PNNL-16148, Rev. 1



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# Dose Assessment for Current and Projected Uses of Source Material Under a U.S. NRC General License and Exemption Criteria

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## **Executive Summary**

Source material is used under specific license, general license, and various exemptions from licensing requirements in Title 10 of the Code of Federal Regulations (CFR) Part 40. Because the Atomic Energy Act of 1946 focused on source material primarily in terms of its significance to the production of special nuclear material, the initial regulation of source material did not, apparently, give much emphasis to public health and safety. This emphasis on the production of special nuclear material continued in later modifications to the regulations. Because the last major update of these regulations was made more than 40 years ago (i.e., in 1961), the U.S. Nuclear Regulatory Commission (NRC) is concerned that the current regulations for source materials may not be consistent with current health and safety regulations.

Naturally occurring uranium and thorium and their decay chains emit alpha, beta, and gamma radiation. Thorium dioxide is classified as a "known carcinogen" by the U.S. Agency for Toxic Substances and Disease Registry and has been linked to lung and liver diseases.<sup>1</sup> Uranium possesses toxic chemical properties that can damage kidney and other tissue when ingested or inhaled in large quantities.<sup>2</sup> Consequently, it is important for the NRC to understand how and in what quantities source material is being used under the general license and various exemptions and how it affects the health of the public. A document published in 2001 by the NRC partially addressed this concern, but it did not focus specifically on the general license uses of source material.<sup>3</sup> The information and analyses presented in this report specifically address current applications and trends in the use of generally licensed source material and provide estimates of dose to those who come in contact with this material.

Current industrial trends indicate a general decline in the use of thorium and uranium. Thorium was traditionally used in ceramics for refractory purposes but now is primarily used in scientific research, in a few electrical applications, as a chemical catalyst, and in optical lenses. Uranium, once popular in pigments and glazes, is now almost entirely limited to military applications that require special licensure and in scientific research. Because of high disposal costs, increasingly stringent regulations, and growing public concerns related to the natural radioactivity of source material, industry is rapidly developing substitutes and alternative processes that do not involve the use of uranium and thorium. For these reasons, the downward trend in the use of source material is expected to continue.

Researchers at Pacific Northwest National Laboratory identified how source materials meeting the exemption and general license requirements of 10 CFR 40.13(a), 40.13(b), and 40.22 are currently being used in industry and by the public. Based on the results of an extensive literature and Internet search, few "truly" new uses of source material were discovered that were not already considered in or represented by previous NRC analyses. Two exceptions were found: (1) the use of uranium and thorium as a thin-film optical coating and (2) the accumulation of uranium at water treatment plants that filter water to meet the U.S. Environmental Protection Agency drinking water standard for uranium of 30  $\mu$ g/L. Analyses of

<sup>&</sup>lt;sup>1</sup> U.S. Department of Health and Human Services, Agency for Toxic Substances and Disease Registry. 1999. "ToxFAQs<sup>TM</sup> for Thorium." [Electronic document accessed at http://www.atsdr.cdc.gov/tfacts147.html on August 8, 2006]

<sup>&</sup>lt;sup>2</sup> U.S. Department of Health and Human Services, Agency for Toxic Substances and Disease Registry. 1999. "ToxFAQs<sup>™</sup> for Uranium." [Electronic document accessed at http://www.atsdr.cdc.gov/tfacts150.html on August 9, 2006]

<sup>&</sup>lt;sup>3</sup> U.S. Nuclear Regulatory Commission. 2001. *Systematic Radiological Assessment of Exemptions for Source and Byproduct Materials*. NUREG-1717, Office of Nuclear Regulatory Research, Washington, D.C.

these scenarios indicate potential radiological exposures to the public above 100 mrem/yr in the following situations: (1) exposure of unprotected workers during manufacture of thin-film optical coatings and (2) filtration of uranium using methods that filter other radionuclides, including radium (e.g., mixed-ion beds, lime softening, reverse osmosis).

In addition, realistic, bounding scenarios were developed and evaluated for use of source material under 10 CFR 40.13(a), 40.13(b), and 40.22. These scenarios are intended to bound doses to members of the public from exposure to source material under "reasonable" conditions of use. Results suggest that reasonable scenarios exist for uses under 10 CFR 40.13(a), 40.13(b), and 40.22 that can potentially result in dose greater than 100 mrem/yr to unregulated workers or members of the public.

This report also provides a discussion of two dosimetric problems associated with thorium. First, unlike natural uranium, elemental thorium does not exist in a well-defined isotopic mixture, because <sup>230</sup>Th is produced from the primordial radioactive decay chain headed by <sup>238</sup>U, while <sup>232</sup>Th is the parent radionuclide of a separate primordial radioactive decay chain. The uranium and thorium series can be found in virtually any ratio in various ores, so the radionuclidic composition of elemental thorium can contain virtually any ratio of <sup>230</sup>Th to <sup>232</sup>Th. The presence of <sup>230</sup>Th is only significant for intakes of thorium, because it produces virtually no penetrating photon radiation. The second dosimetric problem is the unknown amount of ingrowth of the radioactive decay products of <sup>228</sup>Th and <sup>232</sup>Th. External dose rates may vary over a factor of about 2.5 depending on the degree of equilibrium.

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# Abbreviations/Acronyms

ATSDR	U.S. Agency for Toxic Substances and Disease Registry
AZoM	A to Z of Materials
Bq	becquerel(s)
C	Celsius
CDPH	Connecticut Department of Public Health
CERMET	ceramic metal
CFR	Code of Federal Regulations
cm <sup>3</sup>	cubic centimeter(s)
COMPENDEX <sup>®</sup>	COMPuterized ENgineering InDEX database
CRC	Chemical Rubber Company
DOE	U.S. Department of Energy
EHSO	Environmental Health and Safety Online
EPA	U.S. Environmental Protection Agency
eq	equivalent(s)
EUV	extreme ultra violet
FR	Federal Register
ft <sup>3</sup>	cubic feet
g	gram(s)
HPS	Health Physics Society
hr	hour(s)
Inspec <sup>®</sup>	database operated by the Institution of Engineering and Technology
ICRP	International Commission on Radiological Protection
ISCORS	Interagency Steering Committee on Radiation Standards
kg	kilogram(s)
L	liter(s)
LANL	Los Alamos National Laboratory
lb	pound(s)
m	meter(s)
MCL	maximum contaminant level
MEXT	Ministry of Education, Culture, Sports, Science and Technology
mm	millimeter(s)
mol	mole(s)
mrad	millirad(s)
mrem	millirem(s)
mSv	millisievert(s)

NDWC	National Drinking Water Clearinghouse
NHDES	New Hampshire Department of Environmental Services
nm	nanometer(s)
NORM	naturally occurring radioactive material
NRC	U.S. Nuclear Regulatory Commission
nSv	nanosievert(s)
NTIS	National Technical Information Service
OLED	organic light emitting diode
ORAU	Oak Ridge Associated Universities
ORNL	Oak Ridge National Laboratory
PLED	polymer light emitting diode
PNNL	Pacific Northwest National Laboratory
POTW	publicly-owned treatment works
ppm	parts per million
rem	rem(s)
Sv	sievert(s)
TENORM	technically enhanced naturally occurring radioactive material
TFHRC	Turner-Fairbank Highway Research Center
UIC	Uranium Information Center
μg/L	micrograms per liter
μrem	microrem(s)
U.S.	United States
USGS	U.S. Geological Survey
VOC	Volatile Organic Compound
WDOH	Washington State Department of Health
WHO	World Health Organization
wk	week(s)
WNA	World Nuclear Association
wt%	weight percent
yr	year(s)

# **1** Introduction

The objective of this review and assessment is to develop information needed to evaluate the adequacy of the regulations for possession and use of generally licensed and exempt source material addressed in Title 10 of the Code of Federal Regulations (CFR) Part 40. 10 CFR Part 40.4 defines source material as (1) uranium or thorium, or any combination thereof, in any physical or chemical form or (2) ores that contain by weight one-twentieth of one percent (0.05 percent) or more of uranium, thorium, or any combination thereof. Source material does not include special nuclear material (i.e., material artificially enriched). Because the last major update of these regulations was made more than 40 years ago, the U.S. Nuclear Regulatory Commission (NRC) is concerned that the current regulations for source materials may not be consistent with current health and safety regulations.

The focus of this project is on identifying new uses of source materials permitted under 10 CFR 40.13(a), 40.13(b), and 40.22 that had not already been evaluated in NUREG-1717 (NRC 2001). Exempt source materials identified in 10 CFR 40.13(c) were fully evaluated in NUREG-1717 (NRC 2001) and are outside the scope of this project, although any relevant information identified during the conduct of this project was collected and passed on to the NRC. Emphasis was therefore placed on discovery of new or projected uses not reported in NUREG-1717.

In this project, the primary concerns are the health and safety of the public, so data development efforts were focused on current, ongoing uses of source materials and the development of realistic exposure scenarios that potentially could have significant impact on individual dose. In addition, realistic, bounding scenarios were developed and evaluated for use of source material under 10 CFR 40.13(a), 40.13(b), and 40.22. These scenarios are intended to bound potential doses to members of the public from exposure to source material under "reasonable" conditions of use. Population or collective dose considerations are outside the scope of this project.

The report is organized as follows:

Chapter 2 provides a description of the methodology and approach used in the study to identify new uses of source materials and to evaluate their potential doses.

Chapter 3 describes the results of the literature search to identify new uses of source materials and overall trends in the use of source materials.

Chapter 4 provides an overview of the technical issues and limitations associated with performing radiological hazard assessments for thorium.

Chapter 5 describes the analysis and results for the dose assessment of new uses of source materials and bounding scenarios.

Chapter 6 summarizes the results of the study, draws conclusions about the results, and makes recommendations for follow-on work.

Chapter 7 lists references used in the report.

# 2 Approach to Review and Dose Assessment

The objectives of the review and assessment efforts performed by PNNL were fivefold:

- 1. Identify how source material meeting the exemption and general license requirements of 10 CFR 40.13(a), 40.13(b), and 40.22 is currently being used in industry and by the public.
- 2. Characterize the identified new uses of source material in terms of isotopic composition and inventory, material type and form, and how the material is routinely used.
- 3. Develop realistic radiological exposure scenarios for the identified new uses of source material for routine uses, reasonably possible non-routine uses, credible accident situations, and waste disposal possibilities.
- 4. Develop realistic individual dose estimates for the identified new uses of source materials permitted under 10 CFR 40.22.
- 5. Develop bounding individual dose estimates for source materials permitted under 10 CFR 40.13(a), 40.13(b), and 40.22.

These objectives were completed in three phases:

#### Phase I: Literature Search

In this phase, an extensive search was conducted to identify new uses and potential new uses of source materials in the United States. The required information was gathered by (1) evaluating NRC's existing information obtained from source material suppliers in 1986 and 2005, (2) searching the Internet to identify products currently being marketed and the suppliers of these products, (3) searching library and on-line database resources, (4) using existing knowledge and experience of the Pacific Northwest National Laboratory (PNNL) staff, and (5) querying PNNL staff contacts in trade societies, government, and industry to further identify source material products and suppliers. Resources for the literature and Internet search included commercial, government, and public internet sites, health-physics related user groups, the Hanford Technical Library, and scientific literature databases (e.g., COMPENDEX<sup>®</sup>, Inspec<sup>®</sup>, NTIS, PubMed, Science Citation Index Expanded). This search for new uses of source material used data previously collected by the NRC during industry surveys and phone contacts with knowledgeable individuals in relevant government organizations and professional societies.

#### Phase II: Scenario Development and Dose Analysis

After the identification of the potential uses, individual exposure scenarios were developed for each type of use or selected specific uses. The philosophy followed during the scenario development phase was to attempt to define realistic ways that people could interact with the radioactive materials or items. This approach was intended to define ways in which people might have extended-duration, relatively close-range exposures without being overly conservative. Parameters such as exposure time, exposure distance,

or inhalation rate were selected on the basis of daily exposure, usually in an occupational setting, at typical radionuclide concentrations. The approach is similar to that used in NUREG-1717, with more of a focus on the production and/or use stages of the product life cycle (because the quantities of radioactive material currently or projected to be in use are small and the potential for population dose from retail or waste management activities is also small). Calculations were made with appropriate computer codes to standardize the data for a large number of radionuclides or variants of exposure. Dose rates from sources external to the human body were estimated using tabulated factors taken from Federal Guidance Report No. 12 (Eckerman and Ryman 1993) or point-kernel integration computer codes such as MicroShield (Grove Engineering 2003). Radiation dose commitments from inhalation or ingestion were estimated using dose coefficients from the International Commission on Radiological Protection (ICRP) Publications 68 and 72 (ICRP 1994, 1996a). These dose coefficients are relatively recent guidance, and differ somewhat from the values used in NUREG-1717. Although the work plan for this project suggested that the computer code GENII (Napier et al. 1988; Napier 2006) might be used for some analyses, no scenarios of environmental dispersion and accumulation were evaluated to a level of detail that required this capability.

#### Phase III: Refine Scenarios and Dose Analysis

Develop and implement an assessment plan to acquire additional information to refine bounding dose scenarios and individual dose estimates from Phase II.

According to the Phase II results, the committed effective dose to unprotected workers during the manufacture of thorium thin-film optical coatings could approach about 0.8 rem, for the bounding case of a single worker processing up to 150 lb/yr of thorium with progeny. Additional information was needed on the design and operation of industrial sputtering processes to determine if the bounding exposure scenario is overly conservative. The NRC determined it necessary for PNNL to augment the source material information compiled in Phase I and refine the bounding dose scenarios and conservative individual dose estimates developed in Phase II related to the manufacture of thin-film optical coatings that utilize source material. PNNL first developed an assessment plan for collecting the additional information and refining the analyses. The assessment plan contained the following:

- identification of known gaps in the source material data collected in Phase I and where overly conservative assumptions had been made in the Phase II bounding dose scenarios due to the lack of information
- identification of 9 organizations to be contacted to obtain the additional source material information
- a questionnaire to be used to obtain the additional source material information
- identification of other sources of information to be pursued.

PNNL implemented the actions identified in the assessment plan after receiving NRC review and approval, including collecting additional source material information from the identified organizations using the questionnaire developed in the assessment plan. PNNL prepared this revised report (PNNL-16148, Rev. 1) to provide the additional source material information gathered during the manufacturer survey.

## **3** Trends in Usage of Source Material

An extensive literature and Internet search was conducted to identify new uses, and potential new uses, of source materials in the United States. This search for new uses used data previously collected by the NRC in industry surveys. It also included telephone contacts with knowledgeable individuals from government organizations and professional societies who have knowledge of how source materials are being used today.

In general, the use of source material (uranium and thorium) in consumer products and building materials is declining. For thorium, the U.S. Geological Survey reports that this decline is the result of "highdisposal costs, increasingly stringent regulations, and public concerns related to thorium's natural radioactivity" (Hedrick 2006). It is believed the same conclusion holds true for uranium as companies look to reduce potential liabilities by finding alternative materials for their products. As a common example, the Coleman Company now uses yttrium hydroxide and cerium hydroxide for the production of its gas lantern mantles rather than thorium nitrate (Frame 2003). Further detail on the results of this research are provided below.

#### 3.1 Thorium Industry and Uses

Thorium can be found in almost all rocks, soils, water bodies, plants, and animals with most soils having an average thorium concentration of 6 ppm (EPA 2006a; UIC 2006). Almost all the mass of natural thorium exists as <sup>232</sup>Th, but <sup>230</sup>Th and <sup>228</sup>Th can be significant in the environment, and all three naturally occurring isotopes are radioactive (EPA 2006a). Monazite, a rare-earth thorium-phosphate mineral, is the most common mineral containing thorium with up to 12 percent being composed of thorium oxide but averaging 6 to 7 percent (UIC 2006). Monazite was last produced in the United States in 1994, but 4930 kg of thorium compounds (e.g., oxides, nitrates), having an estimated value of \$145,000, were imported in 2005 (Hedrick 2006).

Thorium oxide has been widely used in refractory products because of its high melting point of 3330°C, which is the highest of all oxides (LANL 2003). Many ceramic products that must withstand high temperatures contain thorium oxide (also known as thoria), including crucibles, firebrick, and fibers in fabrics and tape used on space shuttles (LANL 2003; Thermal Ceramics, Inc. 2000). Magnesium-thorium alloys were also used for their high-temperature tolerance in the aerospace industry, but lanthanides, zirconium, and yttrium can now be used as substitutes for the thorium in these alloys (USGS 1999). Investment casting—also known as the lost-wax process—usually includes the application of zircon sand after wax molds are dipped into ceramic slurry, and both the ceramic and zirconium can contain trace amounts of thorium (Beatty 2006a). Where products containing trace amounts of radioactive materials are unacceptable, zirconium powders (used in bioceramics for the construction of artificial joint implants) are specifically manufactured so that virtually all radioactive impurities are eliminated (Wright Medical Technology, Inc. 2004; Heimann 2001). Gas lantern mantles containing thorium were once popular because at 2000°C the mantle is still solid and emits an incandescent glow (ORAU 2004d). The thorium compounds in lantern mantles have now largely been replaced with yttrium compounds (USGS 1999). However, high-intensity discharge lamps used in outdoor and industrial settings still do contain thorium (NRC 2001). The overall trend is for ceramic and gas mantle industries to continue replacing thorium with less or nonradioactive substitutes.

Thorium has other uses besides being a refractory material. It has been used in cigar/cigarette lighter flints and ceramic tableware glazes (NRC 2001). Welding rods contain thorium to increase the current carrying capacity of the electrode and reduce contamination of the weld (ORAU 2004e). Magnetron tubes containing thoriated cathodes have been used in radar systems for air traffic control, surveillance, and weather monitoring (Hedrick 1999). The magnetron tube is also the type of vacuum tube used in microwave ovens (Hedrick 1999; Sprague 2006). Thorium can be used as a target material for x-rays and in radiation detectors (Hedrick 1999). Thorium powder is used as a pigment to color glass (Beatty 2006b). Thorium also increases the index of refraction in glass so that high-quality lenses (e.g., camera lenses) can be built with less curvature, which makes the lenses thinner and lighter (Frame 2004a). Thorium fluoride is used as an anti-reflective coating in the ultraviolet, visible, and infrared light spectrums (CERAC Inc. 2006). Currently, thorium is still used in welding electrodes and magnetrons, but applications for thorium have largely disappeared as new technologies and substitute materials replace an element that arouses public concern, has a high disposal cost, and is subject to stringent regulations (Hedrick 2006).

However, the unique properties of thorium and the expense of potential alternatives have not allowed it to be replaced in all applications. Thorium remains in common use for lighting, welding electrodes, and chemical catalysts (e.g., conversion of ammonia to nitric acid, petroleum cracking, and production of sulfuric acid), with a continued shift away from refractory applications (Hedrick 2006; LANL 2003). Thorium also continues to be used in microwave ovens, high-refractivity glass, and photo-conductive films (Short 2006), and is now being used in Japan as an exhaust catalyst to decrease soot emissions from diesel engines (PBI Media 2003). Thorium-based thin films are under development for use in extreme ultraviolet lithography, microscopes, and astronomy (Allred and Turley 2003a). Thorium consumption in the United States during the past decade has remained at historical lows, as shown in Figure 1 depicting apparent U.S. consumption of thorium by year from 1902 to 2002. Non-energy applications of thorium in the United States are expected to remain low (Hedrick 2006).

#### 3.2 Uranium Industry and Uses

Uranium is found at low levels in almost all rock, soil, and water bodies, but significant concentrations do occur in some environments (EPA 2006b). Certain ores or minerals, such as phosphate or bentonite, can contain elevated levels of uranium; therefore, products made from these ores or minerals (e.g., fertilizers and cat litter) can be slightly radioactive (ORAU 2004a, c). In some geographical areas, the uranium concentration may be so high that the filter materials used in publically owned water treatment plants can accumulate enough uranium to classify the filter residue as source material (Gatchett 1994). Coal ash can also accumulate uranium and thorium, which becomes an issue when the ash is used in road construction or other commercial applications (EHSO 2006; TFHRC 2006). Oil and gas production may produce scales on pipes that concentrate uranium and thorium, so careful monitoring is warranted when reusing these pipes (EPA 2006c). Other minerals and ores associated with uranium include aluminum, copper, fluorspar, gypsum, molybdenum, niobium, potassium, tin, titanium, tungsten, and vanadium (EPA 2006c). Waste rock, mill tailings, and wash slimes associated with these minerals and ores should be screened to prevent the distribution of concentrated naturally occurring uranium.



**Figure 1.** Apparent U.S. consumption of thorium by year from 1902 to 2002. Note that U.S. thorium consumption during the past decade is near historical lows. Based on data from USGS (DiFrancesco and Hedrick 2004).

Almost all natural uranium exists as <sup>238</sup>U, approximately 99.2745 atom percent, while <sup>235</sup>U and <sup>234</sup>U are the only other natural isotopes, occurring at 0.7200 atom percent and 0.0055 atom percent, respectively. All three of these uranium isotopes are radioactive (CRC 1994; EPA 2006b). Enriched uranium, which has a somewhat higher specific activity than naturally occurring uranium because of its higher <sup>235</sup>U content, is the basis for commercial nuclear reactor fuel with a <sup>235</sup>U concentration typically around 2 to 3 percent or higher (EPA 2006b; ATSDR 1999b). Large quantities of uranium with depleted amounts of <sup>235</sup>U remain after enrichment. The isotopic concentrations of depleted uranium are around 99.75 percent <sup>238</sup>U, 0.25 percent <sup>235</sup>U, and 0.0005 percent <sup>234</sup>U (Voss 2001).

Naturally occurring uranium has traditionally been used as a dye and has been used to color glass since the 1830s (ORAU 2004g). Uranium or vaseline glass, a type of glass colored with uranium and developed in the 1830s, continues to be a popular decorative glass (ORAU 2004g; Beatty 2006c). Until World War II, the glass was colored with uranium dioxide, but depleted uranium replaced uranium dioxide when glass production resumed in 1959 after a 15-year break (ORAU 2004g). Glass enamel, such as that found in Cloisonné jewelry (ORAU 2005a), was permitted by exemption to contain not more than 10 percent by weight of source material prior to July 25, 1983 (10 CFR 40.13(c)(2)(iv)). Glazes containing uranium were developed in the 1930s in many vibrant colors; these glazes also acted as a sealant for ceramics (ORAU 2005b; Frame 2004b). Uranium glazes have a history similar to uranium glass, but commercial use of uranium glazes ended by the late 1980s (ORAU 2005b; Frame 2004b). Uranium was even used as a fluorescent pigment in porcelain dentures starting in the 1940s, but this practice was discontinued around 1986 (Binney and Scherpelz 1977; ORAU 2004f). Aside from still being used as a pigment for glass, uranium is also currently used as a stain or dye for wood and leather and as a mordant for wood and silk (ATSDR 1999c). Public concern over the radiation in these products has limited their consumer audience, although hobbyists do persist.

Uranium was once a popular quack cure. With the fame of hot springs in the 1800s came the assumption that radiation was the source of a spa's restorative powers (Frame 1989). Beginning in 1912, products were created that would add radon to drinking water via containers or disks made with a combination of cement and uranium (Frame 1989; ORAU 1998). The practice of drinking radiated water ended by the mid-1930s, but the idea that radiation could be beneficial to health did not (Frame 1989). Japanese textile manufacturers currently market thorium-containing ceramic fabrics for wrist bands, undergarments, socks, sheets, and clothing (ORAU 1998; MEXT 2004; Fujibo Holdings Inc. 2005). Ceramic disks, tobacco cards, and kitchenware that have small amounts of thorium are also marketed in Japan, promoting the idea that radiation possesses cleansing, deodorizing, and germicidal properties (ORAU 1998). The use of uranium as a supposed health aid has disappeared, but the benefits of natural radiation are still touted in the form of these thorium-containing ceramics.

The majority of current applications for uranium, however, are scientific. Uranyl acetate is used in analytical chemistry as an indicator or titrant because of its ability to precipitate sodium (Fujibo Holdings Inc. 2005; Reckhow 2006). It can also be used as a stain in electron microscopy (Monash University 2005). Uranium carbide is reportedly a good catalyst for the production of ammonia (ATSDR 1999c). Uranium oxide is under development as a highly reflective coating for mirrors in astrophysics applications (Allred et al. 2002; Allred and Turley 2003b; Artioukov et al. 2004; Squires et al. 2000). Oak Ridge National Laboratory is also researching uranium oxide, investigating its role as a catalyst for the oxidative destruction of volatile organic compounds (VOCs), halogenated VOCs, and other short-chain alkanes (ORNL 2002; Taylor and O'Leary 2000). The semiconductive properties of uranium oxides are under scrutiny, especially with regard to solar cells (Meek et al. 2001). Additional research includes using uranium in rechargeable batteries and the development of uranium alloys that provide special properties to steel with respect to elastic limit and tensile strength (Haire 2004; AZoM 2006).

Other uses for uranium are either diminishing in popularity or exceed the 15-pound weight limit in 10 CFR 40.22. Uranyl nitrate has been used as a photosensitive salt since the mid-1800s and can also be used as a toner to alter an image created by another metallic salt, but only photography hobbyists still use it today because of the superiority of silver halides (ORAU 2006b). The facing material of frizzens used in flintlock rifles was made with depleted uranium as uranium can produce sparks by friction (ORAU 2004b). This process began in the early 1970s only to disappear by the mid-1990s because of concerns regarding the fragments of uranium that became airborne after the flintlock was discharged (ORAU 2004b). Uranium dioxide has been used to extend the lives of filaments in large incandescent lamps (ATSDR 1999c). Uranium metal can be used as an x-ray target for the production of high-energy x-rays (EPA 2002). Depleted uranium is receiving consideration as a radiation shielding material and could potentially replace lead in aprons or steel in ceramic metallic (CERMET) composite waste packages used for the containment of spent nuclear fuel (Haire and Price 2001). Depleted uranium is used as a ballast and penetrator for missiles, as a counterweight for aircraft control surfaces, and in gyro compasses and other inertial guidance devices (EPA 2002, 2006b; LANL 2004; ORAU 2006a). Many of the

applications that use depleted uranium require a specific license for production and require registration for use under the general license at 10 CFR 40.25, so would not be covered by 10 CFR 40.22.

#### 3.3 National Security Experience

Because of increased security awareness during the past 5 years, more information is becoming known about the radioactive content of consumer products and building materials. This is partially the result of the increased number of radiation portal monitors being installed throughout the United States (e.g., buildings, train stations, border crossings, ports of entry, railways). To reduce the number of false alarms at these facilities, developers are pursuing a better understanding of what types of radioactive materials are commonly in use (Kouzes et al. 2006). The ranges of specific activities of <sup>40</sup>K, <sup>226</sup>Ra, <sup>238</sup>U, and <sup>232</sup>Th observed in common cargo materials are reported in Table 1. The specific activities reported in Table 1 were used to calculate whether these cargo materials would be considered an "unimportant quantity" of source material under 10 CFR 40.13(a) (i.e., uranium and thorium is by weight less than 0.05 percent of the mixture, compound, solution, or alloy). The uranium and thorium percent of total mass is reported in Table 2 as "Source Material." Note that of these common cargo materials, only the higher concentrations of monazite sand would not qualify as an exempt concentration of source material under 10 CFR 40.13(a). Cargo items known to contain significant levels of naturally occurring radioactive material (NORM) and technically enhanced naturally occurring radioactive material (TENORM) are listed in Table 3. Cargo items that have been observed to cause alarms at border crossings are listed in Table 4. These cargo items were each previously addressed in NUREG-1717, providing some anecdotal evidence of limited new applications of source material or borderline source material.

#### 3.4 Identified New Uses of Source Material

Based on the results of the research into current uses of source material, only two new uses were identified that were sufficiently different from uses previously evaluated by NRC to warrant further evaluation. These new uses are:

- 1. Thorium and uranium thin-film optical coatings. Thin-film optical coatings are used on the surface of glass lenses to reduce reflection and glare in the ultraviolet, visible, and infrared light spectra and to increase reflection in extreme ultraviolet (EUV) and soft x-ray spectra. Thorium has been used to reduce reflection, while uranium and thorium have been used to increase reflection.
- 2. Filtration of uranium from drinking water at municipal water treatment plants. The EPA has promulgated new regulations that included a maximum contaminant level (MCL) for uranium of 30 µg/L in drinking water (65 FR 76708). The regulation requires that community water systems of all sizes be in compliance by December 31, 2007. Filtering uranium is of potential concern to the NRC because several available remediation alternatives act to concentrate uranium.

A third potential new use was identified: thorium exhaust catalysts. As discussed previously, thorium is now also being used in Japan as an exhaust catalyst to decrease soot emissions from diesel engines (PBI Media 2003). However, the NRC is currently addressing a petition to amend 10 CFR 40.13(c) to exempt the use of thorium as an exhaust catalyst (Hee 2003). Based on this action, no further evaluation of this potential use was determined to be necessary.

	Specific Activity (Bq/kg) of Isotopes in Substances							
	4	٥K	226	Ra	:	<sup>238</sup> U	23	<sup>2</sup> Th
Substance	Low	High	Low	High	Low	High	Low	High
Adobe	300	2000	20	90	20	90	23	200
Alum shales (Sweden)	10,300	_	2200	_	2200	_	6.1	_
Banana	130	_	0.037	_	_	_	_	_
Basalt	3900	_	37	_	37	_	37	_
Beer	14	_	-	_	_	_	_	_
Brazil nuts	210	_	37	259	_	_	_	_
Carrot	130	_	0.022	0.07	_	_	_	_
Cat litter	200	300	_	_	21	136	18	43
Coal ash	44	1060	22	220	56	440	44	280
Concrete	150	500	40	_	40	_	40	_
Diorite	5200	_	51	_	51	_	24	_
Feldspar	2000	4000	40	100	40	100	70	200
Fertilizers	40	8000	20	1000	230	2300	20	30
Gabbro	2100	_	33	_	33	_	20	_
Granite	600	10,000	30	500	30	500	40	70
Granodiorite	7800	_	99	_	99	_	73	_
Light salt	8060	_	_	_	_	_	_	_
Lima Bean (raw)	170	_	0.074	0.19	_	_	_	_
Limestones	780	_	15	-	15	_	5.3	_
Marble	40	200	20	30	20	30	20	_
Marble tile	965	_	_	_	63	_	220	_
Monazite sand	40	70	30	1000	30	1000	50	3000
Peridotite	2300	_	18	_	18	_	12	_
Phosphates	_	_	5	800	150	2300	5	170
Red Meat	110	_	_	_	_	_	-	_
Sandstone	40	1000	70	_	70	_	70	_
Scotch Brite <sup>™</sup> pads	210	_	_	_	350	_	310	_
Shales	7800	_	40	_	40	_	4.5	_
Slate	1000	_	70	—	70	_	70	_
US Soils	10,300	_	160	—	140	_	130	_
White Potatoes	130	_	0.093	_	_	_	_	_

**Table 1.** Ranges of specific activity in common cargo materials (from Kouzes et al. 2006). Single valuesreported by Kouzes et al. (2006) are conservatively recorded here as a low value because theactual range of values may include higher values.

	Percentage of Substance by Mass						
	<sup>226</sup> I	Ra	238	U	232	Th	Source Material
Substance	Low	High	Low	High	Low	High	High
Adobe	0.000	0.000	0.000	0.001	0.001	0.005	0.006
Alum shales (Sweden)	0.000	_	0.018	_	0.000	_	0.018
Banana	0.000	_	_	_	_	_	_
Basalt	0.000	_	0.000	_	0.001	_	0.001
Beer	_	_	_	_	_	_	_
Brazil nuts	0.000	0.000	_	_	_	_	_
Carrot	0.000	0.000	_	_	_	_	_
Cat litter	_	_	0.000	0.001	0.000	0.001	0.002
Coal ash	0.000	0.000	0.000	0.004	0.001	0.007	0.010
Concrete	0.000	_	0.000	_	0.001	_	0.001
Diorite	0.000	_	0.000	_	0.001	_	0.001
Feldspar	0.000	0.000	0.000	0.001	0.002	0.005	0.006
Fertilizers	0.000	0.000	0.002	0.019	0.000	0.001	0.019
Gabbro	0.000	_	0.000	_	0.000	_	0.001
Granite	0.000	0.000	0.000	0.004	0.001	0.002	0.006
Granodiorite	0.000	_	0.001	_	0.002	-	0.003
Light salt	_	_	_	_	_	_	_
Lima Bean (raw)	0.000	0.000	_	_	_	-	_
Limestones	0.000	_	0.000	_	0.000	-	_
Marble	0.000	0.000	0.000	0.000	0.000	_	0.001
Marble tile	_	_	0.001	_	0.005	_	0.006
Monazite sand	0.000	0.000	0.000	0.008	0.001	0.074	0.082
Peridotite	0.000	_	0.000	_	0.000	_	_
Phosphates	0.000	0.000	0.001	0.019	0.000	0.004	0.023
Red Meat	-	—	_	—	—	_	_
Sandstone	0.000	_	0.001	_	0.002	_	0.002
Scotch Brite <sup>™</sup> pads	-	_	0.003	_	0.008	-	0.010
Shales	0.000	_	0.000	_	0.000	_	_
Slate	0.000	_	0.001	_	0.002	-	0.002
U.S. Soils	0.000	_	0.001	_	0.003	-	0.004
White Potatoes	0.000	_	_	_	_	_	_

Table 2. Ranges of percent of mass of three radionuclides in common cargo materials (calculated from Kouzes et al. 2006). The column labeled "Source Material" contains the sum of the maximum reported values of <sup>238</sup>U and <sup>232</sup>Th. Note that only the higher concentrations of monazite sand qualifies as source material.

Table 3.	Cargo items known to	contain significant	levels of NORM	and TENORM	as uranium or
	thorium (adapted from	1 Kouzes et al. 2006	, Table 2).		

Cargo Item	<sup>238</sup> U	<sup>232</sup> Th
Aircraft counterweights	Х	
Camera lenses and other optical glass		х
Cloisonné jewelry	Х	
Colored ceramic glazes	Х	
Dental ceramics	Х	
Firebrick		Х
Gas lantern mantles		Х
Gyroscope rotors	Х	
Magnesium-thorium alloys (aircraft parts)		х
Manufactured anhydride (byproduct gypsum)	Х	Х
Phosphogypsum	Х	
Polishing powder		Х
Specialty glass	Х	
Strike plate for flintlock muskets (misch metal)	Х	
Thoriated tungsten welding electrodes (welding rods)		х
Tundish nozzles used in steel making and smelting		х
Zirconium sand		Х

**Table 4.** Cargo items observed to cause alarms at border crossings (adapted from Kouzes et al. 2006,<br/>Table 4).

Cargo Item			
Cat litter	Medical isotopes (In, I, Tc, Tl)		
Abrasives, scouring pads	Refractory material		
Mica	Fertilizer, potash		
Granite, marble slabs	Ceramics, tile, toilets		
Trucks, cars	Aluminum		
Earth	Bentonite		
Salt	Other metal		
Television sets	Gas tankers		
Smoke detectors	Other		

# 4 Limitations of Thorium Exposure Assessment

### 4.1 Background

Radiological hazard assessments for natural uranium generally assume an isotopic composition of 99.2745 atom percent <sup>238</sup>U, 0.7200 atom percent <sup>235</sup>U, and 0.0055 atom percent <sup>234</sup>U (CRC 1994). While some exceptions do exist in nature, the majority of the world's resources of uranium have this composition and so there is little uncertainty when making this assumption.

On the other hand, the isotopic composition of natural thorium, specifically the <sup>230</sup>Th/<sup>232</sup>Th activity ratio, can vary widely between ores. This issue is addressed by Stannard (1988) who quotes Hursh (1974) and amplifies his arguments that there is no such thing as "natural thorium." The dose assessments reported in Chapter 5 of this report assume natural and processed thorium to be nearly 100 percent <sup>232</sup>Th by mass (50 percent by activity) and 50 percent <sup>228</sup>Th by activity. However, as was discussed in NUREG-1717 (NRC 2001) and further supplemented in the discussion below, the wide variability in this composition assumption for natural thorium can yield widely varying results.

# 4.2 Variability in the <sup>230</sup>Th/<sup>232</sup>Th Ratio of Natural Thorium

Radiological hazard assessments for thorium generally ignore the <sup>230</sup>Th content of thorium. This is an acceptable approach for monazite ores that typically have a <sup>230</sup>Th/<sup>232</sup>Th ratio of about 0.08 to 0.12 (Frondel 1958). However, <sup>230</sup>Th must be considered in dose assessments for products containing thorium from other source ores.

For reference, the thorium (4n) and uranium (4n+2) series are depicted in Figures 2 and 3. Depending on the source of the thorium, there may be very little <sup>230</sup>Th (e.g., thorium from monazite, which contains very little uranium), or there may be a significant amount. Monazite ore is the most common source for thorium, not so much for its thorium content that is relatively low (10 percent by weight), but because of its availability following processing as a rare-earth ore (USGS 2006). If demand for thorium were to increase, it is likely that higher grade thorium ores would be in demand (USGS 2006), most of which have a higher <sup>230</sup>Th/<sup>232</sup>Th ratio compared to monazite ore.

Numerous data are reported in geologic literature on <sup>238</sup>U-<sup>230</sup>Th disequilibrium, as techniques exist to date and characterize the formation of basalts based on the disequilibrium. Many of these studies report <sup>230</sup>Th/<sup>232</sup>Th activity ratios that range from 0.738 to 1.960 (e.g., Fretzdorff et al. 2003; Pyle et al. 1988; Bourdon et al. 1996; Zou et al. 2002). However, <sup>230</sup>Th/<sup>232</sup>Th activity ratios above 100 are not uncommon in some Brannerite and Davidite ore samples and even above 1000 in some Betafite and Uraninite ore samples (Frondel 1958). Figure 4 shows the <sup>230</sup>Th/<sup>232</sup>Th activity ratio for eight types of thorium ore. It is notable that monazite ores are low in uranium content, but that many higher-grade thorium ores contain significant amounts of uranium. Many urananite and betafite ores have a very high <sup>230</sup>Th/<sup>232</sup>Th activity ratio. In summary, although it is somewhat common practice to assume natural thorium is 100 percent <sup>232</sup>Th, this can result in a substantial underestimation of dose from intakes of ores with a high <sup>230</sup>Th/<sup>232</sup>Th activity ratio. If thorium usage increases substantially in the future, this assumption should be re-addressed to ensure that regulations governing the use of source materials remain protective of human health.

# 4.3 Variability in the <sup>228</sup>Th Content of Processed Thorium

When <sup>232</sup>Th is in thorium ore, it comes into secular equilibrium with all of its radioactive decay products all the way through stable <sup>208</sup>Pb, as listed in Table 5. When thorium metal (<sup>232</sup>Th and <sup>228</sup>Th present in the same activity concentration) is separated from thorium ore, <sup>232</sup>Th exists in varying states of equilibrium with its decay products, including <sup>228</sup>Th, as shown in Figure 5. Immediately after separation, there is no <sup>228</sup>Ra or <sup>228</sup>Ac (a potent gamma-emitter) present. At 4.55 years after separation, <sup>228</sup>Th, which is normally supported by <sup>228</sup>Ra, drops to 42.2 percent of equilibrium, and with it, all of its decay products through <sup>208</sup>Pb. By 10 years, it approaches 60 percent of equilibrium, and by 41.5 years after separation, it is at 99 percent of equilibrium.

Dose rate constants for <sup>232</sup>Th in equilibrium or disequilibrium with its decay products are not available in common reference materials. Although of low specific activity (4.06 MBq/kg or 110  $\mu$ Ci/kg of the parent nuclide), <sup>232</sup>Th in equilibrium with its decay products is a potent gamma-emitter on an activity basis. The specific exposure rate constant of <sup>232</sup>Th in equilibrium with its decay products is 1.55 R/h m<sup>2</sup>/Ci (of <sup>232</sup>Th)<sup>4</sup>, higher than for <sup>60</sup>Co. Constants for ambient dose equivalent rate ( $\Gamma_{H^*}$ ), directional dose equivalent rate ( $\Gamma_{H^*}$ ), and air kerma rate ( $\Gamma_K$ ), calculated using ICRP-60 tissue weighting factors ( $w_T$ )<sup>5</sup>, are given in Table 6. Non-zero contributions from 429 x- and  $\gamma$ -photons are included. The top row in Table 6 is for nearly the minimum activity of <sup>228</sup>Th at around 4.4 years, while the second row is for decay products in equilibrium with the <sup>232</sup>Th parent.

Massive sources of thorium metal will, of course, absorb or down-scatter photons. However, <sup>208</sup>Tl emits a 2.614-MeV gamma in virtually all of its transitions, and despite the fact that the branching ratio to <sup>208</sup>Tl is just under 36 percent, this highly penetrating gamma photon is the largest single contribution to  $\Gamma_{H^*}$  and  $\Gamma_{\kappa}$ , and is a hallmark of the presence of <sup>232</sup>Th in a material.

Five kinds of dose rate constants for external irradiation are presented in Table 7 for <sup>232</sup>Th in secular equilibrium with its decay products, calculated from the results in Eckerman (2004). These can be used for dose assessments for various exposure scenarios. The coefficients are, respectively,

<sup>&</sup>lt;sup>4</sup> Calculated August 28, 2006, using MicroShield Version 7.01 by Steven J. Maheras, Battelle Columbus Operation, Columbus, Ohio.

<sup>&</sup>lt;sup>5</sup> Calculated August 29, 2006, based on ICRP Publication 38 radionuclide emission data

<sup>(</sup>x and gamma) from ICRP38.exe v. 1.0 (March 25, 2003) and ICRP Pub. 74 fluence-to-air-kerma, ambient-doseequivalent-per-unit-air-kerma, and directional-dose-equivalent-per-unit-air-kerma data from Rad Toolbox v. 1.0.0 (December 31, 2003). Photons below 10 keV were ignored because ICRP Pub. 74 (ICRP 1996b) does not have data below 10 keV. Bremsstrahlung from  $\beta^-$  emissions was not included. Both codes can be downloaded from http://ordose.ornl.gov/downloads.html. Calculations done by Daniel J. Strom, Pacific Northwest National Laboratory, Richland, Washington.

- the specific air kerma rate constant<sup>6</sup>
- the effective dose rate constant for submersion
- the effective dose rate constant for a ground plane source
- the effective dose rate constant for a soil volume
- the effective dose rate constant for water immersion.

The submersion and soil volume coefficients can be converted from volume units (mL) to "conventional" mass units (g) by multiplying by 0.00120 g/mL (at 20°C and 1 atmosphere) and 1.6 g/mL, respectively.

Similar coefficients are given in Table 8 for elemental thorium about 4.4 years after separation from its decay products. This period of time is very close to the minimum activity of <sup>228</sup>Th, and the coefficients are about 42 percent of the values for full equilibrium.

In summary, <sup>228</sup>Th may not be in equilibrium with <sup>232</sup>Th in newly refined samples. Figure 6 shows the decline in <sup>228</sup>Th activity following separation of thorium from impurities, including radium. The 1.91-year <sup>228</sup>Th decays, only to be re-supported by the ingrowth of 5.75-year <sup>228</sup>Ra. Because of the noted variability, analyses for thorium were conducted for pure thorium and thorium with all progeny.

### 4.4 Summary of Internal and External Dosimetry Issues for Thorium

Because there is no well-defined mixture of radioactive thorium isotopes, dose assessment scenarios for thorium that include intakes by ingestion or inhalation require the assumption of some mixture of <sup>232</sup>Th and its decay product <sup>228</sup>Th along with <sup>230</sup>Th, which originates in the <sup>238</sup>U decay series. Assuming that all <sup>232</sup>Th decay products are in equilibrium maximizes the projected dose from an intake of <sup>232</sup>Th. Assuming some plausible ratio of <sup>230</sup>Th to <sup>232</sup>Th is required to complete the dose assessment. The value of <sup>230</sup>Th activity equaling 11 percent of <sup>232</sup>Th activity adopted in Section 3.1.5 of NUREG-1717 may not be conservative if thorium originates from ores other than monazite.

For external irradiation, the assumption that all decay products of <sup>232</sup>Th are in equilibrium may overestimate the dose by up to a factor of about 4 for some values of time since separation of elemental thorium from ore, but this is "conservative" in the sense of overestimating dose from an assessment perspective. If a significant fraction of the activity is <sup>230</sup>Th, then the photon dose rate from <sup>226</sup>Ra and its decay products may predominate over time. The photon dose rate grows in over time with the 1599-year half-time of <sup>226</sup>Ra. By 100 years of age, <sup>226</sup>Ra reaches 4.25 percent of the initial <sup>230</sup>Th activity, 12.2 percent by 300 years, and 35.3 percent by 1000 years, as shown in Figure 7.

<sup>&</sup>lt;sup>6</sup> According to Eckerman (2004, p. 655), the "Specific Air Kerma Rate Constant" is calculated using mass energy *absorption* coefficients  $\mu_{en}/\rho$  rather than mass energy *transfer* coefficients  $\mu_{tr}/\rho$  so strictly speaking, they are specific air absorbed dose rate constants and are somewhat smaller than specific air kerma rate constants.



**Figure 2.** Thorium (2*n*) decay series that includes  $^{232}$ Th, highlighting the thoron decay chain and alpha contributors.



**Figure 3.** Uranium (4n+2) decay series that includes <sup>230</sup>Th, highlighting the radon decay chain and alpha contributors.



**Figure 4.** Ratio of <sup>230</sup>Th activity to <sup>232</sup>Th activity in various ores by weight percent of thorium in ore. Data from Frondel (1958).



Activity of Th-232 and progeny

**Figure 5.** Return to equilibrium of decay products of  $^{232}$ Th following separation of thorium metal ( $^{232}$ Th and  $^{228}$ Th) from thorium ore.



**Figure 6.** Decay and ingrowth of <sup>228</sup>Th after separation of thorium from impurities.



Figure 7. Decay of <sup>230</sup>Th and ingrowth of <sup>226</sup>Ra over time

		Atomic Mass, A	Specific Activity of Pure Nuclide
Nuclide	Half-life, $T_{1/2}$	(g/mol)	(Bq/kg)
<sup>232</sup> Th	$1.405 \times 10^{10}  yr$	232.038050	4,057,224
<sup>228</sup> Ra	5.75 yr	228.03	$1.01 \times 10^{16}$
<sup>228</sup> Ac	6.15 hr	228.03	$8.27 \times 10^{19}$
<sup>228</sup> Th	1.912 yr	228.028731	$3.03 \times 10^{16}$
<sup>224</sup> Ra	3.66 d	224.020202	$5.89 \times 10^{18}$
<sup>220</sup> Rn	55.6 s	220.011384	$3.41 \times 10^{22}$
<sup>216</sup> Po	0.145 s	216.001905	$1.33 \times 10^{25}$
<sup>212</sup> Pb	10.64 hr	212.0	$5.14 \times 10^{19}$
$^{212}$ Bi	60.55 min	211.991271	$5.42 \times 10^{20}$
<sup>212</sup> Po	0.299 μs	211.988852	$6.59 \times 10^{30}$
<sup>208</sup> T1	3.053 min	207.98	$1.10 \times 10^{22}$
<sup>208</sup> Pb	stable	207.976636	0

**Table 5.** The  $^{232}$ Th (4*n*) Decay Chain

**Table 6.** Ambient dose equivalent rate, directional dose equivalent rate, and air kerma rate constants for  $^{232}$ Th in two extremes of equilibrium with its decay products, calculated using ICRP-60 tissue weighting factors ( $w_{\tau}$ ) and data from ICRP Publications 38 and 74 (ICRP 1984, 1996b).

		$\Gamma_{H^*}$ (Ambient		
		Dose	$\Gamma_{H'}$ (Directional	
Parent		Equivalent;	Dose Equivalent;	
Nuclide	Equilibrium	rem/h m <sup>2</sup> /Ci)	rem/h m²/Ci)	$\Gamma_K$ (Air Kerma; rad/h m <sup>2</sup> /Ci)
<sup>232</sup> Th	4.4 y	0.546	0.671	0.593
<sup>232</sup> Th	Secular	1.307	1.574	1.384

	Specific	Effective Dose Rate Coefficient (rem/hour)/				fficient(ren/hour)/ Specific <u>Effective Dose Rate Coefficie</u>				
	Air Kenna Rate Constant	ė <sub>511</sub> 5	<sup>é</sup> grd	e soil	e <sub>imm</sub>	Air Kenna Rate Constant	é <u>suð</u>	ë grd	ê <sub>scil</sub>	ė <sub>imm</sub>
	(rad/h					(Gy/s)				
Nuclide	m <sup>2</sup> /Ci)	(µCimL <sup>-1</sup> )	(µCicm <sup>2</sup> )	(µCimL <sup>-1</sup> )	(µCimL <sup>-1</sup> )	(m²/Bq)	(Bq m <sup>-3</sup> )	(Bq m <sup>-2</sup> )	(Bq m <sup>-3</sup> )	(Bq m <sup>-3</sup> )
Th-232	2.98E-02	9.64E-02	6.06E-05	3.25E-05	2.18E-04	2.24E-18	7.24E-18	4.55E-19	2.44E-21	1.64E-20
Ra-228	0	0	0	0	0	0	0	0	0	0
Ac-228	5.94E-01	5.98E+02	1.25E-01	4.04E-01	1.29E+00	4.46E-17	4.49E-14	9.39E-16	3.03E-17	9.70E-17
Th-228	3.41E-02	1.08E+00	2.84E-04	5.11E-04	2.40E-03	2.56E-18	8.10E-17	2.13E-18	3.84E-20	1.80E-19
Ra-224	6.21E-03	5.71E+00	1.22E-03	3.37E-03	1.25E-02	4.66E-19	4.29E-16	9.15E-18	2.53E-19	938E-19
Rn-220	1.97E-04	2.29E-01	4.92E-05	1.53E-04	4.98E-04	1.48E-20	1.72E-17	3.69E-19	1.15E-20	3.74E-20
Po-216	8.38E-06	1.03E-02	2.14E-06	7.01E-06	2.24E-05	6.29E-22	7.75E-19	1.61E-20	5.26E-22	1.68E-21
Рь-212	1.43E-01	8.31E+01	1.80E-02	4.61E-02	1.82E-01	1.08E-17	6.24E-15	1.35E-16	3.46E-18	137E-17
Bi-212	1.33E-01	1.19E+02	3.00E-02	7.94E-02	2.53E-01	1.00E-17	8.95E-15	2.25E-16	5.96E-18	190E-17
Po-212	0	0	0	0	0	0	0	0	0	0
TI-208	4.93E-01	8.10E+02	1.42E-01	5.61E-01	1.75E+00	3.70E-17	6.08E-14	1.07E-15	4.21E-17	131E-16
Totals	1.435	1617	0317	1.094	3.492	1.08E-16	1.21E-13	2.38E-15	8.21E-17	2.62E-16

**Table 7.** Dose coefficients for <sup>232</sup>Th in equilibrium with its decay products (calculated from Eckerman [2004]). Calculations include the branching fractions of 0.3597 to <sup>208</sup>Tl and 0.6403 to <sup>212</sup>Po.

**Table 8.** Dose coefficients for elemental thorium about 4.4 years after separation (calculated from<br/>Eckerman [2004]). Calculations include the branching fractions of 0.3597 to <sup>208</sup>Tl and 0.6403 to<br/><sup>212</sup>Po, as well as the fractional equilibrium factors listed below.

	8		Air	Effective	Dose Rate (	Coefficient (1	en/hour)/	S pecific	Effective	Dose Rate	Coefficient	(Svs <sup>-1</sup> )/
			Kerma Rate Corstant	ê <u>10</u> 6	<sup>é</sup> grd	ê <sub>soil</sub>	e <sub>imm</sub>	Air Kenna Rate Constant	é <sub>suð</sub>	ë grd	ë <sub>soil</sub>	ė <sub>imm</sub>
	Branch	Equilib-	(rad/h					(Gy/s)	्	00	्	
Nuclide	ing	ninn	m²/Ci)	(µCimL <sup>-1</sup> )	(µCicm <sup>2</sup> )	(µCimL <sup>-1</sup> )	(µCimL <sup>-1</sup> )	(m²/Bq)	(Bq m <sup>3</sup> )	(Bq m <sup>-2</sup> )	(Bq m <sup>3</sup> )	(Bq m <sup>3</sup> )
Th-232	1	1	2.98E-02	9.64E-02	6.06E-05	3.25E-05	2.18E-04	2.24E-18	7.24E-18	4.55E-19	2.44E-21	1.64E-20
Ra-228	1	0.4064	0	0	0	0	0	0	0	0	0	0
Ac-228	1	0.4064	2.42E-01	2.43E+02	5.08E-02	1.64E-01	5.25E-01	1.81E-17	1.82E-14	3.82E-16	1.23E-17	394E-17
Th-228	1	0.4217	1.44E-02	4.55E-01	1.20E-04	2.16E-04	1.01E-03	1.08E-18	3.42E-17	8.98E-19	1.62E-20	7.59E-20
Ra-224	1	0.4218	2.62E-03	2.41E+00	5.14E-04	1.42E-03	5.27E-03	1.97E-19	1.81E-16	3.86E-18	1.07E-19	396E-19
Rn-220	1	0.4218	8.30E-05	9.66E-02	2.07E-05	6.46E-05	2.10E-04	6.23E-21	7.26E-18	1.56E-19	4.85E-21	1.58E-20
Po-216	1	0.4218	3.53E-06	4.35E-03	9.05E-07	2.96E-06	9.44E-06	2.65E-22	3.27E-19	6.79E-21	2.22E-22	7.09E-22
Рь-212	1	0.4218	6.05E-02	3.51E+01	7.59E-03	1.94E-02	7.70E-02	4.54E-18	2.63E-15	5.69E-17	1.46E-18	5.78E-18
Bi-212	1	0.4218	5.62E-02	5.03E+01	1.26E-02	3.35E-02	1.07E-01	4.22E-18	3.78E-15	9.49E-17	2.51E-18	8.01E-18
Po-212	0.6403	0.4218	0	0	0	0	0	0	0	0	0	0
TI-208	0.3597	0.4218	2.08E-01	3.42E+02	6.00E-02	2.36E-01	7.38E-01	1.56E-17	2.56E-14	4.51E-16	1.78E-17	5.54E-17
Totals			0.613	673	0.132	0.455	1.453	4.60E-17	5.05E-14	9.89E-16	3.42E-17	1.09E-16

# 5 Source Material Dose Analyses

10 CFR 40.22 is a general license authorizing commercial and industrial firms; research, educational, and medical institutions; and Federal, State, and local government agencies to use and transfer not more than 15 pounds of source material at any one time for research, development, educational, commercial, or operational purposes. Not more than a total of 150 pounds of source material can be received in any one calendar year. Such receipt, possession, use, or transfer under this general license provides exemption from the provisions of 10 CFR Parts 19, 20, and 21, so long as the exemption shall not apply to any such person who is also in possession of source material under a specific license under 10 CFR Part 40. Additionally, persons who receive, possess, use, or transfer source material pursuant to this general license are prohibited from administering source material, or the radiation therefrom, either externally or internally, to human beings except as may be authorized by NRC in a specific license.

Uses under 10 CFR 40.22 were not evaluated as part of NUREG-1717 (NRC 2001). Under the general license, source materials identified and characterized include uranium and thorium metals,  $UO_2$ ,  $U_3O_8$ ,  $UO_3$ ,  $ThF_4$ , and  $ThOF_2$ , and so on, that meet the quantity requirements of 10 CFR 40.22. For this assessment, PNNL staff assume thorium and uranium thin-film optical coatings are possessed or used under 10 CFR 40.22 and are in common use. However, source materials specifically exempted under 10 CFR 40.13(c) are not considered in this assessment.

This general license allows one to possess up to 15 pounds of natural uranium and thorium metal, oxide, and fluoride. Types of materials include sources used in industrial and research and development applications, material collected by hobbyists and collectors (e.g., uranium metal, yellowcake), and thorium-coated lenses. United Nuclear (http://www.unitednuclear.com), for example sells refined source material products, including thorium nitrate [Th(NO<sub>3</sub>)<sub>4</sub>], uranium metal, and uranium glass, to public, industry, commercial, and research and development organizations.

Potential new uses that will be described and analyzed below are thorium and uranium thin-film optical coatings and filtration for removal of uranium at drinking water facilities.

### 5.1 Thorium and Uranium Thin-Film Optical Coatings

Thin-film optical coatings are used on the surface of lenses to reduce reflection and glare in the ultraviolet, visible, and infrared light spectra and to increase reflection in EUV and soft x-ray spectra. Thorium has been used to reduce reflection, while uranium and thorium have been used to increase reflection.

The anti-reflective coatings work by using destructive interference of two separate surface reflections: one from the lens surface and one from the coating surface. The coating is designed so that the two reflected waves are exactly one-half wavelength out of phase; thus, the coatings are typically one-quarter wavelength thick. Thorium fluoride,  $ThF_4$ , is a low refractive-index material that is transparent from the ultraviolet to the infrared portions of the spectrum. Evaporated films are durable and chemically stable and exhibit low stress in thicknesses up to 2  $\mu$ m (Borah and Fay 2005). The visible range includes wavelengths of 400 to 700 nm, so the wavelength generally chosen for optical lenses is about 550 nm; lens coatings for this range would be about 140 nm thick. The infrared spectrum, which includes high-power

 $CO_2$  lasers with wavelengths from 8 to 12  $\mu$ m, can require coating thicknesses of 2 to 3  $\mu$ m for  $CO_2$  lasers and up to 10  $\mu$ m for higher wavelength IR applications.

Uranium and thorium optical mirror coatings are being researched at Brigham Young University, MOXTEK, Inc., and elsewhere for advances in the use of EUV and soft x-rays, freznel zone plates, and notch filters (Allred et al. 2002; Allred and Turley 2003b; Artioukov et al. 2004; Johnson et al. 2006; Squires et al. 2000). As applications for EUV radiation have been identified, the demand for better optics has also increased. Uranium and thorium are found to be more reflective in the 4.5 to 9 nm range than traditional reflectors (i.e., nickel, iridium, and gold) and there are potential applications in astronomical, medical, and biological imaging (Johnson et al. 2006). Thorium and thorium oxide thin films (19 to 61 nm thick) have been investigated for use in the EUV range (Johnson et al. 2006), along with uranium thin films (less than 10 nm) in the EUV and soft x-ray ranges (Allred et al. 2002; Allred and Turley 2003b; Artioukov et al. 2004; Squires et al. 2000). Presently, application of this technology is limited to producing high-resolution images for astronomy (i.e., IMAGE satellite); however, there are potential wideranging applications from developing smaller computer chips to producing higher resolution images of organic structures for biological and medical applications (Johnson et al. 2006).

Lenses are coated by sputtering in vacuum chambers by vaporizing the thorium fluoride or uranium and allowing it to deposit on the lens surface. Thorium fluoride evaporates at 800°C. Electron-beam or resistance-heated sources can be used to heat a molybdenum or tantalum hearth on which the thorium fluoride is placed. The recommended substrate temperature is 175°C, with a chamber pressure below 10<sup>-5</sup> Torr. The vapor deposits on the lens, as well as on all exposed surfaces in the vacuum chamber, which may be lined to capture the excess depositions. The manufacturing process generates a significant amount of waste. A similar process is used in the manufacture of uranium thin-film coatings.

#### 5.1.1 Description of Items

Lenses are made in many sizes. For analysis, a 5-cm-diameter lens was assumed. This lens size is reasonable for typical small camera, telescope, and laser applications. Larger lens sizes are expected for space-science applications (e.g., the uranium-coated EUV mirror used on the IMAGE satellite had a 12.3 cm diameter [Sandel et al. 2000]). A high-energy infrared lens with a 2000-nm coating of thorium fluoride would have a total deposition of 0.025 g of ThF<sub>4</sub> containing 0.019 g thorium, or about 77 Bq (assuming 100 percent <sup>232</sup>Th). The same size lens for an EUV application with a 10 nm coating of natural uranium would have a total deposition of 0.00037 g uranium, or about 10 Bq.

#### 5.1.2 Exemption Analysis: Scenario Description and Dose Assessments

Scenarios were developed to analyze normal use, accidents, and manufacturing. In normal use, it is reasonable to assume the primary exposure routes and times to a researcher using the thin-film coated lenses would be similar to those evaluated in NUREG-1717 for other users of thoriated lenses (i.e., television and photographic lenses). Other known applications for thin-film optical coatings, such as optics used in lasers or space telescopes, are expected to be shielded or used remotely, resulting in a lower expected dose. Further, the thin-film coatings contain such a limited amount of thorium or uranium (see Sections 5.1.2.1 and 5.1.2.2 for discussion) that no conceivable end use will result in an exposure of

concern during normal use and accident conditions. As described in Section 5.1.2.3, potential exposure comes during the manufacturing process where larger quantities of source material may be handled.

#### 5.1.2.1 Routine Use

The lenses evaluated in NUREG-1717 had incorporated up to 100 g thorium, while thin-film coated lenses have only about 0.02 g thorium or 0.0004 g uranium. The routine use scenario only has an external dose component. Routine doses from total thorium (i.e., with all progeny in equilibrium) would be about 0.0004 mrem/yr to an individual photographer and about 0.004 mrem/yr to a television camera operator (i.e., 0.02 percent of the values reported in NUREG-1717, Section 3.19.4, for thoriated lenses). Doses from thin-film uranium coatings were estimated using the ratio of gamma constants (Unger and Trubey 1982) for total thorium (2.07 × 10<sup>-6</sup> rem/ $\mu$ Ci-hr) and total uranium (1.32 × 10<sup>-7</sup> rem/ $\mu$ Ci-hr) and the ratio of activities for the example thorium and uranium lens coatings (i.e., 77 Bq for thorium and 10 Bq for uranium). This approach yielded routine doses from total uranium of about  $3.3 \times 10^{-6}$  mrem/yr to an individual photographer and about  $3.3 \times 10^{-5}$  mrem/yr to a television camera operator, about two orders of magnitude lower than those for thin-film thorium optical coatings. Individuals using coated lenses in recreational camera optics are expected to receive much lower doses. Using the same approach, doses were calculated for thorium, thorium progeny, uranium, and uranium short-lived progeny, with results shown as Table 9. Because of limited time between thorium and uranium separation and subsequent use, only short-lived progeny are considered for uranium and all progeny are considered for thorium. A description of these calculations is provided in Appendix A.

Table 9.	Radiation Dose	Assessment	for Routine	Use of	Thorium	and	Uranium	Thin-Film	Optical
	Coatings								

Source	Annual Scenario Dose (mrem)			
	Photographer	TV Camera Operator		
Thorium-Coated Optical Lens				
- Thorium Only	$2.9 \times 10^{-5}$	$2.9 \times 10^{-4}$		
- Thorium Progeny	$3.7 \times 10^{-4}$	$3.7 \times 10^{-3}$		
- Thorium + Progeny	$4.0  imes 10^{-4}$	$4.0 \times 10^{-3}$		
Uranium-Coated Optical Lens				
- Uranium Only	$2.4 \times 10^{-7}$	$2.4 \times 10^{-6}$		
- Uranium Short-Lived Progeny	$3.1 \times 10^{-6}$	$3.1 \times 10^{-5}$		
- Uranium + Short-Lived Progeny	$3.3 \times 10^{-6}$	$3.3 \times 10^{-5}$		

#### 5.1.2.2 Accidents

The generic accident methodology developed in NUREG-1717, Appendix A.2, was found to be reasonable and has been adapted for exposures to thorium and uranium coated lenses. It is assumed, first, that 10 percent of the available material would be deposited somewhere on the skin of an individual and, second, that 0.1 percent of this deposited material would be ingested before bathing removed the material from the body. Thus, the direct ingestion intake is estimated to be  $1 \times 10^{-4}$  times the total amount of available material on the lens. The same value is adopted for inhalation. This results in an ingestion and

inhalation of about 0.0077 Bq of thorium and about 0.001 Bq of natural uranium. The committed dose coefficients from ICRP Publication 72 (ICRP 1996a) were used to estimate the inhalation and ingestion doses for thorium and natural uranium (activity weighted average for  $^{234}$ U,  $^{235}$ U, and  $^{238}$ U) in 100 percent equilibrium with short-lived progeny. The committed dose for thorium was estimated to be about 1 µrem for ingestion and about 60 µrem for inhalation. The committed dose for natural uranium was estimated to be about 0.0005 µrem for ingestion and about 0.3 µrem for inhalation. Using the same approach, doses were calculated for thorium, thorium progeny, uranium, and uranium short-lived progeny, with results shown as Table 10. Because of limited time between thorium and uranium separation and subsequent use, only short-lived progeny are considered for uranium and all progeny are considered for thorium. A description of these calculations is provided in Appendix A.

Source	Annual Scenario Dose (mrem)				
	Ingestion	Inhalation	Effective <sup>†</sup>		
Thorium-Coated Optical Lens					
- Thorium Only	$2.3 \times 10^{-4}$	$5.9 \times 10^{-2}$	$5.9 \times 10^{-2}$		
- Thorium Progeny	$5.9 \times 10^{-4}$	$4.5 \times 10^{-3}$	$5.1 \times 10^{-3}$		
- Thorium + Progeny	$8.2 \times 10^{-4}$	$6.4 \times 10^{-2}$	$6.5 \times 10^{-2}$		
Uranium-Coated Optical Lens					
- Uranium Only	$4.7 \times 10^{-6}$	$3.2 \times 10^{-4}$	$3.2 \times 10^{-4}$		
- Uranium Short-Lived Progeny	$1.7 \times 10^{-7}$	$3.2 \times 10^{-7}$	$4.9 \times 10^{-7}$		
- Uranium + Short-Lived Progeny	$4.9 \times 10^{-6}$	$3.2 \times 10^{-4}$	$3.2 \times 10^{-4}$		

 Table 10.
 Radiation Dose Assessment for Accidents Involving Thorium and Uranium Thin-Film Optical Coatings

#### 5.1.2.3 Manufacturing

The manufacture of coated lenses primarily involves the use of thorium fluoride, with very limited use of uranium. The approach used in NUREG-1717 for estimating releases into a laboratory-type room was found to be reasonable and applicable for approximating releases in a small manufacturing facility. The scenario assumes resuspension of a fraction of the inventory in a laboratory-type room with an enclosure volume of 180 m<sup>3</sup>, a room height of 3 m, a ventilation rate of six volume changes per hour, and a breathing rate of 1.2 m<sup>3</sup>/hr. The time-integrated air concentration is adjusted to account for settling loss rate using the following approach.

$$\frac{d(Activity)}{dt} = -Activity \times \frac{ventilation \ rate}{room \ volume} - Activity \times \frac{deposition \ velocity}{room \ height}$$

The loss rate is then

loss rate = air exchange rate + 
$$\frac{deposition \ velocity}{room \ height}$$

For these high-density particles a deposition velocity of 0.01 m/s is appropriate, resulting in a loss rate of 18 hr<sup>-1</sup> (i.e., 6 hr<sup>-1</sup> + 0.01 m/s × 3600 s/hr ÷ 3 m). The settling loss (12 hr<sup>-1</sup>) dominates, even in the laboratory setting with six air exchanges per hour.

NUREG-1717 uses a generic release factor of  $10^{-3}$  for powders. The same factor  $(10^{-3})$  is used in NUREG-0706 as part of an empirical expression to represent the initial release of plutonium oxide from a falling drum and also is reported to be valid for  $U_3O_8$  (NRC 1980). This release factor  $(10^{-3})$  may be conservative because (1) the source materials are used in crystal form (i.e., not as a powder) for sublimation in a high temperature vacuum chamber and (2) the thin-film coatings, especially thorium, are durable and stable.

It is assumed that a small manufacturing facility uses 150 pounds/year of source material. Lenses are placed in a vacuum chamber with thorium fluoride or natural uranium, the chamber is evacuated, and the source material is vaporized and is deposited on surfaces within the chamber. Upon opening and cleaning of the chamber,  $10^{-3}$  of the deposited source material is assumed to be dispersed into the room as a respirable powder.

The total annual inhalation intake by a worker would be 0.0252 g (i.e., 150 lb × 454 g/lb × 0.001 ×  $[1/(180 \text{ m}^3) (18 \text{ hr}^{-1})] \times 1.2 \text{ m}^3/\text{hr})$ . This results in an inhalation of about 100 Bq of thorium and about 630 Bq of natural uranium. The committed inhalation dose coefficients from ICRP Publication 68 (ICRP 1994) were used to estimate the inhalation doses for thorium and natural uranium. The committed inhalation dose was estimated to be about 560 mrem for thorium and about 120 mrem for natural uranium. This is an upper bound, based on exposure of a single worker daily to the process. Using the same approach, doses were calculated for thorium, thorium progeny, uranium, and uranium short-lived progeny, with results shown as Table 11. Because of limited time between thorium and all progeny are considered for thorium.

The ingestion dose resulting from intake by a worker was also calculated. The EPA Exposure Factors Handbook offers a soil ingestion rate of 50 mg/day (EPA 1997). This value includes all soil, dust, and dirt consumed with foods. Beyond the NRC requirements, the scenario also is shaped by other industrial hygiene regulations, such as those of the U.S. Occupational Safety and Health Administration (29 CFR 1910). It is assumed that the manufacturer complies with the intent of 10 CFR 20.1201(e), which limits intake of soluble uranium to 10 mg/week to avoid the effects of heavy metal toxicity. Using this ingestion intake rate (10 mg/week) for source material results in a dose for thorium of 206 mrem/yr and uranium of 60 mrem/yr. Using the same approach, doses were calculated for thorium, thorium progeny, uranium, and uranium short-lived progeny, with results shown as Table 11. Because of limited time between thorium and uranium separation and subsequent use, only short-lived progeny are considered for uranium and all progeny are considered for thorium. A description of these calculations is provided in Appendix A.

#### 5.1.2.4 Additional Analyses Based on Manufacturer Survey

In an attempt to determine the level of conservativeness in the manufacturing analyses, several manufacturers were contacted to obtain additional information on the manufacturing environment associated with the application of uranium and thorium thin-film optical mirror and lens coatings. The questions largely centered around the manufacturing materials, processes, products, and environments related to production. Because of the limited response from manufacturers to the information inquiries the level of conservativeness of the scenario is not certain; however, some aspects of the scenario do appear to

be conservative. Personnel and workplace monitoring for radiation does not appear to be common; however, precautions are taken as employees reportedly are required to wear masks, gloves, and coats when handling thorium. In addition, thorium is reported to be stored with shielding and is handled in glove boxes or with enhanced ventilation. Cleaning, maintenance, material processing, and manufacture, are each reported to be less than full-time activities; however, for a small company a single individual may be expected to perform all of these tasks. Industry wide, the use of thorium thin-film coating is expected to increase because of rising demand and the lack of a suitable alternative.

According to the results reported in Table 11, the committed effective dose to unprotected workers during the manufacture of thorium thin-film optical coatings could approach about 0.8 rem, for the bounding case of a single worker processing up to 150 lb/yr of thorium with progeny. The survey, although with a limited response, revealed that manufacturers of thin-film optical coatings are using far less than 150 lb/yr uranium. If an assigned protection factor for respirator use of 10 can be credited (i.e. value for half facepiece, negative pressure air purifying respirator or a disposable negative pressure filtering facepiece, provided a fit factor of at least 100 is demonstrated by use of a validated or evaluated, qualitative or quantitative fit test) (10 CFR Part 20, Appendix A) and the annual use of uranium is limited to 15 lb, then the results in Table 11 would be reduced by a factor of 100 (i.e., the committed effective dose could approach about 8 mrem).

Table 11.	Radiation Dose Assessment	for Manufacture	of Thorium	and Uraniu	n Thin-Film	Optical
	Coatings					

Source	Annual Scenario Dose (mrem)				
	Ingestion	Inhalation	Effective <sup>†</sup>		
Thorium-Coated Optical Lens					
- Thorium Only	58	520	578		
- Thorium Progeny	148	42	190		
- Thorium + Progeny	206	562	768		
Uranium-Coated Optical Lens					
- Uranium Only	58	117	175		
- Uranium Short-Lived Progeny	2	0.2	2.2		
- Uranium + Short-Lived Progeny	60	117	177		
† Contribution from external dose considered negligible.					

### 5.1.3 Summary

The end use of thin-film optical coatings made with source materials appears to be fairly innocuous, with normal use doses around  $10^{-3}$  mrem/yr and accident committed effective doses around  $10^{-1}$  mrem. However, committed effective doses to unprotected workers during the manufacturing process could reach about 0.8 rem, for the bounding case of a single worker processing up to 150 lb/yr of source material. Additional information is needed on the design and operation of industrial sputtering processes to determine if the bounding exposure scenario is overly conservative.

#### 5.2 Drinking Water Filtration for Removal of Uranium

On December 7, 2000, the EPA promulgated new drinking water regulations that included an MCL for uranium of 30  $\mu$ g/L (65 FR 76708). The regulation requires that community water systems of all sizes be

in compliance by December 31, 2007. Filtering uranium is of potential concern to the NRC because several available remediation alternatives act to concentrate uranium. A number of these technologies can elevate uranium concentration above the exemption limit in 10 CFR 40.13(a) of 0.05 percent by weight and total uranium above the general license limit in 10 CFR 40.22 of 15 pounds. Because of their shared responsibility for protecting public health and the environment, the NRC has been in communication with the EPA about this regulation. On March 2, 2006, the NRC staff issued a policy recommendation (SECY-06-0049) to the Commission that included background information on the EPA drinking water regulations, methods for compliance, and disposal of uranium; along with alternatives for licensing or deferring licensing (NRC 2006). The NRC staff recommended developing a "new general license through rulemaking, applicable to drinking water treatment facilities that concentrate uranium in excess of 0.05 percent by weight" (NRC 2006).

The new regulation may affect a large number of community water systems because many areas of the United States produce groundwater with uranium concentrations in excess of 30  $\mu$ g/L. The EPA estimates that 795 community water systems will need to take some action to come in compliance with the new regulations (EPA 2001). One survey of domestic wells reported that 88 of 2390 wells surveyed (nearly 4 percent) exceeded the MCL for uranium of 30  $\mu$ g/L (Focazio et al. 2006), while other surveys reported exceedance rates much below 1 percent (Hess et al. 1985; Sloto 2000).

Uranium filtration technologies are widely reported (e.g., Boodoo 2005; CDPH 2001; NDWC 2000; NHDES 2004, 2005; NRC 2006), with an excellent summary provided by *A Regulators' Guide to the Management of Radioactive Residuals from Drinking Water Treatment Processes* (EPA 2005). The available technologies include anion exchange, reverse osmosis, lime softening, green sand filtration, co-precipitation with barium sulfate, electrodialysis/electrodialysis reversal, pre-formed hydrous manganese oxide filtration, activated alumina, and coagulation/filtration.

The radiation and chemical toxicity effects from acute exposure to uranium are well known, primarily from experiences of uranium processing workers associated with projects of the Manhattan Engineer District and the Atomic Energy Commission. Several studies attempt to address the health effects from chronic uranium exposure from drinking water (e.g., Svensson et al. 2005; Weir 2004; WDOH 1997; WHO 2004; Zamora et al. 1998); however, most of the reported results are not conclusive for what is a safe level of chronic exposure. Several of the studies report that increased chronic exposure to uranium is correlated with increased urinary excretion of alkaline phosphatase, calcium, and  $\beta_2$ -microglobulin—all associated with possible malfunction of the proximal tubule in the kidney. Svennson et al. (2005) concludes that these effects are observed with uranium concentrations in water of a few hundred µg/L and may exist at lower concentrations.

An abundance of experience with uranium filtration is reported for other applications and industries, including from *in-situ* leach uranium mining (e.g., Mackin et al. 2001; Marston 1998; NRC 1980, 1997, 2003; SRI International 1979), publically owned treatment works (POTWs) (e.g., Bastian et al. 2005; ISCORS 2005; Wolbarst et al. 2006), and uranium facilities (e.g., Rich et al. 1988).

#### 5.2.1 Description of Items

Dose assessments are performed for a large and small anion exchange resin column containing (1) 0.05 percent uranium by weight and (2) 15 pounds of uranium. The large anion exchange column is

assumed to be 20-ft tall and 15-ft diameter, the size reported for the drinking water filtration system in Oswego, Illinois (Hutchinson 2006). The small anion exchange column is assumed to be 5.2-ft high and 1-ft diameter. Both columns are modeled with a 5 mm steel wall thickness.

#### 5.2.2 Exemption Analyses: Scenario Description and Dose Assessments

It should be noted that some uranium filtration technologies are capable of removing additional radionuclides, along with the uranium. For example, lime softening and reverse osmosis remove both radium and uranium. In addition, mixed-bed filtration that combines cation and anion exchange will remove radium and uranium. Although not as efficient as cation exchange for radium removal, anion exchange is reported to remove radium with an efficiency of 35 to 60 percent, attributable to the presence of anionic radium compounds (STUK 2000). The dose assessments reported here are for anion exchange and only include results for uranium, uranium with short-lived progeny, and uranium with all progeny. A dose assessment for other filtration technologies would need to consider a variety of radionuclides in addition to uranium and its progeny. Along with uranium, anion exchange will filter other contaminants such as nitrates, sulfates, arsenic, chromates, molybdenum, selenium, and tannins (Boodoo 2005; Dow 2007b; O'Connor et al. 2005).

The EPA reports that the "...actual relationship between mass concentration (mg/L) and activity (pCi/L) varies somewhat in drinking water sources, since the relative amounts of the radioactive isotopes that make up naturally occurring uranium (<sup>238</sup>U, <sup>235</sup>U, and <sup>234</sup>U) vary between drinking water sources. The typical conversion factors that are observed in drinking water range from 0.67 up to 1.5 pCi/mg" (65 FR 76708). For these analyses, it is assumed the specific activity of natural uranium is  $6.77 \times 10^{-1} \,\mu$ Ci/g as reported in 10 CFR Part 20, Appendix B.

Of the available treatment technologies, strongly basic anion exchange resin provides one the highest filtration efficiencies for uranium at greater than 95 percent (Huikuri and Salonen 2000; STUK 2000; Vaaramaa et al. 2000). At low uranium concentrations typical for groundwater and surface water systems the uranium filtration efficiency is primarily dependent on the flow rate through the exchange column, with efficiencies near 95 percent with higher flow rates and approaching 100 percent with lower flow rates (Vaaramaa et al. 2000). Water quality does not appear to influence the uranium filtration efficiency at low uranium concentrations, but will affect the regeneration cycle time (Marston 1998; STUK 2000). Uranium filtration efficiencies approach 100 percent for sampling and airborne filtration applications, but these efficiencies are not yet attainable for large throughput water filtration applications such as those designed for use at POTWs (Rich et al. 1988).

#### 5.2.2.1 Routine Use – Resin with 15 Pounds Uranium

For routine use, the dose assessment is modeled as described in Section 5.2.2 using MicroShield Version 6.2 for large and small resin columns containing 15 pounds of uranium. Separate analyses were performed assuming the resin contains uranium only, uranium with short-lived progeny, and uranium with all progeny.

For the large column, assuming a resin density of 2.0 g/cm<sup>3</sup> and a resin volume of  $1.00 \times 10^8$  cm<sup>3</sup> (i.e., total resin mass of  $2.00 \times 10^8$  g), holding 15 pounds uranium would be 0.00341 percent uranium by weight

(i.e., does not exceed the 0.05 percent uranium by weight limit). With a 5-mm-thick steel wall and an exposure distance of 1 m at mid height, MicroShield Version 6.2 calculated an exposure rate of about 0.001 mrad/hr or an annual dose of about 0.25 mrem for a worker who is near the anionic exchange column for 2000 hr/yr.

A similar scenario was developed for a smaller anion exchange column (156-cm height and 30-cm diameter [i.e., 4 ft<sup>3</sup>]) holding 15 pounds uranium, maintaining previous 5-mm-thick steel wall and 1-m exposure distance at mid height. This geometry results in a uranium concentration of about 4 lb/ft<sup>3</sup>, which equates to about 0.28 mol/L or about 3.1 percent uranium by weight (i.e., does exceed the 0.05 percent uranium by weight limit). The capacity of the Rohm and Haas Amberjet 4400, an industrial-use anionic resin, is advertised to have a capacity of 1.4 eq/L (Rohm and Hass 2006). The Dow Chemical Company offers an anionic resin for potable water systems (DOWEX<sup>TM</sup> 1) that also has a capacity of 1.4 eq/L (Dow 2007a). Carbonate and sulfate complexes of uranium exist in the -4 state so the Amberjet 4400 and DOWEX<sup>TM</sup> 1 capacities for uranium become 0.35 mol/L or about 5 lb/ft<sup>3</sup>. There is some anecdotal evidence (Egidi et al. 2004) that the concentration of uranium in resin could reach 2 to 4 lb/ft<sup>3</sup> for drinking water systems. Under certain conditions (e.g., those maintained for an *in-situ* leach uranium mining operation) these anionic resins are capable of holding 10 lb/ft<sup>3</sup> or more of uranium (Mackin et al. 2001; Marston 1998). For this scenario, MicroShield Version 6.2 calculated an exposure rate of about 0.009 mrad/hr or an annual dose of about 20 mrem for a worker that is near the anionic exchange column for 2000 hr/yr.

Using the same approach, doses were calculated for uranium only, uranium with short-lived progeny, and uranium with all progeny, with results shown as Table 12. A description of these calculations is provided in Appendix A.

	Annual Scenar	io Dose (mrem)
Source	Large Column	Small Column
Resin Column Containing 15 Pounds Uranium		
- Uranium Only	0.064	5.68
- Uranium + Short-Lived Progeny	0.25	20
- Uranium + Progeny	1.3	1060
Resin Column Containing 0.05 wt% Uranium		
- Uranium Only	0.93	0.09
- Uranium + Short-Lived Progeny	3.7	0.32
- Uranium + Progeny	194	17

Table 12. Radiation Dose Assessment for Large and Small Anion Resin Exchange Columns Containing15 Pounds Uranium or 0.05 wt% Uranium.

#### 5.2.2.2 Routine Use – Resins at 0.05 Percent Uranium by Weight

For routine use, the dose assessment is modeled as described in Section 5.2.1 using MicroShield Version 6.2 for large and small resin columns containing 0.05 percent by weight uranium. Separate analyses were performed assuming the resin contains uranium only, uranium with short-lived progeny, and

uranium with all progeny.

For the large column, assuming a resin density of 2.0 g/cm<sup>3</sup> and a resin volume of  $1.00 \times 10^8$  cm<sup>3</sup>, (i.e., total resin mass of  $2.00 \times 10^8$  g), holding 0.05 percent by weight uranium would be about 220 pounds of uranium. This amount of uranium (220 pounds) is about 1 ounce/ft<sup>3</sup>, much less than anecdotal 4 lb/ft<sup>3</sup>. With a 5-mm-thick steel wall and an exposure distance of 1 m, MicroShield Version 6.2 calculated an exposure rate of about 0.002 mrad/hr or an annual dose of about 4 mrem for a worker that is near the anionic exchange column for 2000 hr/yr.

For the small column, assuming a resin density of 2.0 g/cm<sup>3</sup> and a resin volume of about  $1.10 \times 10^5$  cm<sup>3</sup>, (i.e., total resin mass of  $2.20 \times 10^5$  g), holding 0.05 percent by weight uranium would be about 0.24 pounds of uranium. With a 5-mm-thick steel wall and an exposure distance of 1 m, MicroShield Version 6.2 calculated an exposure rate of about 0.00016 mrad/hr or an annual dose of about 0.3 mrem for a worker that is near the anionic exchange column for 2000 hr/yr.

Using the same approach, doses were calculated for uranium only, uranium with short-lived progeny, and uranium with all progeny, with results shown as Table 12. A description of these calculations is provided in Appendix A.

#### 5.2.3 Summary

Analyses showed that for a worst case scenario, filtration of water to remove uranium could lead to doses to workers near 1.1 rem/yr for a small resin column (4 ft<sup>3</sup>) containing up to 15 pounds of uranium with progeny. Calculations indicate that assuming a uranium loading this high (i.e., 4 lb/ft<sup>3</sup>) is reasonable. Assuming on average a half-full anionic exchange column (i.e., the column contains no uranium at the beginning of its duty cycle, with uranium concentration increasing to a maximum at the end of its duty cycle) gives 0.27 mrad/hr or about 550 mrem/working year. Analyses for natural uranium (not accounting for radium or other progeny) gives a fairly low dose rate of less than 6 mrem/yr, assuming a fully loaded anionic exchange column and an exposure time of a full working year (2000 hr/yr). Use of specific filtering technologies that also would filter other radionuclides (e.g., radium), the presence of anionic radium compounds in source waters, or the use of a long duty cycle that would permit buildup of radon decay products, each may require review in greater detail to ensure adequate dose protection. Additional precautions, such as limiting access (e.g., not locating occupied work areas near the anionic exchange columns) or adding some shielding (e.g., concrete wall), would reduce the already low external dose rates to insignificant levels. Consideration of these uncertainties and selective use of precautions, such as those described here, would likely be sufficient to provide adequate safety.

#### 5.3 Source Material Bounding Dose Assessments

Realistic, bounding scenarios were developed and evaluated for use of source material under 10 CFR 40.13(a), 40.13(b), and 40.22. These scenarios are intended to bound doses to workers and members of the public from exposure to source material under "reasonable" conditions of use. A description of analyses and results follow, organized by regulatory area; 10 CFR 40.13(a), 40.13(b), and 40.22.

#### 5.3.1 10 CFR 40.22 Bounding Dose Assessment

A bounding assessment for the use of source material under 10 CFR 40.22 was developed for an individual that uses 150 pounds of uranium or thorium powder in a small room. This was considered to be an operation such as a commercial public pottery facility in which an owner/employee works full time preparing pottery, mixing pigments, and firing customer creations. The worker regularly mixes powdered uranium/thorium pigments, applies glazes, and cleans up after customers. This is a revision of the Section 5.1.2.3 occupational scenario, with a smaller room and lower ventilation rate. It is still a dusty work environment. Cleanliness standards are lower than in the industrial operation (i.e., a small business, in which the dusty operations occur in a small room away from most customers). Studies related to art students (Stopford 2003) indicate that activities that generate dust include:

- ceramic glaze spraying
- slip decorating with splattering
- clay and slip cleanup
- clay sanding
- clay vacuuming.

Note that no powder pouring was evaluated. Respirable fractions of the dust thus generated were relatively low, from 3 to 4 percent. This study also measured dust in a community ceramic studio where dust levels ranged 130 to  $370 \ \mu g/m^3$ .

The U.S. Department of Energy publication, *Airborne Release Fractions/Rates and Respirable Fractions* for Nonreactor Nuclear Facilities, DOE-HDBK-3010-94 (DOE 1994), indicates a median release fraction of  $3 \times 10^{-4}$  with a respirable fraction of 0.5 for spills of powders (and a bounding release fraction of  $2 \times 10^{-3}$  with a respirable fraction of 0.3); adding other physical activities may raise the average total release fraction to near 0.001 (i.e., the same as the industrial value).

Studies of bakery workers indicate flour dust on the order of 2 to 5 mg/m<sup>3</sup> is common; however, flour has about a 0.2 respirable fraction (Lidén et al. 1998). For these analyses it is assumed that a 20 percent respirable fraction is appropriate for U/Th pigment powders as well.

Assuming a 30 m<sup>3</sup> room, with 0.5 air exchange/hour (the median national residential rate as determined by Murray and Burmaster [1995]), accounting for settling rate using approach used in Section 5.1.2.3 gives a total loss rate of 12.5 hr<sup>-1</sup>. The settling loss rate component (12 hr<sup>-1</sup>) dominates over the room ventilation loss rate (0.5 hr<sup>-1</sup>).

The inhalation intake will then be 150 lb × 454 g/lb ×  $10^{-3}$  release fraction ×  $[1/(30 \times 12.5)]hr/m^3 \times 0.9 m^3/hr = 0.16$  grams/year (650 Bq Th or 4000 Bq U). The committed inhalation dose coefficients from ICRP Publication 68 (ICRP 1994) were used to estimate the inhalation doses for thorium and natural uranium. The committed inhalation dose was estimated to be about 4 rem for thorium and about 0.7 rem for natural uranium. This is an upper bound, based on exposure of a single worker daily to the process. Using the same approach, doses were calculated for thorium, thorium progeny, uranium, and uranium short-lived progeny, with results shown as Table 13. Because of limited time between thorium and

uranium separation and subsequent use, only short-lived progeny are considered for uranium and all progeny are considered for thorium.

The ingestion dose resulting from intake by the owner/employee was also calculated. The EPA *Exposure Factors Handbook* offers a soil ingestion rate of 50 mg/day (EPA 1997). It is assumed that the ceramics shop owner/employee spends the equivalent of 8 hours/week (1 working day per week) mixing pigments, cleaning up, and vacuuming. Scaling the EPA *Exposure Factors Handbook* value for ingestion of 50 mg/day from surfaces, gives 50 mg/wk (2.5 g/yr). This ingestion intake rate (50 mg/week) results in a dose for thorium of about 1 rem/yr and uranium of 300 mrem/yr. Using the same approach, doses were calculated for thorium, thorium progeny, uranium, and uranium short-lived progeny, with results shown in Table 13. Because of limited time between thorium and uranium separation and subsequent use, only short-lived progeny are considered for uranium and all progeny are considered for thorium. A description of these calculations is provided in Appendix A.

Table 13. Radiation Dose Assessment for 10 CFR 40.22 Bounding Scenario

	Annual Scenario Dose (mrem)				
Source	Ingestion	Inhalation	Effective <sup>†</sup>		
Handling 150 lb/yr of Thorium Powder					
- Thorium Only	290	3380	3670		
- Thorium Progeny	740	272	1012		
- Thorium + Progeny	1030	3650	4680		
Handling 150 lb/yr of Uranium Powder					
- Uranium Only	290	740	1030		
- Uranium Short-Lived Progeny	10	1	11		
- Uranium + Short-Lived Progeny	300	740	1040		

<sup>†</sup> Contribution from external dose considered negligible.

#### 5.3.2 10 CFR 40.13(a) Bounding Dose Assessment

10 CFR 40.13(a) exempts from regulation the receipt, possession, use, transfer, or delivery of source material (uranium and thorium) in any chemical mixture, compound, solution, or alloy in which the source material is by weight less than 0.05 percent of the mixture, compound, solution, or alloy. This exemption allows one to possess unlimited quantities of source material as long as the source material content is less than 0.05 weight percent of the total material mass.

Examples of this category of material were identified, characterized, and evaluated in Section 3.2 of NUREG-1717 (NRC 2001) and included coal fly ash, water treatment sludges, metal mining and processing wastes/tailings, dental products, and eyeglasses. The general conclusions that the presence of thorium and uranium in consumer products is declining or is no longer used remains valid. This is particularly true of uranium in dental products and eyewear. As for waste products containing source material, pressure remains high to find alternative disposal methods, including reuse, because of limitations on acceptance at landfills and rising costs of disposal. The use of coal combustion products in

road construction (e.g., as a replacement for Portland cement in concrete applications and as structural fill) remains common in several States including Wisconsin that enacted legislation to streamline the reuse of industrial byproducts (WAC NR 538).

For this evaluation, emphasis was placed on characterizing those materials in which the source material content equals or exceeds the exemption limit of 0.05 weight percent, although information on material containing a lower source material content was reviewed for completeness.

A bounding assessment for the use of source material under 10 CFR 40.13(a) was developed for an individual who works in an environment with airborne source material that is 0.05 percent uranium or thorium by weight. For this analysis, it is assumed that airborne source material (uranium and thorium) is maintained at the threshold limit value for nuisance dust, 10 mg/m<sup>3</sup> (ACGIH 2001). At this level, it is reasonable to assume that no respiratory protection would be used. Because a given airborne concentration is assumed (i.e., 10 mg/m<sup>3</sup>), no credit is given for ventilation rate, and the person could be assumed to be working indoors or outdoors in these conditions. A breathing rate of 1.2 m<sup>3</sup>/hr is assumed. Given a 10 mg/m<sup>3</sup> loading rate, 1.2 m<sup>3</sup>/hr breathing rate, 0.05 percent by weight uranium or thorium, and 2000 hr exposure time, the resulting total annual inhalation intake is 12 mg of either uranium or thorium. This results in an inhalation of about 49 Bq of thorium or about 320 Bq of natural uranium. The committed inhalation dose coefficients from ICRP Publication 68 (ICRP 1994) were used to estimate the inhalation doses for thorium, thorium progeny, uranium, and uranium short-lived progeny, with results shown as Table 14. The committed inhalation dose was estimated to be about 275 mrem for thorium and about 59 mrem for natural uranium. A description of these calculations is provided in Appendix A.

Source	Annual Scenario Dose (mrem)
10 CFR 40.13(a) Bounding	Inhalation
Thorium Nuisance Dust	
- Thorium	255
- Thorium Progeny	20
- Thorium + Progeny	275
Uranium Nuisance Dust	
- Uranium	59
- Uranium Short-Lived Progeny	0.08
- Uranium + Short-Lived Progeny	59

Table 14. Radiation Dose Assessment for 10 CFR 40.13(a) Bounding Scenario

#### 5.3.3 10 CFR 40.13(b) Bounding Dose Assessment

10 CFR 40.13(b) exempts any person from the regulations in 10 CFR Part 40 for the receipt, possession, use, or transfer of unrefined and unprocessed ore containing source material; provided, that except as authorized in a specific license, the ore is not refined or processed. This exemption allows one to possess unlimited quantities of unrefined and unprocessed ore containing source material (uranium or thorium).

Examples of this category of material were identified, characterized, and evaluated in Section 3.3 of NUREG-1717 (NRC 2001) and included mined ores, ores collected by hobbyists and collectors, and ore used as construction materials. The NUREG-1717 scenario for amateur geologist assumed possession of 100 uranium ore samples (1 percent weight uranium in equilibrium with its short-lived progeny) and 100 monazite samples (0.03 percent weight thorium). At a distance of 1.8 m and with an exposure time of 2000 hr/yr, NUREG-1717 reported an annual effective dose equivalent of 0.4 mrem for uranium ore and 0.9 mrem for thorium ore. It was noted, for example, that United Nuclear (http://www.unitednuclear.com) and other suppliers sell unrefined and unprocessed source material products to public, industry, commercial, and research and development organizations. These source material products include thorium ore and uranium ores (e.g., torbernite, carnotite, uraninite, gummite, pitchblende, and uranophane) advertised up to 500,000 counts per minute for combined beta and gamma radiation measured at 1 in. from the sample. These higher-activity ores are evaluated here for an amateur geologist that possesses such a sample.

A typical uranium ore contains 0.14 percent  $U_3O_8$  (DOE 2006). However, some high-grade ores such as pitchblende (an amorphous, black, pitchy form of the crystalline uranium oxide mineral uraninite that is one of the primary mineral ores of uranium) can contain up to 50 to 80 percent uranium (Encyclopædia Britannica 2006). It is reported that African-origin pitchblende and torbernite processed during the 1940s at the Linde Ceramics Plant as part of a Manhattan Engineer District project contained 3 to 20 percent  $U_3O_8$  by weight with a density of 3.25 g/cm<sup>3</sup> (ORAU et al. 2006). It is not unreasonable to assume that these high-activity ore samples would be desirable to a hobbyist or collector.

For this bounding analysis, it is assumed that a hobbyist obtains and displays a 1-kg piece of pitchblende ore. The ore is assumed to contain 500 grams of <sup>238</sup>U (approximately 50 percent  $U_3O_8$  by weight), with all associated isotopes of uranium in equilibrium with their decay progeny. Additionally, an assessment was performed for uranium only and uranium with short-lived progeny. Note that similar analyses reported in NUREG-1717 accounted only for short-lived progeny, so it did not include significant external dose contribution from <sup>226</sup>Ra. Uranium ore that has not undergone chemical processing will contain all members of the uranium decay series, including radium and its progeny (ORAU et al. 2006). The activities (assuming secular equilibrium) of the various radionuclides in the ore sample (reported in curies) are provided as Table 15.

The ore sample is assumed to be a sphere of density about 5 g/cm<sup>3</sup>, with a diameter of 6 cm. The dose rate for uranium with all progeny calculated using MicroShield Version 6.2 is about 0.5 mrem/hr at a distance of one meter. If the hobbyist keeps the sample on display on his desk, at which he sits 2000 hr/yr, the dose would be approximately 1.1 rem/yr. Results for uranium only, uranium with short-lived progeny, and uranium with all progeny is shown as Table 16. A description of these calculations is provided in Appendix A.

Radionuclide	Activity (Ci)
<sup>238</sup> U	$1.67  imes 10^{-4}$
<sup>234</sup> Th	$1.67 \times 10^{-4}$
<sup>234m</sup> Pa	$1.67 \times 10^{-4}$
<sup>234</sup> Pa	$2.16 \times 10^{-7}$
<sup>234</sup> U	$1.67 \times 10^{-4}$
<sup>230</sup> Th	$1.67 \times 10^{-4}$
<sup>226</sup> Ra	$1.67 \times 10^{-4}$
<sup>222</sup> Rn	$1.67 \times 10^{-4}$
<sup>218</sup> Po	$1.67 \times 10^{-4}$
<sup>214</sup> Pb	$1.66 \times 10^{-4}$
<sup>214</sup> Bi	$1.67 \times 10^{-4}$
<sup>214</sup> Po	$1.66 \times 10^{-4}$
<sup>210</sup> T1	$3.33 \times 10^{-8}$
<sup>210</sup> Pb	$1.67 \times 10^{-4}$
<sup>210</sup> Bi	$1.67 \times 10^{-4}$
<sup>210</sup> Po	$1.67 \times 10^{-4}$
<sup>208</sup> T1	$2.16 \times 10^{-10}$
<sup>235</sup> U	$7.49 \times 10^{-6}$
<sup>231</sup> Th	$7.49 \times 10^{-6}$
<sup>231</sup> Pa	$7.49 \times 10^{-6}$
<sup>227</sup> Ac	$7.49 \times 10^{-6}$
<sup>227</sup> Th	$7.39 \times 10^{-6}$
<sup>223</sup> Fr	$1.05 \times 10^{-7}$
<sup>223</sup> Ra	$7.49 \times 10^{-6}$
<sup>219</sup> Rn	$7.49 \times 10^{-6}$
<sup>215</sup> Po	$7.49 \times 10^{-6}$
<sup>211</sup> Pb	$7.49 \times 10^{-6}$
<sup>211</sup> Bi	$7.49 \times 10^{-6}$
<sup>211</sup> Po	$2.10 \times 10^{-8}$
<sup>207</sup> Tl	$7.47 \times 10^{-6}$

Table 15. Activities of Various Radionuclides for the Hobbyist Ore Assessment

 Table 16.
 Radiation Dose Assessment for High-Grade Uranium Ore Sample

Source	Annual Scenario Dose (mrem)			
High-Grade Uranium Ore Sample				
- Uranium Only	5.7			
- Uranium + Short-Lived Progeny	20			
- Uranium + Progeny	1060			

# 6 Insights, Conclusions, and Recommendations

The primary objective of this project was to identify new uses of source material permitted under 10 CFR 40.13(a), 40.13(b), and 40.22, and to assess the potential individual radiation dose to workers and members of the public from identified new 10 CFR 40.22 uses. New uses identified and assessed were:

- thorium and uranium thin-film optical coatings
- filtration of uranium from drinking water at municipal water treatment plants.

A second objective was to develop and assess bounding but realistic scenarios for public exposure to source materials permitted under 10 CFR 40.13(a), 40.13(b), and 40.22. These scenarios are intended to bound any potential doses to workers and members of the public from exposure to source material under "reasonable" conditions of use.

The results of these radiation dose assessments are summarized in Table 17. The results suggest the following:

- Uranium and thorium-coated optical lenses yield minimal dose under routine use and accident conditions. However, the unprotected occupational worker exposed to thorium and uranium powders during the sputtering process can potentially receive an annual internal radiation dose up to 560 mrem. Additional information is needed on the design and operation of industrial sputtering processes to determine if this bounding exposure scenario is overly conservative.
- Filtration or ion exchange of uranium from drinking water by municipal water treatment plants pose little health risk from the uranium being removed from the drinking water. Substantially greater health risk is potentially posed to the occupational worker from long-lived uranium progeny or other radionuclides that may be removed during filtration by the water treatment system.
- As might be expected, the dose results for each of the bounding scenarios are not inconsequential. Those results need to be considered in light of the fact that the scenarios were developed with the specific intent of being conservatively bounding, but realistic.

However, as with any analysis, assumptions were made that could be refined with additional information and which could potentially impact these conclusions. Specifically, the occupational exposure scenario for the manufacture of uranium and thorium-coated lenses would benefit from a more detailed review and understanding of the manufacturing processes for these materials. Also, a more comprehensive understanding of how municipal water treatment plants are implementing the requirement to remove uranium from drinking water would improve the analysis of this scenario.

Scenario	Annual Scenario Dose (mrem/yr)		nrem/yr)
Routine Use	Photographer	TV Camera	
Thorium-Coated Optical Lens		Operator	
- Thorium Only	$2.9 \times 10^{-5}$	$2.9 \times 10^{-4}$	
- Thorium Progeny	$3.7 \times 10^{-4}$	$3.7 \times 10^{-3}$	
- Thorium + Progeny	$4.0 \times 10^{-4}$	$4.0 \times 10^{-3}$	
Uranium-Coated Optical Lens			
- Uranium Only	$2.4 \times 10^{-7}$	$2.4 \times 10^{-6}$	
- Uranium Short-Lived Progeny	$3.1 \times 10^{-6}$	$3.1 \times 10^{-5}$	
- Uranium + Short-Lived Progeny	$3.3 \times 10^{-6}$	$3.3 \times 10^{-5}$	
Accident	Ingestion	Inhalation	Effective <sup>†</sup>
Thorium-Coated Optical Lens			
- Thorium Only	$2.3 \times 10^{-4}$	$5.9 \times 10^{-2}$	$5.9 \times 10^{-2}$
- Thorium Progeny	$5.9 \times 10^{-4}$	$4.5 \times 10^{-3}$	$5.1 \times 10^{-3}$
- Thorium + Progeny	$8.2 \times 10^{-4}$	$6.4 \times 10^{-2}$	$6.5 \times 10^{-2}$
Uranium-Coated Optical Lens			
- Uranium Only	$4.7 \times 10^{-6}$	$3.2 \times 10^{-4}$	$3.2 \times 10^{-4}$
- Uranium Short-Lived Progeny	$1.7 \times 10^{-7}$	$3.2 \times 10^{-7}$	$4.9 \times 10^{-7}$
- Uranium + Short-Lived Progeny	$4.9 \times 10^{-6}$	$3.2 \times 10^{-4}$	$3.2 \times 10^{-4}$
Manufacturing	Ingestion	Inhalation	Effective <sup>†</sup>
Thorium-Coated Optical Lens	_		
- Thorium Only	58	520	578
- Thorium Progeny	148	42	190
- Thorium + Progeny	206	562	768
Uranium-Coated Optical Lens			
- Uranium Only	58	117	175
- Uranium Short-Lived Progeny	2	0.2	2.2
- Uranium + Short-Lived Progeny	60	117	177
Uranium Filtration	Large Column	Small Column	
Resin Column Containing 15 Pounds Uranium	-		
- Uranium Only	0.064	5.68	
- Uranium + Short-Lived Progeny	0.25	20	
- Uranium + Progeny	1.3	1060	
Resin Column Containing 0.05 wt% Uranium			
- Uranium Only	0.93	0.09	
- Uranium + Short-Lived Progeny	3.7	0.32	
- Uranium + Progeny	194	17	
10 CFR 40.22 Bounding	Ingestion	Inhalation	Effective †
Handling 150 lb/yr of Thorium Powder	5		
- Thorium Only	290	3380	3670
- Thorium Progeny	740	272	1012
- Thorium + Progeny	1030	3650	4680
Handling 150 lb/yr of Uranium Powder			
- Uranium Only	290	740	1030
- Uranium Short-Lived Progeny	10	1	11
- Uranium + Short-Lived Progeny	300	740	1040

# Table 17. Summary of Radiation Dose Assessment

Scenario	Annual Scenario Dose (mrem/yr)
10 CFR 40.13(a) Bounding	Inhalation
Thorium Nuisance Dust	
- Thorium	255
- Thorium Progeny	20
- Thorium + Progeny	275
Uranium Nuisance Dust	
- Uranium	59
- Uranium Short-Lived Progeny	0.08
- Uranium + Short-Lived Progeny	59
10 CFR 40.13(b) Bounding	External
High-Grade Uranium Ore Sample	
- Uranium Only	5.7
- Uranium + Short-Lived Progeny	20
- Uranium + Progeny	1060
† Contribution from external dose considered negligible.	

Table 17. (contd.)

# 7 References

10 CFR Part 19. Code of Federal Regulations, Title 10, *Energy*, Part 19, "Notices, Instructions, and Reports to Workers: Inspection and Investigations."

10 CFR Part 20. Code of Federal Regulations, Title 10, *Energy*, Part 20, "Standards for Protection Against Radiation."

10 CFR Part 21. Code of Federal Regulations, Title 10, *Energy*, Part 21, "Reporting of Defects and Noncompliance."

10 CFR Part 40. Code of Federal Regulations, Title 10, *Energy*, Part 40, "Domestic Licensing of Source Material."

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Appendix A

### **Description of Calculations**

The following are descriptions of calculations used to generate the results presented in this report. The description titles used below are presented in order of appearance in the report and reference their associated report section number in parentheses. Descriptions are provided for analyses of thorium and uranium thin-film optical coatings (routine use, accidents, and manufacturing), drinking water filtration for removal of uranium (resin with 15 lb uranium and resin with 0.05 percent uranium by weight), and source material bounding dose assessments for 10 CFR 40.22, 40.13(a), and 40.13(b).

### Radiation Dose Assessment for Routine Use of Thorium and Uranium Thin-Film Optical Coatings (Section 5.1.2.1, Routine Use)

The lenses evaluated in NUREG-1717 (NRC 2001) had incorporated up to 100 g thorium, while thin-film coated lenses have only about 0.02 g thorium or 0.0004 g uranium. The routine use scenario only has an external dose component. Because of limited time between thorium and uranium separation and subsequent use, only short-lived progeny are considered for uranium and all progeny are considered for thorium.

#### Thorium

Routine doses from total thorium (i.e., with all progeny in equilibrium) would be about 0.0004 mrem/yr to an individual photographer and about 0.004 mrem/yr to a television camera operator (i.e., 0.02 percent of the values reported in NUREG-1717, Section 3.19.4, for thoriated lenses). The 0.02 percent scaling approximation is based on the difference in thorium content between the NUREG-1717 scenario (100 g thorium) and this scenario (0.02 g thorium). The separate dose contributions from thorium and thorium progeny were determined by first calculating the percent contribution to dose from thorium (7.14 percent) and thorium progeny (92.86 percent), as shown in Table A-1. Results for routine use of thorium coated lenses are then calculated as follows:

Individual Photo	ographer (NU	JREG-1717 Section $3.19.4.1.2 = 2 \text{ mrem/yr for } 100 \text{ g thorium}$
Thorium On	ly	2 mrem/yr × 0.0002 × 0.0714 = $2.9 \times 10^{-5}$ mrem/yr
Thorium Pro	geny	$2 \text{ mrem/yr} \times 0.0002 \times 0.9286 = 3.7 \times 10^{-4} \text{ mrem/yr}$
Thorium + P	rogeny	0.0004 mrem/yr
TV Camera ope	rator ((NUR	EG-1717 Section 3.19.4.1.1 = 20 mrem/yr for 100g thorium)
Thorium On	ly	$20 \text{ mrem/yr} \times 0.0002 \times 0.0714 = 2.9 \times 10^{-4} \text{ mrem/yr}$
Thorium Pro	geny	$20 \text{ mrem/yr} \times 0.0002 \times 0.9286 = 3.7 \times 10^{-3} \text{ mrem/yr}$
Thorium + P	rogeny	0.004 mrem/yr

#### Uranium

Doses from thin-film uranium coatings were estimated using the ratio of gamma constants (Unger and Trubey 1982) for total thorium  $(2.07 \times 10^{-6} \text{ rem/}\mu\text{Ci-hr})$  and total uranium  $(1.32 \times 10^{-7} \text{ rem/}\mu\text{Ci-hr})$  and the ratio of activities for the example thorium and uranium lens coatings (i.e., 77 Bq for thorium and 10 Bq for uranium). The gamma constant for total thorium  $(2.07 \times 10^{-6} \text{ rem/}\mu\text{Ci-hr})$  is taken from Table A-1 as the sum of dose contribution in units of rem/ $\mu\text{Ci-hr}$ . Similarly, the gamma constant for total uranium  $(1.32 \times 10^{-7} \text{ rem/}\mu\text{Ci-hr})$  is taken from Table A-2 as the sum of dose contribution in units of rem/ $\mu\text{Ci-hr}$ . This approach yielded routine doses from total uranium of about  $3.3 \times 10^{-6} \text{ mrem/yr}$  to an individual photographer and about  $3.3 \times 10^{-5} \text{ mrem/yr}$  to a television camera operator, about two orders of magnitude lower than those for thin-film thorium optical coatings. The separate dose contributions from uranium and uranium short-lived progeny were determined by first calculating the percent contribution to dose from

uranium (58.80 percent) and uranium short-lived progeny (41.20 percent), as shown in Table A-2. Results for routine use of uranium coated lenses are then calculated as follows:

Individual Photographer (ra Uranium Only Uranium Progeny	tio to thorium of gamma constants [rem/ $\mu$ Ci-hr] and inventory [Bq]) 3.31 × 10 <sup>-6</sup> mrem/yr × 0.5880 = 1.95 × 10 <sup>-6</sup> mrem/yr 3.31 × 10 <sup>-6</sup> mrem/yr × 0.4120 = 1.36 × 10 <sup>-6</sup> mrem/yr
Uranium + Progeny	$0.0004 \text{ mrem/yr} \times (10 / 77) \times (1.32 \times 10^{-7} / 2.07 \times 10^{-6}) = 3.31 \times 10^{-6} \text{ mrem/yr}$
TV Camera operator (ratio t	o thorium of gamma constants [rem/µCi-hr] and inventory [Bq])
Uranium Only	$3.31 \times 10^{-5} \text{ mrem/yr} \times 0.5880 = 1.95 \times 10^{-5} \text{ mrem/yr}$
Uranium Progeny	$3.31 \times 10^{-5}$ mrem/yr $\times 0.4120 = 1.36 \times 10^{-5}$ mrem/yr
Uranium + Progeny	$0.004 \text{ mrem/yr} \times (10 / 77) \times (1.32 \times 10^{-7} / 2.07 \times 10^{-6}) = 3.31 \times 10^{-5} \text{ mrem/yr}$

**Table A-1.** External Dose Contributions for Thorium and Progeny.

Nuclide	Yield	Specific Gamma-Ray Dose Constant (mSv/MBq-hr) <sup>†</sup>	Specific Gamma-Ray Dose Constant (rem/µCi-hr)	Dose Contribution (rem/µCi-hr) <sup>‡</sup>	Dose Contribution (percent)
Th-232	1	1.848E-05	6.838E-08	6.838E-08	3.307E-02
Ra-228	1	0.000E+00	0.000E + 00	0.000E + 00	0.000E+00
Ac-228	1	2.278E-04	8.429E-07	8.429E-07	4.077E - 01
Th-228	1	2.142E-05	7.925E-08	7.925E-08	3.834E-02
Ra-224	1	2.967E-06	1.098E - 08	1.098E-08	5.310E-03
Rn-220	1	9.607E-08	3.555E-10	3.555E-10	1.719E-04
Po-216	1	2.421E-09	8.958E-12	8.958E-12	4.333E-06
Pb-212	1	7.389E-05	2.734 E - 07	2.734E-07	1.322E-01
Bi-212	1	5.252E-05	1.943 E - 07	1.943E-07	9.400E-02
Po-212	0.64	0.000E+00	0.000E + 00	0.000E+00	0.000E+00
T1-208	0.3593	4.497E-04	1.664E-06	5.978E-07	2.892E-01

† (Unger and Trubey 1982)

‡ Dose contribution is product of yield and specific gamma-ray dose constant.

**Table A-2.** External Dose Contributions for Uranium and Short-Lived Progeny.

Nuclide	Percent Activity <sup>†</sup>	Specific Gamma-Ray Dose Constant (mSv/MBq-hr) <sup>‡</sup>	Specific Gamma-Ray Dose Constant (rem/µCi-hr)	Dose Contribution (rem/µCi-hr) *	Dose Contribution (percent)
U-238	0.4872	1.763E-05	6.523E-08	3.178E-08	2.407E-01
Th-234	0.4872	2.038E-05	7.541E-08	3.674E-08	2.783E-01
Pa-234m	0.4872	2.776E-06	1.027 E - 08	5.004E-09	3.791E-02
U-234	0.4896	2.097E-05	7.759E-08	3.799E-08	2.877E-01
U-235	0.0232	9.159E-05	3.389E-07	7.862E-09	5.955E-02
Th-231	0.0232	1.473 E - 04	5.450E-07	1.264E-08	9.578E-02

† (Voss 2001)

‡ (Unger and Trubey 1982)

\* Dose contribution is product of yield and specific gamma-ray dose constant.

## Radiation Dose Assessment for Accidents Involving Thorium and Uranium Thin-Film Optical Coatings (Section 5.1.2.2, Accidents)

The generic accident methodology developed in NUREG-1717, Appendix A.2, was found to be reasonable and has been adapted for exposures to thorium and uranium coated lenses. It is assumed, first, that 10 percent of the available material would be deposited somewhere on the skin of an individual and, second, that 0.1 percent of this deposited material would be ingested before bathing removed the material from the body. Thus, the direct ingestion intake is estimated to be  $1 \times 10^{-4}$  times the total amount of available material on the lens. The same value  $(1 \times 10^{-4})$  is adopted for inhalation. Because of limited time between thorium and uranium separation and subsequent use, only short-lived progeny are considered for uranium and all progeny are considered for thorium.

#### Thorium

Using the above assumption (i.e., inhalation and ingestion intake fraction of  $1 \times 10^{-4}$ ) results in an ingestion and inhalation of about 0.0077 Bq of thorium for each pathway. The committed dose coefficients from ICRP Publication 72 (ICRP 1996) were used to estimate the inhalation and ingestion doses for thorium, thorium progeny, and thorium with progeny, as shown in Table A-3. Results for accidents involving thorium coated lenses are then calculated as follows:

Accidents (10 percent of thorium on skin, 0.1 percent of that ingested and inhaled)

#### Uranium

Using the above assumption (i.e., inhalation and ingestion intake fraction of  $1 \times 10^{-4}$ ) results in an ingestion and inhalation of about 0.001 Bq of natural uranium for each pathway. The committed dose coefficients from ICRP Publication 72 (ICRP 1996) were used to estimate the inhalation and ingestion doses for natural uranium (activity weighted average for <sup>234</sup>U, <sup>235</sup>U, and <sup>238</sup>U), uranium short-lived progeny, and uranium with short-lived progeny, as shown in Table A-4. Results for accidents involving uranium coated lenses are then calculated as follows:

Accidents (10 percent of uranium on skin, 0.1 percent of that ingested and inhaled)

Ingestion	
Uranium Only	$10 \text{ Bq} \times 0.0001 \times (4.70 \times 10^{-8} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 4.7e-6 \text{ mrem}$
Uranium Progeny	$10 \text{ Bq} \times 0.0001 \times (1.67 \times 10^{-9} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 1.7\text{e}-7 \text{ mrem}$
Uranium + Progeny	$(4.7 \times 10^{-6} \text{ mrem}) + (1.7 \times 10^{-7} \text{ mrem}) = 4.9 \times 10^{-6} \text{ mrem}$
Inhalation	
Uranium Only	$10 \text{ Bq} \times 0.0001 \times (3.20 \times 10^{-6} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 3.2 \times 10^{-4} \text{ mrem}$
Uranium Progeny	$10 \text{ Bq} \times 0.0001 \times (3.23 \times 10^{-9} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 3.2 \times 10^{-7} \text{ mrem}$
Uranium + Progeny	$(3.2 \times 10^{-4} \text{ mrem}) + (3.2 \times 10^{-7} \text{ mrem}) = 3.2 \times 10^{-4} \text{ mrem}$

Nuclide	Inhalation Dose Coefficient (Sv/Bq)	Ingestion Dose Coefficient (Sv/Bq)
Th-232	4.50E-05	2.30E-07
Ra-228	2.60E-06	6.90E-07
Ac-228	1.70E - 08	4.30E-10
Th-228	3.20E-05	7.20E-08
Ra-224	3.00E-06	6.50E-08
Pb-212	1.70E - 07	6.00E-09
Bi-212	3.10E-08	2.60E-10

Table A-3. Thorium Inhalation and Ingestion Dose Coefficients from ICRP Publication 72 (ICRP 1996).

Table A-4. Uranium Inhalation and Ingestion Dose Coefficients from ICRP Publication 72 (ICRP 1996).

Nuclide	Inhalation Dose Coefficient (Sv/Bq)	Ingestion Dose Coefficient (Sv/Bq)
U-238	2.90E-06	4.50E-08
Th-234	6.60E-09	3.40E-09
Pa-234m	0.00E+00	0.00E+00
U-234	3.50E-06	4.90E-08
U-235	3.10E-06	4.70E-08
Th-231	3.10E-10	3.40E-10

## Radiation Dose Assessment for Manufacture of Thorium and Uranium Thin-Film Optical Coatings (Section 5.1.2.3, Manufacturing)

The approach used in NUREG-1717 for estimating releases into a laboratory-type room was found to be reasonable and applicable for approximating releases in a small manufacturing facility. The scenario assumes resuspension of a fraction of the inventory in a laboratory-type room with an enclosure volume of 180 m<sup>3</sup>, a room height of 3 m, a ventilation rate of six volume changes per hour, and a breathing rate of  $1.2 \text{ m}^3/\text{hr}$ . The time-integrated air concentration is adjusted to account for settling loss rate using the following approach:

$$\frac{d(Activity)}{dt} = -Activity \times \frac{ventilation rate}{room volume} - Activity \times \frac{deposition velocity}{room height}$$

The loss rate is then

loss rate = air exchange rate + 
$$\frac{deposition \ velocity}{room \ height}$$

For these high-density particles a deposition velocity of 0.01 m/s is appropriate, resulting in a loss rate of 18 hr<sup>-1</sup> (i.e., 6 hr<sup>-1</sup> + 0.01 m/s × 3600 s/hr ÷ 3 m). The settling loss (12 hr<sup>-1</sup>) dominates, even in the laboratory setting with six air exchanges per hour.

NUREG-1717 uses a generic release factor of  $10^{-3}$  for powders. The same factor ( $10^{-3}$ ) is used in

NUREG-0706 as part of an empirical expression to represent the initial release of plutonium oxide from a falling drum and also is reported to be valid for  $U_3O_8$  (NRC 1980). This release factor  $(10^{-3})$  may be conservative because (1) the source materials are used in crystal form (i.e., not as a powder) for sublimation in a high temperature vacuum chamber and (2) the thin-film coatings, especially thorium, are durable and stable.

It is assumed that a small manufacturing facility uses 150 pounds/year of source material. Lenses are placed in a vacuum chamber with thorium fluoride or natural uranium, the chamber is evacuated, and the source material is vaporized and is deposited on surfaces within the chamber. Upon opening and cleaning of the chamber,  $10^{-3}$  of the deposited source material is assumed to be dispersed into the room as a respirable powder.

The total annual inhalation intake by a worker would be 0.0252 g (i.e., 150 lb  $\times$  454 g/lb  $\times$  0.001  $\times$  [1/(180 m<sup>3</sup>) (18 hr<sup>-1</sup>)]  $\times$  1.2 m<sup>3</sup>/hr). Because of limited time between thorium and uranium separation and subsequent use, only short-lived progeny are considered for uranium and all progeny are considered for thorium.

The ingestion dose resulting from intake by a worker was also calculated. The EPA Exposure Factors Handbook offers a soil ingestion rate of 50 mg/day (EPA 1997). This value includes all soil, dust, and dirt consumed with foods. Beyond the NRC requirements, the scenario also is shaped by other industrial hygiene regulations, such as those of the U.S. Occupational Safety and Health Administration (29 CFR 1910). It is assumed that the manufacturer complies with the intent of 10 CFR 20.1201(e), which limits intake of soluble uranium to 10 mg/week to avoid the effects of heavy metal toxicity.

#### Thorium

Using the above scenario description with an intake of 0.0252 g source material yields an inhalation of about 100 Bq of thorium. The committed dose coefficients from ICRP Publication 68 (ICRP 1994) were used to estimate the inhalation doses for thorium, thorium progeny, and thorium with progeny, as shown in Table A-5. Results for inhalation dose during manufacturing of thorium coated lenses are then calculated as follows:

#### Inhalation

Thorium Only	$100 \text{ Bq} \times (5.20 \times 10^{-5} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 520 \text{ mrem}$
Thorium Progeny	$100 \text{ Bq} \times (4.18 \times 10^{-6} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 42 \text{ mrem}$
Thorium + Progeny	520  mrem + 42  mrem = 562  mrem

Using the above scenario ingestion intake rate (10 mg/week) for source material with a working period of 50 wk/yr yields an annual intake of 500 mg/yr source material. Doses were calculated for thorium, thorium progeny, and thorium with progeny, using the committed dose coefficients from ICRP Publication 68 (ICRP 1994) as shown in Table A-5. Results for ingestion dose during manufacturing of thorium coated lenses are then calculated as follows:

Ingestion

Thorium Only $500 \text{ mg/yr} \times 4.0 \text{ Bq/mg} \times (2.90 \times 10^{-7} \text{ Sv/Bq}) \times (1 \times 10^5 \text{ mrem/Sv}) = 58 \text{ mrem}$ Thorium Progeny $500 \text{ mg/yr} \times 4.0 \text{ Bq/mg} \times (7.42 \times 10^{-7} \text{ Sv/Bq}) \times (1 \times 10^5 \text{ mrem/Sv}) = 148 \text{ mrem}$ Thorium + Progeny58 mrem + 148 mrem = 206 mrem

#### Uranium

Using the above scenario description with an intake of 0.0252 g source material yields an inhalation of about 630 Bq of uranium. The committed dose coefficients from ICRP Publication 68 (ICRP 1994) were used to estimate the inhalation doses for natural uranium (activity weighted average for <sup>234</sup>U, <sup>235</sup>U, and <sup>238</sup>U), uranium short-lived progeny, and uranium with short-lived progeny, as shown in Table A-6. Results for inhalation dose during manufacture of uranium coated lenses are then calculated as follows:

Inhalation	
Uranium Only	$630 \text{ Bq} \times (1.85 \times 10^{-6} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 117 \text{ mrem}$
Uranium Progeny	$630 \text{ Bq} \times (2.60 \times 10^{-9} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 0.16 \text{ mrem}$
Uranium + Progeny	117  mrem + 0.16  mrem = 117  mrem

Using the above scenario ingestion intake rate (10 mg/week) for source material with a working period of 50 wk/yr yields an annual intake of 500 mg/yr source material. Doses were calculated for uranium, uranium short-lived progeny, and uranium with short-lived progeny, using the committed dose coefficients from ICRP Publication 68 (ICRP 1994) as shown in Table A-6. Results for ingestion dose during manufacture of uranium coated lenses are then calculated as follows:

Ingestion	
Uranium Only	$500 \text{ mg/yr} \times 25 \text{ Bq/mg} \times (4.65 \times 10^{-8} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 58.1 \text{ mrem}$
Uranium Progeny	$500 \text{ mg/yr} \times 25 \text{ Bq/mg} \times (1.67 \times 10^{-9} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 2.1 \text{ mrem}$
Uranium + Progeny	58.1  mrem + 2.1  mrem = 60  mrem

Nuclide	Inhalation Dose Coefficient (Sv/Bq)	Ingestion Dose Coefficient (Sv/Bq)
Th-232	2.90E-05	2.20E-07
Ra-228	1.70E-06	6.70E-07
Ac-228	1.20E-08	4.30E-10
Th-228	2.30E-05	7.00E-08
Ra-224	2.40E-06	6.50E-08
Pb-212	3.30E-08	5.90E-09
Bi-212	3.90E-08	2.60E-10

Table A-5. Thorium Inhalation and Ingestion Dose Coefficients from ICRP Publication 68 (ICRP 1994).

Table A-6. Uranium Inhalation and Ingestion Dose Coefficients from ICRP Publication 68 (ICRP 1994).

Nuclide	Inhalation Dose Coefficient (Sv/Bq)	Ingestion Dose Coefficient (Sv/Bq)
U-238	1.60E-06	4.40E-08
Th-234	5.30E-09	3.40E-09
Pa-234m	0.00E+00	0.00E+00
U-234	2.10E-06	4.90E-08
U-235	1.80E-06	4.60E-08
Th-231	3.70E-10	3.40E-10

# Radiation Dose Assessment for Large and Small Anion Resin Exchange Columns Containing 15 Pounds Uranium (Section 5.2.2.1, Routine Use – Resin with 15 Pounds Uranium)

For routine use, the dose assessment is modeled using MicroShield Version 6.2 (Grove Engineering 2003) for large and small resin columns containing 15 pounds of uranium. The large anion exchange column is assumed to be 20-ft tall and 15-ft diameter, the size reported for the drinking water filtration system in Oswego, Illinois (Hutchinson 2006). The small anion exchange column is assumed to be 5.2-ft high and 1-ft diameter. Both columns are assumed to have a 5 mm steel wall thickness. Separate analyses were performed assuming the resin contains uranium only, uranium with short-lived progeny, and uranium with all progeny. The MicroShield output value selected for reporting is the effective dose estimated using the rotational geometry factors from ICRP Publication 51 (ICRP 1987).

For the large column, assuming a resin density of 2.0 g/cm<sup>3</sup> and a resin volume of  $1.00 \times 10^8$  cm<sup>3</sup> (i.e., total resin mass of  $2.00 \times 10^8$  g), holding 15 pounds uranium would be 0.0034 percent uranium by weight (i.e., does not exceed the 0.05 percent uranium by weight limit). With an exposure distance of 1 m at mid height, MicroShield Version 6.2 calculated exposure rates in units of mrad/hr that were approximated as equivalent to mrem/hr, then multiplied by 2000 hr/yr exposure time to obtain an annual scenario dose, as follows:

Large	Exchange	Column
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Uranium Only	$3.18 \times 10^{-5} \text{ mrad/hr} \times 2000 \text{ hr/yr} = 0.064 \text{ mrem}$
Uranium + Short-Lived Progeny	$1.25 \times 10^{-4} \text{ mrad/hr} \times 2000 \text{ hr/yr} = 0.25 \text{ mrem}$
Uranium + Progeny	$6.60 \times 10^{-3} \text{ mrad/hr} \times 2000 \text{ hr/yr} = 1.3 \text{ mrem}$

A similar scenario was developed for a smaller anion exchange column (156-cm height and 30-cm diameter [i.e., about 4 ft<sup>3</sup>]) holding 15 pounds uranium, maintaining previous 5-mm-thick steel wall and 1-m exposure distance. This geometry results in a uranium concentration of about 4 lb/ft<sup>3</sup>, which is equivalent to about 3.1 percent uranium by weight (i.e., does exceed the 0.05 percent uranium by weight limit). MicroShield calculated exposure rates in units of mrad/hr that were approximated as equivalent to mrem/hr, then multiplied by 2000 hr/yr exposure time to obtain an annual scenario dose, as follows:

Small Exchange Column	
Uranium Only	$2.84 \times 10^{-3}$ mrad/hr × 2000 hr/yr = 5.68 mrem
Uranium + Short-Lived Progeny	$1.01 \times 10^{-2} \text{ mrad/hr} \times 2000 \text{ hr/yr} = 20 \text{ mrem}$
Uranium + Progeny	$5.32 \times 10^{-1} \text{ mrad/hr} \times 2000 \text{ hr/yr} = 1060 \text{ mrem}$

# Radiation Dose Assessment for Large and Small Anion Resin Exchange Columns Containing 0.05 wt% Uranium (Section 5.2.2.2, Routine Use – Resins at 0.05 Percent Uranium by Weight)

For routine use, the dose assessment is modeled using MicroShield Version 6.2 (Grove Engineering 2003) for large and small resin columns containing 0.05 percent by weight uranium. The large anion exchange column is assumed to be 20-ft tall and 15-ft diameter, the size reported for the drinking water filtration system in Oswego, Illinois (Hutchinson 2006). The small anion exchange column is assumed to be 5.2-ft high and 1-ft diameter. Both columns are assumed to have a 5 mm steel wall thickness. Separate analyses were performed assuming the resin contains uranium only, uranium with short-lived progeny, and uranium with all progeny. The MicroShield output value selected for reporting is the effective dose estimated using

the rotational geometry factors from ICRP Publication 51 (ICRP 1987).

For the large column, assuming a resin density of 2.0 g/cm<sup>3</sup> and a resin volume of  $1.00 \times 10^8$  cm<sup>3</sup>, (i.e., total resin mass of  $2.00 \times 10^8$  g), holding 0.05 percent by weight uranium would be about 220 pounds of uranium. Because of identical geometry and source term, the results from Section 5.2.2.1 are scaled by the mass of uranium (i.e., 220 lb / 15 lb), as follows:

$3.18 \times 10^{-5}$ mrad/hr × 2000 hr/yr × (220 lb / 15 lb) = 0.93 mrem
$1.25 \times 10^{-4} \text{ mrad/hr} \times 2000 \text{ hr/yr} \times (220 \text{ lb} / 15 \text{ lb}) = 3.7 \text{ mrem}$
$6.60 \times 10^{-3} \text{ mrad/hr} \times 2000 \text{ hr/yr} \times (220 \text{ lb} / 15 \text{ lb}) = 194 \text{ mrem}$

For the small column, assuming a resin density of 2.0 g/cm<sup>3</sup> and a resin volume of about  $1.10 \times 10^5$  cm<sup>3</sup>, (i.e., total resin mass of  $2.20 \times 10^5$  g), holding 0.05 percent by weight uranium would be about 0.24 pounds of uranium. Because of identical geometry and source term, the results from Section 5.2.2.1 are scaled by the mass of uranium (i.e., 0.24 lb / 15 lb), as follows:

Small Exchange Column	
Uranium Only	$2.84 \times 10^{-3}$ mrad/hr × 2000 hr/yr × (0.24 lb / 15 lb) = 0.09 mrem
Uranium + Short-Lived Progeny	$1.01 \times 10^{-2}$ mrad/hr × 2000 hr/yr × (0.24 lb / 15 lb) = 0.32 mrem
Uranium + Progeny	$5.32 \times 10^{-1} \text{ mrad/hr} \times 2000 \text{ hr/yr} \times (0.24 \text{ lb} / 15 \text{ lb}) = 17 \text{ mrem}$

# Radiation Dose Assessment for 10 CFR 40.22 Bounding Scenario (Section 5.3.1, 10 CFR 40.22 Bounding Dose Assessment)

The bounding scenario is assumed as an individual that uses 150 pounds of uranium or thorium powder per year in a small room. The calculations described here are similar to those performed in Section 5.1.2.3, except with a smaller room and a lower ventilation rate. Assuming a 30 m<sup>3</sup> room, with 0.5 air exchanges per hour (the median national residential rate as determined by Murray and Burmaster [1995]), accounting for settling rate using approach used in Section 5.1.2.3 yields a total loss rate of 12.5 hr<sup>-1</sup> loss rate component (12 hr<sup>-1</sup>) dominates over the room ventilation loss rate (0.5 hr<sup>-1</sup>). Because of limited time between thorium and uranium separation and subsequent use, only short-lived progeny are considered for uranium and all progeny are considered for thorium.

The inhalation intake will then be 150 lb × 454 g/lb ×  $10^{-3}$  release fraction × [1/(30 × 12.5)] hr/m<sup>3</sup> × 0.9 m<sup>3</sup>/hr = 0.16 g/yr (650 Bq thorium or 4000 Bq uranium). The committed inhalation dose coefficients from ICRP Publication 68 (ICRP 1994) were used to estimate the inhalation doses for thorium and natural uranium (Tables A-5 and A-6). Using the same approach, doses were calculated for thorium, thorium progeny, uranium, and uranium short-lived progeny, as follows:

Thorium

Inhalation

Thorium Only Thorium Progeny Thorium + Progeny	650 Bq × (5.20 × $10^{-5}$ Sv/Bq) × (1 × $10^{5}$ mrem/Sv) = 3380 mrem 650 Bq × (4.18 × $10^{-6}$ Sv/Bq) × (1 × $10^{5}$ mrem/Sv) = 272 mrem 3380 mrem + 272 mrem = 3652 mrem
Uranium	
Inhalation	
Uranium Only	4000 Bq × $(1.85 \times 10^{-6} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 740 \text{ mrem}$
Uranium Progeny	4000 Bq × $(2.60 \times 10^{-9} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 1 \text{ mrem}$
Uranium + Progeny	740  mrem + 1  mrem = 741  mrem

The ingestion dose resulting from intake by the owner/employee was also calculated. The EPA *Exposure Factors Handbook* offers a soil ingestion rate of 50 mg/day (EPA 1997). It is assumed that the ceramics shop owner/employee spends the equivalent of 8 hours/week (1 working day per week) mixing pigments, cleaning up, and vacuuming. Scaling the EPA *Exposure Factors Handbook* value for ingestion of 50 mg/day from surfaces, gives 50 mg/wk (2.5 g/yr). The committed ingestion dose coefficients from ICRP Publication 68 (ICRP 1994) were used to estimate the ingestion doses for thorium and natural uranium (Tables A-5 and A-6). Using the same approach, doses were calculated for thorium, thorium progeny, uranium, and uranium short-lived progeny, as follows:

Thorium

Ingestion	
Thorium Only	2.5 g/yr × 4000 Bq/g × (2.90 × $10^{-7}$ Sv/Bq) × (1 × $10^{5}$ mrem/Sv) = 290 mrem
Thorium Progeny	2.5 g/yr × 4000 Bq/g × $(7.42 \times 10^{-7} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 740 \text{ mrem}$
Thorium + Progeny	290 mrem + 740 mrem = 1030 mrem/yr

Uranium

Ingestion	
Uranium Only	$2.5 \text{ g/yr} \times (24,900 \text{ Bq/g}) \times (4.65 \times 10^{-8} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 290 \text{ mrem}$
Uranium Progeny	2.5 g/yr × (24,900 Bq/g) × (1.67 × $10^{-9}$ Sv/Bq) × (1 × $10^{5}$ mrem/Sv) = 10 mrem
Uranium + Progeny	290  mrem + 10  mrem = 300  mrem/yr

# Radiation Dose Assessment for 10 CFR 40.13(a) Bounding Scenario (Section 5.3.2, 10 CFR 40.13(a) Bounding Dose Assessment)

A bounding assessment for the use of source material under 10 CFR 40.13(a) was developed for an individual who works in an environment with airborne source material that is 0.05 percent uranium or thorium by weight. For this analysis, it is assumed that airborne source material (uranium and thorium) is maintained at the threshold limit value for nuisance dust, 10 mg/m<sup>3</sup> (ACGIH 2001). At this level, it is reasonable to assume that no respiratory protection would be used. Because a given airborne concentration is assumed (i.e., 10 mg/m<sup>3</sup>), no credit is given for ventilation rate, and the person could be assumed to be working indoors or outdoors in these conditions. A breathing rate of 1.2 m<sup>3</sup>/hr is assumed. Given a 10 mg/m<sup>3</sup> loading rate, 1.2 m<sup>3</sup>/hr breathing rate, 0.05 percent by weight uranium or thorium, and 2000 hr exposure time, the resulting total annual inhalation intake is 12 mg of either uranium. The committed inhalation dose coefficients from ICRP Publication 68 (ICRP 1994) were used to estimate the inhalation doses were calculated for thorium, thorium progeny, uranium, and uranium short-lived progeny, as follows:

Thorium

Inhalation	
Thorium Only	49 Bq × $(5.20 \times 10^{-5} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 255 \text{ mrem}$
Thorium Progeny	49 Bq × (4.18 × 10 <sup>-6</sup> Sv/Bq) × (1 × 10 <sup>5</sup> mrem/Sv) = 20 mrem
Thorium + Progeny	255 mrem + 20 mrem = 275 mrem
Uranium	
Inhalation	
Uranium Only	$320 \text{ Bq} \times (1.85 \times 10^{-6} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 59 \text{ mrem}$
Uranium Progeny	$320 \text{ Bq} \times (2.60 \times 10^{-9} \text{ Sv/Bq}) \times (1 \times 10^{5} \text{ mrem/Sv}) = 0.08 \text{ mrem}$
Uranium + Progeny	59  mrem + 0.08  mrem = 59  mrem

# Radiation Dose Assessment for 10 CFR 40.13(b) Bounding Scenario (Section 5.3.3, 10 CFR 40.13(b) Bounding Dose Assessment)

For this bounding analysis, it is assumed that a hobbyist obtains and displays a 1-kg piece of pitchblende ore. The ore is assumed to contain 500 grams of  $^{238}$ U (approximately 50 percent  $U_3O_8$  by weight), with all associated isotopes of uranium in equilibrium with their decay progeny. Additionally, an assessment was performed for uranium only and uranium with short-lived progeny. Uranium ore that has not undergone chemical processing will contain all members of the uranium decay series, including radium and its progeny (ORAU et al. 2006). The activities (assuming secular equilibrium) of the various radionuclides in the ore sample (reported in curies) are provided as Table 15 in the main report.

The ore sample is assumed to be a sphere of density about 5 g/cm<sup>3</sup>, with a diameter of 6 cm. The dose rate for uranium with all progeny calculated using MicroShield Version 6.2 is about 0.5 mrem/hr at a distance of one meter. If the hobbyist displays the sample on a desk that is occupied 2000 hr/yr, the dose would be approximately 1.1 rem/yr. Results for uranium only, uranium with short-lived progeny, and uranium with all progeny are calculated as follows:

MicroShield (6-cm diameter sphere wi	th density 5 g/cm <sup>3</sup> containing 500 g natural uranium with progeny)
Uranium Only	$(2.84 \times 10^{-3} \text{ mrad/hr}) \times 2000 \text{ hr/yr} = 5.68 \text{ mrem}$
Uranium + Short-Lived Progeny	$(1.01 \times 10^{-2} \text{ mrad/hr}) \times 2000 \text{ hr/yr} = 20.2 \text{ mrem}$
Uranium + Progeny	$(5.32 \times 10^{-1} \text{ mrad/hr}) \times 2000 \text{ hr/yr} = 1064 \text{ mrem/yr}$

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10 CFR Part 40. Code of Federal Regulations, Title 10, *Energy*, Part 40, "Domestic Licensing of Source Material."

29 CFR Part 1910. Code of Federal Regulations, Title 29, *Labor*, Part 1910, "Occupational Safety and Health Standards."

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