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Estimation of Neutron Flux and Neutron Dose Rates from Alpha-Neutron Reactions in Uranium and Thorium Compounds	ORAUT-OTIB-0024	Rev. 01
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PUBLICATION RECORD

EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
04/07/2005	00	New technical information bulletin to provide estimation of neutron doses from uranium and thorium compounds that contain low atomic number components. Incorporates formal internal and NIOSH review comments. Initiated by Cindy W. Bloom.
10/08/2020	01	Revision initiated to update neutron flux from uranium and thorium compounds that contain low atomic number components using Monte Carlo calculation methods. Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by Matthew H. Smith.

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ACRONYMS AND ABBREVIATIONS

Ci	curie
cm	centimeter
DOE	U.S. Department of Energy
ENDF	Energy Nuclear Data Format
ft	feet
g	gram
hr	hour
MCNP	Monte Carlo N-Particle
MeV	megaelectron-volt (1 million electron-volts)
n	neutron
NIOSH	National Institute for Occupational Safety and Health
NIST	National Institute for Standards and Technology
ORAU	Oak Ridge Associated Universities
s	second
SRDB Ref ID	Site Research Database Reference Identification (number)
TBD	technical basis document
TENDL	TALYS Evaluated Nuclear Data Library
TIB	technical information bulletin
U.S.C.	United States Code
Z	atomic number
α	alpha particle
§	section or sections

1.0 INTRODUCTION

Technical information bulletins (TIBs) are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historical background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). TIBs may be used to assist NIOSH staff in the completion of individual dose reconstructions.

In this document, the word “facility” is used as a general term for an area, building, or group of buildings that served a specific purpose at a site. It does not necessarily connote an “atomic weapons employer facility” or a “Department of Energy (DOE) facility” as defined in the Energy Employees Occupational Illness Compensation Program Act of 2000 [42 U.S.C. § 7384l(5) and (12)].

1.1 PURPOSE

Based on a review of Revision 00 of this document – specifically its use of older experimental data – the Oak Ridge Associated Universities (ORAU) Team conducted a study to examine the feasibility of using modern computer codes such as the MCNP (Monte Carlo N-Particle) 6.2 transport code – in conjunction with the TENDL (TALYS Evaluated Nuclear Data Library) data libraries, the Sources 4C code, and the modified Sources 4C-m code – to calculate accurate values for the magnitude of the neutron flux from the following compounds: UO_2, UO_3, U_3O_8, $Na_2U_2O_7$, UF_4, UF_6, ThF_4, and $Th(NO_3)_4$. TENDL is a cross-section library that was produced with results from the TALYS code. It is used by MCNP to simulate particle interactions. TALYS is a software package that simulates nuclear reactions. In addition, neutron dose rate values for these compounds were calculated for use in project technical basis documents (TBD) for sites that processed the compounds listed above. In most cases, the data presented here will be used by site TBD authors and subject matter experts to derive annual neutron dose from work with these compounds to assign to energy employees who may not have been monitored for neutron radiation. Examples of these sites include: Mallinckrodt Chemical Company, Linde Ceramics Plant, Nuclear Materials and Equipment Corporation, Battelle Laboratories, and United Nuclear Corporation in Hematite, Missouri.

1.2 SCOPE

Sources 4C and Sources 4C-m calculate neutron magnitude and spectra from (α,n) interactions, spontaneous fission, and delayed neutrons. The (α,n) spectra from these outputs as the source in MCNP are used. With these sources, MCNP was used to simulate the transport of the neutrons and to calculate the fluence both within the material, at 1 ft, and 3 ft.

TENDL is a cross-section library. MCNP uses cross-section libraries to model the transport of particles through materials. Unlike cross-section libraries provided with the MCNP 6.2 distribution, TENDL contains (α,n) cross-sections. Therefore, if proven to be accurate, TENDL would enable MCNP to model the (α,n) reaction, eliminating the need for Sources 4C and 4C-m.

Volume-averaged neutron flux was calculated for the first seven compounds of interest using four methods:

1. Calculations based on data from Revision 00 of this document (ORAUT 2005);
2. Combined application of Sources 4C and MCNP 6.2;
3. Combined application of Sources 4C-m and MCNP 6.2; and
4. Application of MCNP 6.2 with TENDL (α,n) cross-section libraries.

Volume-averaged neutron flux for the eighth compound, $Th(NO_3)_4$, was calculated with the first and last method only due to software limitations.

ORAUT (2020a) provides the Sources 4C and 4C-m input files. ORAUT (2020b) and ORAUT (2020c) contain the MCNP 6.2 input files based on Sources 4C and 4C-m neutron production rates, respectively. ORAUT (2020d) provides the MCNP input files based on TENDL data libraries, and ORAUT (2020e) discusses the TENDL data.

Results from each method were compared.

Neutron dose rate calculations (at distances of 1 ft and 3 ft) were then derived from the neutron flux information – specifically from Sources 4C and 4C-m results. The 1 ft and 3 ft distances are reasonable bounding distances for activities performed by energy employees. Other distances and specific geometries can be modeled, if needed, using the methods discussed in this document.

2.0 NEUTRON FLUX CALCULATIONS

2.1 CALCULATION OF NEUTRON PRODUCTION RATES WITH SOURCES 4C

The Sources 4C code calculates the neutron production rates and neutron spectra from spontaneous (α,n) fission reactions and delayed neutron emission (Perry and Wilson 1981). The production rates and spectra can be calculated in four situations: a homogeneous mixture of alpha-emitting material and low atomic number (low-Z) target material, an interface between a region of alpha-emitting material and a region of target material, a monoenergetic alpha beam incident on a low-Z target, or a three-region interface in which a third region lies between a region of alpha-emitting material and a region of low-Z target material. In this study, Sources 4C was used to calculate neutron production rates for the compounds. These production rates were later applied in the development of source definitions in MCNP 6.2.

All compounds – as listed above in Section 1.1 – were modeled as solid, homogeneous mixtures. The atomic fraction of each element was determined from the chemical formula of the compound. Natural abundance data for each isotope were taken from "Atomic Weights and Isotopic Compositions with Relative Atomic Masses" from the National Institute for Standards and Technology (NIST) Physical Measurement Laboratory (Coursey et al. 2020). The atomic fraction of each target isotope was determined by multiplying the natural abundance of the isotope by the atomic fraction of its element in the compound. For all compounds, the atomic density N_i of each alpha emitter was determined as:

$$N_i = \frac{\gamma_i \rho N_A}{M} \qquad (2\text{-}1)$$

where

γ_i	=	abundance of isotope *i*
ρ	=	physical density of the compound (g/cm^3)
N_A	=	Avogadro's number
M	=	molecular weight of the compound (g)

The isotopic abundances were also taken from Coursey et al. (2020). The physical densities for UO_2, U_3O_8, UF_4, and UF_6 were taken from *Characteristics of Uranium and its Compounds* (DOE 2001). The physical density of UO_3 was taken from *Cameco UO_3 Materials Analysis* (Hill et al. 2012). The physical density for $Na_2U_2O_7$ was taken as the average of the values in Table A.28 of *Stimulant Basis for the Standard High Solids Vessel Design* (Peterson et al. 2017). The physical density for ThF_4 was taken from WolframAlpha (2020) to maintain consistency with Revision 00. The density for $Th(NO_3)_4$ was taken as the average from Table 12 of *Analytical Characterization of the Thorium Nitrate Stockpile* (Mattus, Hermes, and Terry 2003).

The molecular weights for $Na_2U_2O_7$ and $Th(NO_3)_4$ were calculated from standard atomic weights in Coursey et al. (2020). The molecular weight for all other compounds was taken from the National Institute of Standards and Technology "Chemistry WebBook" (DOC 2020).

Sources 4C presents several limitations. The primary limitation is a hardcoded 6.5-MeV alpha energy threshold for oxygen and a 6.0-MeV threshold for fluorine. This threshold disables the evaluation of all compounds through the full decay chain. Therefore, the Sources 4C method was only applied to uranium compounds without progeny, uranium oxides assumed to be in equilibrium through ^{223}Ra, and thorium tetrafluoride assumed to be in equilibrium through ^{224}Ra. Neutron production rates from $Th(NO_3)_4$ could not be obtained with Sources 4C due to a lack of nuclide-level branching data for ^{14}N.

2.2 CALCULATION OF NEUTRON PRODUCTION RATES WITH SOURCES 4C-m

Sources 4C-m is a modified version of Sources 4C, which enables the evaluation of neutron production from (α,n) reactions over an extended alpha energy range (Montague 2018) by removing the hardcoded energy limit for the (α,2n) energy threshold. Neutron production rates were extended to these higher energies with the application of 2005 TENDL cross-section data. Sources 4C-m was then used to calculate neutron production rates for the compounds. These production rates were later applied in the development of source definitions in MCNP 6.2.

Neutron production rates were determined with Sources 4C-m for all uranium compounds without progeny, all uranium compounds assumed to be in equilibrium through ^{223}Ra, and all uranium compounds assumed to be in equilibrium through the entire decay chain.

Specifically, there were three cases considered for all uranium compounds:

1. No alpha-emitting progeny present,

2. Alpha-emitting progeny are assumed to be present and in secular equilibrium through ^{226}Ra in the ^{238}U chain and through ^{223}Ra in the ^{235}U chain, and

3. Alpha-emitting progeny are assumed to be present and in secular equilibrium through ^{210}Po in the ^{238}U chain and through ^{211}Po in the ^{235}U chain.

The first case was examined with all three modeling techniques. Based on results and resource requirements, Sources 4C and Sources 4C-m were chosen for examination of cases 2 and 3. Case 3 was only examined with Sources 4C-m because the emission energies exceed the capabilities of Sources 4C.

Neutron production rates were determined with Sources 4C-m for thorium tetrafluoride assumed to be in equilibrium through ^{224}Ra, and for thorium tetrafluoride assumed to be in equilibrium through the entire decay chain. Neutron production rates from $Th(NO_3)_4$ could not be obtained with Sources 4C-m due to a lack of nuclide-level branching data for ^{14}N.

Sources 4C has a pedigree that Sources 4C-m lacks. The original code has been validated against measured spectra and the development of the code has been published in reviewed journals. To the best of my knowledge, Sources 4C-m has only been validated against Sources 4C and has not been published in a reviewed journal.

2.3 MCNP 6.2 MODELING

MCNP 6.2 is a commonly used radiation transport code with a wide range of applications (Werner et al. 2013). In combination with Sources 4C and Sources 4C-m, MCNP 6.2 was used to simulate the volume-averaged neutron flux from each compound. Sources 4C and Sources 4C-m do not account for neutron interactions with matter, only production rates. This study used MCNP 6.2 to account for self-attenuation.

Each compound was modeled as a solid sphere with a radius of 1 cm ($V = 4.18879\ cm^3$) in a vacuum. All materials were defined using atomic fractions determined by multiplying the natural abundance of the isotope by the atomic fraction of its element in the compound. A neutron source was homogenously distributed throughout the sphere. The energy spectrum of the source was determined by the (α,n) neutron production rates calculated with Sources 4C and Sources 4C-m. The volume-averaged neutron flux was tallied over a volume of the sphere in which charged particle equilibrium exists ($r = 0.99$ cm; $V = 4.06438\ cm^3$). This constraint was chosen for comparison purposes with Sources 4C (and 4C-m), which assume the material is infinite.

All materials were defined using atomic fractions determined by multiplying the natural abundance of the isotope by the atomic fraction of its element in the compound. All neutron cross-section data were pulled from the ENDF71x cross-section library (continuous energy neutron cross-section data tables published by Los Alamos National Laboratory) (Conlin et al. 2013).

MCNP 6.2 Modeling Using TENDL Cross-Section Libraries
MCNP 6.2 was also used to determine the volume-averaged neutron flux without the aid of Sources 4C or Sources 4C-m. The same geometry and material definitions were used as in previous cases, but the source was defined as an alpha emitter with an energy probability distribution corresponding to the decay of alpha emitters in the material. For each isotope, alpha emission energy and intensity data were taken from Rad Toolbox (Eckerman and Sjoreen 2013). The probability of each alpha energy was determined by multiplying the corresponding intensity by the relative activity of the parent isotope.

TENDL cross-section libraries were applied to model alpha interactions within the material (TENDL is a database of cross-section data that can be used by MCNP to model the interactions of particles in materials). The TENDL libraries are nuclear data libraries developed by the Paul Scherrer Institute and the International Atomic Energy Agency (previously developed by NRG Petten) using the T6 codes (Koning and Rochman 2012). For a more in-depth discussion, the reader is referred to ORAUT (2020e). Neutron interactions were determined by the ENDF71x cross-section library as in the previous section.

Low count rates lead to high statistical errors for the uranium oxides. Therefore, for each of these compounds, two files were run with altered material compositions: one in which all oxygen was assumed to be ^{17}O and one in which all oxygen was assumed to be ^{18}O. The total volume-averaged neutron flux was determined in post-processing as:

$$\Phi = f_{a,\text{O-17}}\Phi_{\text{O-17}} + f_{a,\text{O-18}}\Phi_{\text{O-18}} \tag{2-2}$$

where

$f_{a,i}$ = atomic fraction of isotope i
Φ_i = volume-averaged neutron flux of isotope *i*.

Calculation Based on Revision 00 (ORAUT 2005)
The volume-averaged neutron flux Φ was back-calculated from the dose rates and isotope properties in Tables 5-1, 5-2, 6-1, and 6-2 of Revision 00 as (ORAUT 2005):

$$\Phi = \rho_j \times \sum \left(\omega_i \times a_i \times Y_i \right) \tag{2-3}$$

where

- ρ_j = physical density of element j in the compound (g/cm^3)
- ω_i = mass fraction of isotope i
- a_i = specific activity of isotope i (Ci/g)
- Y_i = neutron yield of isotope i [1/(s-Ci)].

Flux was calculated for all uranium compounds covered in Revision 00 (ORAUT 2005) without progeny and all thorium compounds covered in Revision 00 were assumed to be in equilibrium through ^{224}Ra.

3.0 NEUTRON FLUX RESULTS

Magnitudes of the volume-averaged neutron flux for each compound and each method of calculation [calculations based on Revision 00 data (ORAUT 2005)], combined application of Sources 4C and MCNP 6.2, combined application of Sources 4C-m and MCNP 6.2, and application of MCNP 6.2 with TENDL (α,n) cross-section libraries are displayed in Table 3-1.

Table 3-1. Neutron flux results.

1 Compound	2 Revision 00 (n/s-cm^3)	3 Sources 4C and MCNP 6.2 (n/s-cm^3)	4 Sources 4C-m and MCNP 6.2 (n/s-cm^3)	5 MCNP 6.2 and TENDL (n/s-cm^3)	6 % difference between Columns 3 and 5
UO_2	6.87E-03	1.89E-03	1.91E-03	1.67E-03[a]	12.4[a]
UO_3	4.32E-03	1.53E-03	1.54E-03	1.67E-03[a]	8.8[a]
U_3O_8	5.00E-03	1.69E-03	1.66E-03	1.21E-03[a]	33.1[a]
$Na_2U_2O_7$	1.29E-02	5.01E-03	6.65E-03	9.08E-04[a]	138.6[a]
$Na_2U_2O_7$	1.29E-02	5.01E-03	6.65E-03	1.47E-03[b]	109.3[b]
UF_4	3.02E-01	1.55E-01	1.47E-01	1.43E-01[a]	8.1[a]
UF_6	1.88E-01	1.09E-01	1.09E-01	9.94E-02[a]	9.2[a]
ThF_4	3.24E-01	1.60E-01	1.57E-01	1.67E-01[a]	4.3[a]
$Th(NO_3)_4$	5.67E-04	Not applicable	Not applicable	2.32E-04[a]	Not applicable

a. Results produced with TENDL 2014 cross-section library.
b. Results produced with TENDL 2015 cross-section library.

Compounds evaluated in this study are listed in the first column. The volume-averaged neutron flux as calculated from the values in Revision 00 (ORAUT 2005) are listed in column 2. These values are consistently higher than those calculated with simulations. The third column lists the volume-averaged neutron flux as calculated with MCNP 6.2 using the neutron production rates from Sources 4C. Revision 00 results and Sources 4C results show disagreement with a percent difference ranging from 53% to 114%. Column 4 lists the volume-averaged neutron flux as calculated with MCNP 6.2 using the neutron production rates from Sources 4C-m. These values are in good agreement with those in the previous column with a maximum percent difference of 28%. The fifth column lists the volume-averaged neutron flux determined by MCNP 6.2 calculations alone using TENDL cross-section libraries.

The final column lists the percent difference between the column 3 flux values (as calculated with MCNP 6.2 using Sources 4C production rates) and the column 5 flux values (determined by MCNP

6.2 calculations using TENDL cross-section libraries). For the majority of the compounds (UO_2, UO_3, UF_4, UF_6, ThF_4), the values agree within statistical uncertainty. Sources 4C results typically show ±17% agreement with measured results (Perry and Wilson 1981). MCNP 6.2 results for these compounds were calculated with 10% relative error.

The MCNP 6.2 results for U_3O_8 were calculated to approximately 5% error due to statistical uncertainty. Although the values from U_3O_8 do not show agreement within statistical error, the absolute difference is only 4.8×10^{-4} n/s-cm^3.

$Na_2U_2O_7$ was the only compound that showed greater than a 100% difference between the flux as calculated with MCNP 6.2 and Sources 4C production rates and the flux determined by MCNP 6.2 calculations alone using TENDL libraries. Since 2008, an updated TENDL library has been released every year or two. At the time this work was completed, 9 libraries had been released (now 10). Unlike other isotopes contained in the compounds of interest, the ^{23}Na (α,n) cross-sections showed significant differences between the 2014 release and the 2015 release. Based on the cross-section data used in Sources 4C (from published sources) and the other TENDL releases, the increased ^{23}Na (α,n) cross-sections in the 2015 release were likely in error.

4.0 NEUTRON DOSE RATE CALCULATIONS

Neutron production rates and spectra from (α,n) interactions for UO_2, UO_3, U_3O_8, $Na_2U_2O_7$, UF_4, and UF_6 were determined with Sources 4C. The neutron spectra were used as the energy distribution of a neutron point source in MCNP simulations used to calculate an ambient dose rate equivalent. In these simulations, the neutron fluence per source neutron was tallied at 1 and 3 ft from the neutron point source in air. ICRP Publication 74 conversion coefficients were used to convert the neutron fluence into ambient dose rate equivalent. The ambient dose rate equivalent from 1 g of natural uranium or natural thorium was calculated as the product of the ambient dose rate equivalent per source neutron, the neutron production magnitude (as determined by Sources 4C), and the density of uranium or thorium in the compound (ICRP 1997).

4.1 URANIUM COMPOUNDS

Neutron dose rate values for the compounds discussed in this document were calculated and are shown in this Section (uranium compounds) and Section 4.2 (thorium compounds) for use in dose reconstruction project TBDs for sites that processed these compounds. In most cases, the data presented here will be used by site TBD authors and subject matter experts to derive annual neutron dose from work with these compounds to assign to energy employees who may not have been monitored for neutron radiation. Examples of these sites include (but are not limited to) Mallinckrodt Chemical Company, Linde Ceramics Plant, Nuclear Materials and Equipment Corporation, Battelle Laboratories, and United Nuclear Corporation in Hematite, Missouri.

Table 4-1 shows the ambient dose rate equivalents at 1 and 3 ft from 1 g of natural uranium when no alpha-emitting progeny are considered. Table 4-2 shows the ambient dose rate equivalents at 1 and 3 ft from 1 g of natural uranium when alpha-emitting progeny through ^{223}Ra and ^{226}Ra are assumed to be in secular equilibrium.

Table 4-1. Natural uranium per-gram ambient dose rate equivalents without alpha-emitting progeny (rem/hr-g).[a]

Chemical form	1 ft	3 ft
UO_2	3.22E-12	3.57E-13
UO_3	4.19E-12	4.65E-13
U_3O_8	3.90E-12	4.32E-13
$Na_2U_2O_7$	1.31E-11	1.45E-12
UF_4	4.45E-10	4.93E-11
UF_6	5.35E-10	5.94E-11

a. Source: Sources 4C results.

Table 4-2. Natural uranium per-gram ambient dose rate equivalents with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain (rem/hr-g).[a]

Chemical form	1 ft	3 ft
UO_2	7.64E-12	8.48E-13
UO_3	9.95E-12	1.10E-12
U_3O_8	9.25E-12	1.03E-12
$Na_2U_2O_7$	2.71E-11	3.01E-12

a. Source: Sources 4C results.

The (α,n) neutron production calculations of Sources 4C are limited to alpha energies below 6.5 MeV for oxygen targets and below 6 MeV for fluorine targets. Therefore, no alpha-emitting progeny were considered for UF_4 or UF_6 and the full chain was not considered for any compounds using Sources 4C. Sources 4C-m extends the (α,n) cross-section library enabling calculations for increased alpha energies. Therefore, the calculations were repeated using Sources 4C-m neutron production rates and spectra. Table 4-3 shows the ambient dose rate equivalents at 1 and 3 ft from 1 g of natural uranium when no alpha-emitting progeny are considered. Table 4-4 shows the resulting equivalents when alpha-emitting progeny through ^{223}Ra and ^{226}Ra are assumed to be in secular equilibrium. Table 4-5 shows the equivalents when alpha-emitting progeny through ^{210}Po and ^{211}Po are assumed to be in secular equilibrium.

Table 4-3. Natural uranium per-gram ambient dose rate equivalents without alpha-emitting progeny (rem/hr-g).[a]

Chemical form	1 ft	3 ft
UO_2	3.21E-12	3.56E-13
UO_3	4.18E-12	4.63E-13
U_3O_8	3.88E-12	4.31E-13
$Na_2U_2O_7$	1.70E-11	1.89E-12
UF_4	4.32E-10	4.79E-11
UF_6	5.19E-10	5.76E-11

a. Source: Sources 4C-m results.

4.2 THORIUM COMPOUNDS

As for the uranium compounds, the ambient dose rate equivalents were calculated for ThF_4 at 1 and 3 ft. Table 4-6 shows the equivalents from 1 g of natural thorium when alpha-emitting progeny through ^{224}Ra are assumed to be in secular equilibrium. Table 4-7 shows the equivalents when alpha-emitting progeny through ^{212}Po are assumed to be in secular equilibrium.

Table 4-4. Natural uranium per-gram ambient dose rate equivalents with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain (rem/hr-g).[a]

Chemical form	1 ft	3 ft
UO_2	7.61E-12	8.44E-13
UO_3	9.91E-12	1.10E-12
U_3O_8	9.22E-12	1.02E-12
$Na_2U_2O_7$	3.54E-11	3.93E-12
UF_4	1.10E-09	1.23E-10
UF_6	1.33E-09	1.47E-10

a. Source: Sources 4C-m results.

Table 4-5. Natural uranium per-gram ambient dose rate equivalents with alpha-emitting progeny in secular equilibrium with ^{210}Po in the ^{238}U chain and ^{211}Po in the ^{235}U chain (rem/hr-g).[a]

Chemical form	1 ft	3 ft
UO_2	3.84E-11	4.26E-12
UO_3	5.01E-11	5.56E-12
U_3O_8	4.65E-11	5.16E-12
$Na_2U_2O_7$	1.21E-09	1.34E-10
UF_4	9.44E-09	1.05E-09
UF_6	1.14E-08	1.26E-09

a. Source: Sources 4C-m results.

Table 4-6. Natural thorium per-gram ambient dose rate equivalents for ThF_4 in secular equilibrium through ^{224}Ra (rem/hr-g).

Method	1 ft	3 ft
Sources 4C	5.33E-10	5.91E-11
Sources 4C-m	5.21E-10	5.78E-11

Table 4-7. Natural thorium per-gram ambient dose rate equivalents for ThF_4 in secular equilibrium through ^{212}Po (rem/hr-g).

Method	1 ft	3 ft
Sources 4C-m	3.67E-09	4.07E-10

Dose rates from $Th(NO_3)_4$ were not calculated due to the lack of nuclide-level branching data for ^{14}N in the Sources 4C and Sources 4C-m codes.

Using the same method, the ambient dose rate equivalents at 1 and 3 ft were calculated for ThF_4 based on the thorium isotope mixture at Mallinckrodt. This mixture is included here based on the information in – and potential use with – Section 5.4.2 of ORAUT-TKBS-0005, *Basis for Development of an Exposure Matrix for the Mallinckrodt Chemical Company St. Louis Downtown Site and the St. Louis Airport Site, St. Louis, Missouri* (ORAUT 2010). This mixture consisted of 88.4% ^{232}Th and 11.6% ^{230}Th. Table 4-8 shows the ambient dose rate equivalents from 1 g of the thorium mixture when ^{232}Th alpha-emitting progeny through ^{224}Ra are assumed to be in secular equilibrium. Table 4-9 shows the equivalents when ^{232}Th alpha-emitting progeny through ^{212}Po are assumed to be in secular equilibrium.

Table 4-8. Thorium isotope mix per-gram ambient dose rate equivalents for ThF_4 in secular equilibrium through ^{224}Ra (rem/hr-g).

Method	1 ft	3 ft
Sources 4C	2.02E-06	2.24E-07
Sources 4C-m	1.89E-06	2.10E-07

Table 4-9. Thorium isotope mix per-gram ambient dose rate equivalents for ThF_4 in secular equilibrium through ^{212}Po (rem/hr-g).

Method	1 ft	3 ft
Sources 4C-m	1.90E-06	2.10E-07

5.0 NEUTRON DOSE ENERGY FRACTIONS

Neutron spectra data from Sources 4C and 4C-m were used to generate Tables 5-1 to 5-33 to assist with the entry of neutron dose information (based on the dose rate data provided in Section 4.0) into IREP. The energy bins in each table correlate to the available bins available in IREP. In addition, the ambient [(*H*10*)] dose conversion values (DCFs) given in OCAS-IG-001, *External Dose Implementation Guideline* [NIOSH 2007], should be used with the data presented here to properly enter organ dose into IREP.

Table 5-1. Neutron ambient dose fractions for UO_2 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	2.50E-06
10–100 keV	5.21E-04
0.1–2 MeV	3.95E-01
2–20 MeV	6.04E-01

a. Source: Sources 4C results.

Table 5-2. Neutron ambient dose fractions for UO_2 sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	3.31E-06
10–100 keV	6.71E-04
0.1–2 MeV	3.64E-01
2–20 MeV	6.35E-01

a. Source: Sources 4C results.

Table 5-3. Neutron ambient dose fractions for UO_3 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	2.51E-06
10–100 keV	5.22E-04
0.1–2 MeV	3.95E-01
2–20 MeV	6.05E-01

a. Source: Sources 4C results.

Table 5-4. Neutron ambient dose fractions for UO_3 sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	3.31E-06
10–100 keV	6.72E-04
0.1–2 MeV	3.64E-01
2–20 MeV	6.35E-01

a. Source: Sources 4C results.

Table 5-5. Neutron ambient dose fractions for U_3O_8 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	2.51E-06
10–100 keV	5.22E-04
0.1–2 MeV	3.95E-01
2–20 MeV	6.04E-01

a. Source: Sources 4C results.

Table 5-6. Neutron ambient dose fractions for U_3O_8 sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	3.31E-06
10–100 keV	6.72E-04
0.1–2 MeV	3.64E-01
2–20 MeV	6.35E-01

a. Source: Sources 4C results.

Table 5-7. Neutron ambient dose fractions for $Na_2U_2O_7$ sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	1.66E-05
10–100 keV	3.78E-03
0.1–2 MeV	8.12E-01
2–20 MeV	1.84E-01

a. Source: Sources 4C results.

Table 5-8. Neutron ambient dose fractions for $Na_2U_2O_7$ sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	1.30E-05
10–100 keV	2.86E-03
0.1–2 MeV	8.36E-01
2–20 MeV	1.61E-01

a. Source: Sources 4C results.

Table 5-9. Neutron ambient dose fractions for UF_4 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	2.77E-06
10–100 keV	6.44E-04
0.1–2 MeV	9.61E-01
2–20 MeV	3.79E-02

a. Source: Sources 4C results.

Table 5-10. Neutron ambient dose fractions for UF_6 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	2.77E-06
10–100 keV	6.42E-04
0.1–2 MeV	9.61E-01
2–20 MeV	3.80E-02

a. Source: Sources 4C results.

Table 5-11. Natural thorium neutron ambient dose fractions for ThF_4 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	4.16E-06
10–100 keV	1.08E-03
0.1–2 MeV	8.26E-01
2–20 MeV	1.73E-01

a. Source: Sources 4C results.

Table 5-12. Thorium isotope mix neutron ambient dose fractions for ThF_4 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	1.35E-06
10–100 keV	4.80E-04
0.1–2 MeV	9.60E-01
2–20 MeV	3.92E-02

a. Source: Sources 4C results.

Table 5-13. Neutron ambient dose fractions for UO_2 sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	3.21E-06
10–100 keV	6.50E-04
0.1–2 MeV	3.68E-01
2–20 MeV	6.32E-01

a. Source: Sources 4C-m results.

Table 5-14. Neutron ambient dose fractions for UO_2 sources with alpha-emitting progeny in secular equilibrium with ^{210}Po in the ^{238}U chain and ^{211}Po in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	2.09E-06
10–100 keV	4.19E-04
0.1–2 MeV	2.68E-01
2–20 MeV	7.31E-01

a. Source: Sources 4C-m results.

Table 5-15. Neutron ambient dose fractions for UO_3 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	2.47E-06
10–100 keV	5.02E-04
0.1–2 MeV	3.99E-01
2–20 MeV	6.01E-01

a. Source: Sources 4C-m results.

Table 5-16. Neutron ambient dose fractions for UO_3 sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	3.21E-06
10–100 keV	6.51E-04
0.1–2 MeV	3.67E-01
2–20 MeV	6.32E-01

a. Source: Sources 4C-m results.

Table 5-17. Neutron ambient dose fractions for UO_3 sources with alpha-emitting progeny in secular equilibrium with ^{210}Po in the ^{238}U chain and ^{211}Po in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	2.09E-06
10–100 keV	4.19E-04
0.1–2 MeV	2.68E-01
2–20 MeV	7.32E-01

a. Source: Sources 4C-m results.

Table 5-18. Neutron ambient dose fractions for U_3O_8 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	2.47E-06
10–100 keV	5.02E-04
0.1–2 MeV	3.99E-01
2–20 MeV	6.01E-01

a. Source: Sources 4C-m results.

Table 5-19. Neutron ambient dose fractions for U_3O_8 sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	3.21E-06
10–100 keV	6.50E-04
0.1–2 MeV	3.68E-01
2–20 MeV	6.32E-01

a. Source: Sources 4C-m results.

Table 5-20. Neutron ambient dose fractions for U_3O_8 sources with alpha-emitting progeny in secular equilibrium with ^{210}Po in the ^{238}U chain and ^{211}Po in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	2.09E-06
10–100 keV	4.19E-04
0.1–2 MeV	2.68E-01
2–20 MeV	7.32E-01

a. Source: Sources 4C-m results.

Table 5-21. Neutron ambient dose fractions for $Na_2U_2O_7$ sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	1.87E-05
10–100 keV	4.48E-03
0.1–2 MeV	8.56E-01
2–20 MeV	1.40E-01

a. Source: Sources 4C-m results.

Table 5-22. Neutron ambient dose fractions for $Na_2U_2O_7$ sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	1.43E-05
10–100 keV	3.35E-03
0.1–2 MeV	8.73E-01
2–20 MeV	1.23E-01

a. Source: Sources 4C-m results.

Table 5-23. Neutron ambient dose fractions for $Na_2U_2O_7$ sources with alpha-emitting progeny in secular equilibrium with ^{210}Po in the ^{238}U chain and ^{211}Po in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	4.24E-06
10–100 keV	9.67E-04
0.1–2 MeV	7.10E-01
2–20 MeV	2.89E-01

a. Source: Sources 4C-m results.

Table 5-24. Neutron ambient dose fractions for UF_4 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	3.34E-06
10–100 keV	8.23E-04
0.1–2 MeV	9.66E-01
2–20 MeV	3.31E-02

a. Source: Sources 4C-m results.

Table 5-25. Neutron ambient dose fractions for UF_4 sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	3.37E-06
10–100 keV	7.72E-04
0.1–2 MeV	9.48E-01
2–20 MeV	5.16E-02

a. Source: Sources 4C-m results.

Table 5-26. Neutron ambient dose fractions for UF_4 sources with alpha-emitting progeny in secular equilibrium with ^{210}Po in the ^{238}U chain and ^{211}Po in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	2.35E-06
10–100 keV	6.29E-04
0.1–2 MeV	6.96E-01
2–20 MeV	3.03E-01

a. Source: Sources 4C-m results.

Table 5-27. Neutron ambient dose fractions for UF_6 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	3.34E-06
10–100 keV	8.22E-04
0.1–2 MeV	9.66E-01
2–20 MeV	3.32E-02

a. Source: Sources 4C-m results.

Table 5-28. Neutron ambient dose fractions for UF_6 sources with alpha-emitting progeny in secular equilibrium with ^{226}Ra in the ^{238}U chain and ^{223}Ra in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	3.37E-06
10–100 keV	7.71E-04
0.1–2 MeV	9.48E-01
2–20 MeV	5.16E-02

a. Source: Sources 4C-m results.

Table 5-29. Neutron ambient dose fractions for UF_6 sources with alpha-emitting progeny in secular equilibrium with ^{210}Po in the ^{238}U chain and ^{211}Po in the ^{235}U chain.[a]

Neutron energy group	Dose fraction
<10 keV	2.34E-06
10–100 keV	6.28E-04
0.1–2 MeV	6.96E-01
2–20 MeV	3.03E-01

a. Source: Sources 4C-m results.

Table 5-30. Natural thorium neutron ambient dose fractions for ThF_4 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	4.37E-06
10–100 keV	1.19E-03
0.1–2 MeV	8.24E-01
2–20 MeV	1.74E-01

a. Source: Sources 4C-m results.

Table 5-31. Natural thorium neutron ambient dose fractions for ThF_4 sources with alpha-emitting progeny in secular equilibrium through ^{212}Po.[a]

Neutron energy group	Dose fraction
<10 keV	1.89E-06
10–100 keV	5.03E-04
0.1–2 MeV	5.83E-01
2–20 MeV	4.17E-01

a. Source: Sources 4C-m results.

Table 5-32. Thorium isotope mix neutron ambient dose fractions for ThF_4 sources without alpha-emitting progeny.[a]

Neutron energy group	Dose fraction
<10 keV	1.86E-06
10–100 keV	6.43E-04
0.1–2 MeV	9.65E-01
2–20 MeV	3.46E-02

a. Source: Sources 4C-m results.

Table 5-33. Thorium isotope mix neutron ambient dose fractions for ThF_4 sources with alpha-emitting progeny in secular equilibrium through ^{212}Po.[a]

Neutron energy group	Dose fraction
<10 keV	1.86E-06
10–100 keV	6.43E-04
0.1–2 MeV	9.64E-01
2–20 MeV	3.52E-02

a. Source: Sources 4C-m results.

6.0 CONCLUSIONS

Volume-averaged neutron flux was calculated for the compounds using four methods. Results show agreement for UO_2, UO_3, U_3O_8, UF_4, UF_6, ThF_4, and $Th(NO_3)_4$, which demonstrates the feasibility of using MCNP 6.2 with TENDL cross-section libraries to produce accurate neutron flux. However, results from the MCNP 6.2 with TENDL cross-section libraries method did not show agreement with the other methods for $Na_2U_2O_7$. Therefore, caution is indicated when applying this method to compounds containing sodium.

Values for neutron dose rate were also calculated (using input from Sources 4C and 4C-m). Dose rate values calculated using the methods presented here for UO_2, UF_4, and ThF_4 compounds differed from previous revision values by as much as 92%, 55%, and 40%, respectively. However, given the small magnitude of these values, the overall effect on calculations of energy employee annual dose based on these data is expected to be small. In addition, there were small overall decreases in the neutron flux values calculated for all compounds in comparison with the previous version of this document. Finally, the methods described here are based on the latest available modeling methods.

Information regarding the neutron energy dose fractions – for use with entering the dose data into IREP – have been updated from the previous revision and are provided based on the spectral data from Sources 4C and 4C-m.

Finally, if site-specific needs require the modeling techniques discussed here to address uranium compounds with different enrichment values, recycled uranium, or non-natural thorium compounds (e.g., triple separated), contact the ORAUT Principal Scientist for External Dosimetry for assistance.

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