1.16E-11
$Na_2U_2O_7$ (soda salt)	5.90E-10	6.55E-11
UF_4/ UF_6	1.19E-08	1.32E-09

6.0 THORIUM COMPOUND DOSE RATES

Following the chemical separation of thorium from other elements in ore, naturally occurring thorium consists of ^{232}Th and ^{228}Th in secular equilibrium. Within 25 days, ^{224}Ra is in secular equilibrium with the ^{232}Th and ^{228}Th; as a consequence, neutron dose rates have been calculated for natural thorium compounds assuming the presence of alpha-emitting progeny in secular equilibrium through ^{224}Ra. In addition, neutron dose rates from the thorium alpha-n reaction have been calculated assuming the presence of alpha-emitting progeny in secular equilibrium through ^{212}Po. As noted for uranium, the presence or absence of progeny and their degree of equilibrium will vary depending on site-specific processes and practices. As a consequence, the presence of progeny and degree of equilibrium should be determined on a site-specific basis.

Table 6-1. Natural thorium isotopes and properties.

Property	**Th-232**	**Th-228**
Weight percent	~100	1.34E-08
Activity percent	5.00E+01	5.00E+01
Half-life (Y)	1.41E+10	1.91E+00
Specific activity (Ci/g)	1.10E-07	8.20E+02

The neutron flux from a gram of natural thorium (~1.0 g ^{232}Th and 1.34×10^{-10} g ^{228}Th) is calculated as the product of the ^{232}Th mass fraction per gram, ^{232}Th specific activity, ^{232}Th neutron yield (in the compound of interest) and the neutron multiplication factor. The neutron multiplication factors for ^{232}Th in secular equilibrium through ^{224}Ra and through ^{212}Po are listed in Table 4-5. The neutron flux for each chemical compound of interest [$Th(NO_3)_4$ and ThF_4] has been calculated. The neutron flux at 1 and 3 ft. is calculated as the quotient of the neutron production rate per gram of natural thorium and the surface area of a sphere with a radius of 1 and 3 ft (surface areas of 1.167×10^4 cm^2 and 1.051×10^5 cm^2, respectively). The neutron flux was converted to dose rate using a conversion factor of 1.3×10^{-3} rem/hr per neutron/cm^2-s. Neutron dose rates at 1 and 3 ft from 1 g of natural thorium in secular equilibrium through ^{224}Ra and through ^{212}Po are listed in Tables 6-2 and 6-3, respectively.

Table 6-2. Natural thorium per gram dose rates (secular equilibrium through ^{224}Ra).

Chemical form	Dose rate rem/hr-gram	
	1 ft	3 ft
$Th(NO_3)_4$	6.89E-12	7.65E-13
ThF_4	7.81E-10	8.68E-11

Table 6-3. Natural thorium per gram dose rates (secular equilibrium through ^{212}Po).

Chemical form	Dose rate rem/hr-gram	
	1 ft	3 ft
$Th(NO_3)_4$	2.87E-11	3.19E-12
ThF_4	4.41E-09	4.90E-10

Thorium bearing ores also contain varying amounts of uranium, and the thorium decay progeny of the naturally occurring isotopes of uranium would also be present in chemically separated thorium. The most important of the thorium isotopes and decay progeny in chemically separated thorium compounds are ^{232}Th, ^{228}Th, ^{224}Ra, ^{230}Th, and ^{227}Th. The half-life of ^{227}Th is only 18.7 days, and its ^{223}Ra progeny has a half-life of only 11.4 days. Following purification of the thorium from ore, the ^{227}Th and ^{223}Ra contribution to neutron dose will decrease rapidly. In addition, the activity ratio of ^{230}Th to ^{227}Th would probably follow the ratio of ^{238}U to ^{235}U in natural uranium (22.2:1.0 on an activity basis). As a consequence, even after initial purification of thorium, the neutron flux component from ^{227}Th would be negligible in comparison to the ^{230}Th neutron flux contribution. As a result, ^{227}Th and ^{223}Ra flux contributions have been neglected.

It is reasonable and conservative to assume secular equilibrium between ^{232}Th, ^{228}Th, and ^{224}Ra in calculating the alpha-neutron dose rate, but the activity of ^{226}Ra is not in secular equilibrium with ^{230}Th until approximately 7,000 years. As a consequence, the contribution to neutron flux from ^{226}Ra can be neglected unless site-specific data or processes indicate that ^{226}Ra would be present.

The neutron flux for a gram mixture containing thorium isotopes (^{232}Th in equilibrium with ^{228}Th and ^{224}Ra and ^{230}Th with no progeny) is calculated as the sum of the ^{232}Th neutron flux contribution discussed above and the ^{230}Th contribution. The mass fraction of ^{232}Th and ^{230}Th will need to be determined on a site-specific basis. This analysis is based on the thorium isotope mixture identified at the Mallinckrodt site, which contained 88.4% ^{232}Th and 11.6% ^{230}Th by weight. The neutron flux is calculated as the sum of the products of the ^{232}Th mass fraction per gram, ^{232}Th specific activity, ^{232}Th neutron yield, and ^{232}Th neutron multiplication factor and the product of the ^{230}Th mass fraction, ^{230}Th specific activity, and ^{230}Th neutron yield.

The neutron flux at 1 and 3 ft. was calculated as the quotient of the neutron production rate per gram of natural uranium and the surface area of a sphere with a radius of 1 and 3 ft (surface areas of 1.167×10^4 cm^2 and 1.051×10^5 cm^2, respectively). The neutron flux was converted to dose rate using a conversion factor of 1.3×10^{-3} rem/hr per neutron/cm^2-s. Neutron dose rates at 1 and 3 ft from 1 g of thorium isotope mix containing ^{232}Th in secular equilibrium through ^{224}Ra and through ^{212}Po are listed in Tables 6-4 and 6-5, respectively.

Table 6-4. Thorium isotope mix per gram dose rates (^{232}Th in secular equilibrium through ^{224}Ra).

Chemical form	Dose rate rem/hr-gram	
	1 ft	3 ft
$Th(NO_3)_4$	3.24E-08	3.60E-09
ThF_4	2.80E-06	3.11E-07

Table 6-5. Thorium isotope mix per gram dose rates (^{232}Th in secular equilibrium through ^{212}Po).

Chemical form	Dose rate rem/hr-gram	
	1 ft	3 ft
$Th(NO_3)_4$	3.25E-08	3.61E-09
ThF_4	2.80E-06	3.11E-07

The analysis of neutron production by the alpha-neutron reaction identified the forms of uranium and thorium that would produce neutrons at the highest rates as UF_4, UF_6, and ThF_4. In addition, the analysis considered the uranium oxide forms and soda salt ($Na_2U_2O_7$) as well as thorium nitrate [$Th(NO_3)_4$]. Little information could be found about UCl_4, so the dose from UCl_4 was bounded by identifying another form (UF_4), which results in a larger dose than the UCl_4 form.

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