

ORAU TEAM Dose Reconstruction Project for NIOSH

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Page 1 of 47

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Document No. ORAUT-TKBS-0043	Revision No. 00	Effective Date: 02/14/2006	Page 2 of 47

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02/14/2006	00	New technical basis document for the W. R. Grace and Company in
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TABLE OF CONTENTS

SECT	<u>ION</u>	<u>TITLE</u>	<u>PAGE</u>
Acron	ovms and	I Abbreviations	6
7 (0.01	.ye ae		
1.0	Introdu	iction	8
2.0		escription, History, and Process	
	2.1	Process Description	9
		2.1.1 Production of Uranium Metal and Uranium Metal Alloys	40
		Enriched up to 12% Uranium-235	
		2.1.1.2 Reduction of Uranium Tetrafluoride to Uranium Metal	
		2.1.1.2 Reduction of Granium Tetrandonde to Granium Metal	
		2.1.1.4 Other Metal Products	
		2.1.1.5 Packaging and Shipping	
		2.1.2 Scrap Recovery Operations	
		2.1.3 Highly Enriched Uranium from Scrap Recovery	
		2.1.3.1 Receiving and Storage	
		2.1.3.2 Sampling	
		2.1.3.3 Head-End Operations	
		2.1.3.4 Final Process Steps	
		2.1.4 Production of Uranium Oxide Mixed with Thorium Oxide	
		and Zirconium Oxide	13
		2.1.4.1 Receiving and Storage	
		2.1.4.2 Solution Concentration	
		2.1.4.3 Precipitation	
		2.1.4.4 Drying and Grinding	
		2.1.4.5 Calcination to Uranium Dioxide	
		2.1.4.6 Blending	
		2.1.4.7 Diluent Addition and Blending	
		2.1.4.8 Binder Addition	
		2.1.4.9 Lubricant Addition and Pressing	
		2.1.4.10 Binder Removal and Sintering	
		2.1.4.12 Tube Loading	
		2.1.5 Production of Mixed Oxide Fuel	14 1 <i>1</i>
		2.1.6 Uranium Hexafluoride Cylinder Washing	
2.0	□ation a	tion of Internal Franceiro	4.4
3.0		tion of Internal Exposure	
	3.1	Uranium Exposures	
		3.1.1 Uranium Air Sampling	
		3.1.3 Uranium Bioassay	
	3.2	Plutonium Exposures	
	5.2	3.2.1 Plutonium Organ Dose Calculations	
		3.2.2 Plutonium Bioassay	
	3.3	Thorium Exposures	
	3.4	Uranium and Plutonium Dioxide- MOX, Mixed Oxide Fuel Exposures	
	3.5	Americium	
	3.6	Occupational Internal Dose Reconstruction Summary And Assumptions -	

References44

LIST OF TABLES

TABL	<u>E</u> <u>TITLE</u>	<u>PAGE</u>
2-1	W.R. Grace and NFS AEC weapons related work time period and locations	10
3-1	Fuel types, chemical form, isotope and enrichment of W.R. Grace process	
	material	
3-2	Uranium source term information	
3-3	Occupational exposures for 93% uranium-aluminum alloy recovery	
3-4	Average breathing-zone samples for 93% uranium-aluminum alloy recovery	17
3-5	Average general air-sampling concentrations for 93% uranium-aluminum	
	alloy recovery	17
3-6	Occupational exposures for 3.6% uranium recovery from slag	
3-7	Average breathing-zone samples for 3.6% uranium recovery from slag	18
3-8	Average general air-sampling concentrations for 3.6% uranium recovery from	4.0
	slag	19
3-9	Estimated uranium inhalation intake rates based on time weighted air	
	concentrations measured during uranium recovery operations	20
3-10	Estimated uranium ingestion rates based on time weighted air concentrations	
	measured during uranium recovery operations	
3-11	Intakes of contaminants in recycled uranium as fractions of uranium intake	21
3-12	Uranium urinalysis recorded results for claims submitted as of 10/01/05 in	
		22
3-13	Activity composition of Hanford reference fuel-grade plutonium mixture (12%)	23
3-14	Organ dose for 1 yr for Hanford reference 10-yr-aged fuel-grade plutonium	
	mixture (12%) and Hanford reference 5-yr-aged fuel-grade plutonium mixture	
	(6%) for an annual chronic intake of 6.24×10^5 pCi/yr and absorption type M	
3-16	Plutonium urinalysis results for claims	25
4-1	Minimum detectable level (MDL) and maximum potential missed photon or	
	beta dose	
4-2	Dosimetry for external whole-body, wrist, and extremity exposures	
4-3	Annual occupational radiation exposures for 1982 to 1994	
4-4	Annual occupational TEDE for 1995 to 2002	31
4-5	Annual organ doses due to exposure to a 5-by-5 stacked array of birdcages	
4-6	Uranium activity fractions	
4-7	External exposure from thorium surface contamination	
4-8	External exposure from ²³³ U/thorium surface contamination	
4-9	External exposure from 93%-enriched uranium surface contamination	
4-10	External exposure from 3.5%-enriched uranium surface contamination	
4-11	External exposure from DU surface contamination	
4-12	External exposure from plutonium surface contamination	
4-13	External exposure from MOX surface contamination	
4-14	Plutonium isotopic content and specific activity	40
4-15	Annual organ doses due to the assumed annual diagnostic chest X-ray	41
4-16	Operational external exposure summary	42
	LIST OF FIGURES	
<u>FIGUI</u>	RE TITLE	PAGE
2-1	Location of Erwin, Tennessee	9
2-2	Plant site at W.R. Grace/NFS	9

Document No. ORAUT-TKBS-0043 Revision No. 00 Effective Date: 02/14/2006 Page 6 of 47

ACRONYMS AND ABBREVIATIONS

ADU ammonium diuranate

AEC U.S. Atomic Energy Commission

CDE committed dose equivalent CFR Code of Federal Regulations

cm centimeter

d day

DCF dose conversion factor
DOE U.S. Department of Energy
dpm disintegrations per minute

DU depleted uranium

EEOICPA Energy Employees Occupational Illness Compensation Program Act of 2000

EU enriched uranium

g gram

GSD geometric standard deviation

HASL Health and Safety Laboratory
HEU highly enriched uranium

hr hour

ICRP International Commission on Radiological Protection

IMBA Integrated Modules for Bioassay Analysis IREP Interactive RadioEpidemiology Program

keV kilo electron volt, 1,000 electron volts

kg kilogram

L liter

LEU low-enriched uranium

m meter

MDA minimum detectable activity
MDC minimum detectable concentration

MDL minimum detectable level

MeV mega electron volt, 1 million electron volts

ml milliliter
MOX mixed oxide
mR milliroentgen
mrad millirad
mrem millirem

nCi nanocurie

NFS Nuclear Fuels Services

NIOSH National Institute for Occupational Safety and Health

NRC U.S. Nuclear Regulatory Commission

pCi picocurie

Document No. ORAUT-TKBS-0043 Revision No. 00 Effective Date: 02/14/2006 Page 7 of 47

SEFOR South-West Experimental Fast Oxide Reactor

TBD technical basis document

U.S.C. United States Code

yr year

μCi microcurie μg microgram μm micrometer 1.0

INTRODUCTION

Technical basis documents (TBDs) and site profile documents are general working documents that provide guidance concerning the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist the National Institute for Occupational Safety and Health (NIOSH) in the completion of the individual work required for each dose reconstruction.

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

EEOICPA, as amended, provides for employees who worked at an AWE facility during the contract period and/or during the residual contamination period.

Employment at an AWE facility is categorized as either (1) during the contract period (*i.e.*, when the AWE was processing or producing material that emitted radiation and was used in the production of an atomic weapon), or (2) during the residual contamination period (*i.e.*, periods that NIOSH has determined there is the potential for significant residual contamination outside of the period in which weapons-related production occurred). For contract period employment, all radiation exposures must be included in dose reconstructions. For residual contamination period employment, only the radiation exposures defined in 42 U.S.C. §7384n(c) (4) [i.e., radiation doses received from DOE/U.S. Atomic Energy Commission (AEC) -related work] must be included in dose reconstructions. This site profile covers only exposures from nuclear weapons-related work. Exposures from non-weapons related work, if applicable, will be covered elsewhere.

This site profile provides specific information concerning documentation of historical practices at the W.R. Grace and Company plant. If the dose reconstructor cannot determine whether the work performed was AEC weapons related work or from non-AEC related work then the dose reconstructor should assume that all of the work performed was from AEC weapons related work to ensure claimant-favorability.

2.0 SITE DESCRIPTION, HISTORY, AND PROCESS

The Davison Chemical Company, a division of W.R. Grace, began processing radioactive materials in late 1957 at the site of the current Nuclear Fuels Services (NFS) facility near Erwin, Tennessee. (NFS 2005) In 1964, NFS was formed by the merger of W.R. Grace, and American Machine and Foundry Company. Ownership of NFS transitioned from Getty Oil to Texaco as part of a stock buyout of Getty Oil. In 1987, NFS Services, Ltd., a private Atlanta-based limited partnership purchased NFS, Inc., from Texaco, Inc. Figure 2-1 shows the location of Erwin, Tennessee. Figure 2-2 shows the principal site infrastructure for the NFS facility as of 1986 (Congress 1986).

Regulatory authority over operations was originally under the AEC (1954 to 1974) and transitioned to the U.S. Nuclear Regulatory Commission (NRC) (1975 to present) under Special Nuclear Material License No. SNM-124, as amended.

The principal operation at the site has been to convert highly enriched uranium (HEU) and low-enriched uranium (LEU) from UF₆ to a product that meets customer requirements. Thorium, depleted uranium (DU), ²³³U, and plutonium have also been processed at various times to oxides or metals with subsequent processing into the form necessary for the manufacture of nuclear fuel. Specifically, the

facility at one time processed ThO₂ mixed with ²³³U to make the light-water breeder reactor fuel for the Shippingport Reactor. Plutonium and DU mixed oxide (MOX) fuel for the South-West Experimental



Figure 2-1. Location of Erwin, Tennessee.

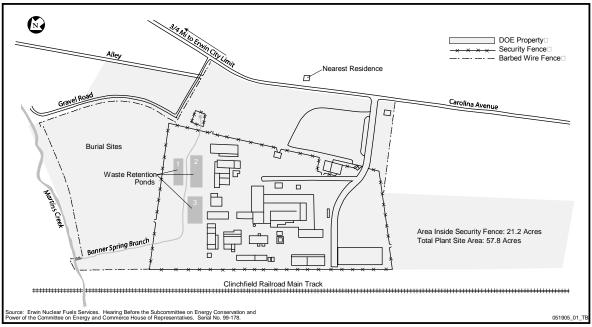


Figure 2-2. Plant site at W.R. Grace/NFS.

Fast Oxide Reactor (SEFOR) was fabricated in Building 234 with laboratory capabilities in Building 110; both operations ceased in 1970. Other operations were associated with the conversion operations such as scrap recovery operations (uranium and other nuclear fuel material) and cleaning and certification of empty cylinders used to transport low-enriched UF $_6$. Table 2-1 lists the approximate processing history of the site.

2.1 PROCESS DESCRIPTION

Brief summaries for the principal operations are given below and additional details can be found in the series of Feasibility Reports provided in the cited references. Inherent in all of the operations is nuclear criticality safety that governs not only the operations and storage but also the movement of material within the facility. Nuclear criticality safety is maintained at the facility through the control of one or more of the conventional parameters of geometry, mass, concentration, and control of neutron interaction between subcritical units. The standard administrative policy at this and other similar facilities is to control two such parameters whenever possible. For birdcage units, the two control

Document No. ORAUT-TKBS-0043	Revision No. 00	Effective Date: 02/14/2006	Page 10 of 47

Table 2-1. W.R. Grace and NFS AEC weapons related work time period and locations.

	Period of AEC Weapons	
Operations	Related Work	Building Location
Thorium (metal & oxide)	Late 1957 to 1970	110C, 110D, 111, 130, 234B,
		234C, 310
Uranium metal (HEU & DU)	Late 1957 to 1970	110, 110E, 111, 130, 135, 234,
		301
Low-enriched UO ₂	Early 1960 to 1970	301, 110E, 111, 130, 135, 301
²³³ U fuel (sometimes mixed with	1961 -1970	234B, 234C, 110C, 110D
ThO ₂)		
Plutonium fuel and MOX	1966 to 1970	234A, 234B, 234C, 110C, 110D
HEU scrap recovery	Late 1957 to 1968	130
	1968 through 1970 ^a	220, 230, 233
HEU Fuel	Early 1966 to 1970 ^a	100, 105, 120, 131, 132/133, 220,
		233, 300, 302, 303, 304, 310, 330
		(Congress 1986, p. 111) 301 (from
		1982) (Congress 1986, p. 132)
LEU Scrap Recovery	1960 to 1970 ^a (AEC 1961), or	111, 220, 230, 233
	(Congress 1986)	
LEU cylinder washing	1970 ^a	111,130

Sources: AEC (1961), Congress (1986, pp. 111-114 and 127-143).

parameters are geometry (birdcage dimensions) and a 235 U mass limit that can be placed within the birdcage. Examples of bird cages that were used included: U-233 storage bird cage that used a shielded 55-gallon drum that contained inserts with spacers; UF $_6$ cylinders storage bird cage that used a 2" × 2" frame made of metal that held the 5" UF $_6$ cylinder in place at its center with a total dimension of 52" high and 30 " wide; and a U_3O_8 storage bird cage that used a 2" × 2" metal frame made of metal that held a 10 3 /4" × 12" cylinder in the center with a total dimension of 36" high and 30 5/8 ". A review of the available literature shows that no criticality accidents have occurred during W.R. Grace and later NFS operations.

2.1.1 <u>Production of Uranium Metal and Uranium Metal Alloys Enriched up to 12% Uranium-235</u>

The UF₆ was supplied in approved standard cylinders and received in approved packaging such as birdcages. The cylinders were check-weighed and placed into storage in special concrete cells or birdcages in one of the warehouse buildings.

2.1.1.1 Conversion of Uranium Hexafluoride to Uranium Metal

The overall process involves vaporization, reduction to UF₄, reduction to uranium metal, pickling, processing into other metal products, packaging, and shipment. The following information is from *Feasibility Report for the Production of Uranium Metal and Uranium Metal Alloys Enriched Up To 12%* ²³⁵*U* (Housholder 1963a).

 UF_6 was received in solid form in cylinders and had to be vaporized to transfer it to the UF_6 to UF_4 reactor (the 6 to 4 unit). Vaporization was accomplished by heating the UF_6 cylinders in an electric oven. Up to six cylinders could be placed in to the oven for processing.

The UF₆ was piped into the 6 to 4 unit where it was reduced with hydrogen. The solid UF₄ powder dropped into a product hopper where it was metered into safe diameter product cans. The off gas flowed through two cyclone separators where any entrained UF₄ dust was collected in additional

a. The end of 1970 is when AEC-weapons related operations ended.

product cans. The off gas was then filtered to remove the last traces of UF₄ dust and was scrubbed with KOH to remove HF vapors. The gas, free of uranium and acid, was vented to the atmosphere through a flame arrester where excess hydrogen was burned off.

2.1.1.2 Reduction of Uranium Tetrafluoride to Uranium Metal

The UF₄ was weighed into a reduction batch and was blended with a reducing agent such as magnesium metal. The charge was then heated under vacuum in an induction furnace to form a uranium metal derby. After cooling, the uranium metal derby was broken out and separated from the slag. The slag was packaged for scrap recovery where any uranium remaining in the slag was recovered.

2.1.1.3 Pickling

The uranium derby was pickled in acid to remove adherent slag and scale. The pickle solution was sent to scrap recovery for recovery of uranium. The pickled derby was then sampled for impurities.

2.1.1.4 Other Metal Products

As necessary, the derbies were broken into smaller pieces before shipment on a large hydraulic press. On other occasions, the derbies were remelted and cast into various shapes such as slugs, rods, and plates. The uranium could also be alloyed with other metals during remelting and casting. Melting was done in a large vacuum induction furnace. Cast pieces could be pickled as previously described.

2.1.1.5 Packaging and Shipping

All enriched uranium (EU) metal products were packaged for shipment in approved birdcages. The products could be stored on site for a time before shipping to the customer.

2.1.2 Scrap Recovery Operations

This operation was designed to recover the scrap uranium from the manufacturing processes. The process steps were assumed to be carried out in a similar manner for scrap with either HEU or LEU (Katine 1960). The exception between handling the different enrichment levels would be in the dissolution and filtration steps. There was potential for different end processes that were adjusted based on the final material form desired by either the customer (for scrap recovery from outside sources) or how the facility was going to reinsert the recovered uranium in the fuel fabrication process.

Dependent of whether the material was greater or less than 2% enrichment, two different batch sizes and dissolver tanks were used. For scrap material equal to less than 2% enrichment, a maximum batch of 87.95 kg uranium (220 pounds) of UO_2 is dissolved in a 500-gallon capacity dissolver tank. For all other enrichments, a 20-gallon dissolver tank was employed and the batch would contain a maximum of 400 g of ^{235}U .

Of note for this process, the memorandum attached to a Feasibility Report for HEU scrap recovery specifically states "the chopping and sawing of solid metal plates, billets, and rods, and the operations of grinding, screening, and blending of the resultant oxides from the calcinations step, could be possible dust sources. Air samples should be obtained at these operations and evaluated at the start

of operations in order to ascertain the effectiveness of the existing ventilation (Glauberman 1962)." A summary of the HEU scrap recovery follows (Housholder and Runion 1962).

2.1.3 <u>Highly Enriched Uranium from Scrap Recovery</u>

2.1.3.1 Receiving and Storage

All scrap would be received in approved shipping containers and birdcages and sent to storage warehouses to await processing.

2.1.3.2 Sampling

Before processing, each container was inspected for the purpose of deciding what head-end steps could be necessary before the scrap could be dissolved. Also at this time, samples were taken for uranium assay for the purpose of determining batch sizes.

2.1.3.3 Head-End Operations

Due to the many types of scrap received, it was often necessary to pretreat the scrap before dissolution and subsequent extraction. Metal chips and turnings, for instance, were routinely received stored under oil, which must be removed before dissolution. This was done by draining the oil off the metal in wire baskets. The metal was then washed with solvents and dried.

Solid metal plates and rods were chopped or sawed into smaller pieces to facilitate handling and weighing into batches. Combustible wastes such as filters, sponges, grinder sludge, sweepings, etc., were calcined to reduce their bulk and to remove hydrogenous and carbonaceous materials, such as water and oil. After calcining, the resultant oxides were ground and screened; material not passing the screen was recycled to the grinder. The oxides were then blended to ensure homogeneity for sampling for uranium assay from which accountability and batch sizes could be determined.

Oxide pellets were ground and screened and could be calcined and blended to facilitate dissolution.

2.1.3.4 Final Process Steps

A review of the Feasibility Reports showed there were different final process steps that appear to depend on the final material form needed either to be reinserted into the facility process or to be shipped back to the customer. The extracted uranium liquid (also known as O.K. Liquor) could be converted into a solid through either a boil-down and crystallization process (Housholder and Runion 1962) or by precipitation and filtration followed by calcination and blending (AEC 1959). A summary of both final process steps follows:

- <u>Boil-down and Crystallization</u>: The pure uranium solution could be boiled down and evaporated to produce uranyl salts, which were packaged for shipment. At times, the concentrated solution from the boil-down step was packaged in bottles for shipment as a solution.
- <u>Precipitation/Filtration and Calcination/Blending</u>: The extracted uranium liquid was precipitated by batch process with ammonium hydroxide in an approved container with an agitator. The precipitated slurry was transferred to the filtration hood for filtration with a large Buchner funnel. The filter cake was loaded into metal trays and transferred to a hooded muffle furnace where it was calcined to U₃O₈. Once cooled, the material was transferred to a blending station

for introduction into a ball mill for grinding and blending. After ball milling, the U₃O₈ was sampled, packaged, weighed, and returned to the storage area ready for shipment.

2.1.4 Production of Uranium Oxide Mixed with Thorium Oxide and Zirconium Oxide

This line of production (233 UO₂/ThO₂ and 233 UO₂/ZrO₂) was in operation from approximately 1961 to 1969 for the light-water breeder reactor fuel of the Shippingport Reactor. This process was more involved than other process lines with eleven separate steps to produce the 233 U fuel (Housholder 1963b). A brief summary of the process follows.

2.1.4.1 Receiving and Storage

²³³U was received as a uranyl nitrate solution in an approved shipping cask. A typical receipt shipment was approximately 7 kg of ²³³U stored in storage columns waiting processing.

2.1.4.2 Solution Concentration

The received uranyl nitrate solution had a concentration of approximately 150 g/L ²³³U. Before precipitation, a higher concentration was required and was obtained through evaporation.

2.1.4.3 Precipitation

The uranyl nitrate solution was measured into safe batches and the uranium was precipitated by the addition of a precipitating agent such as NH_4OH .

2.1.4.4 Drying and Grinding

The resultant precipitate was dried at a low temperature and ground to a fine powder.

2.1.4.5 Calcination to Uranium Dioxide

The dried ground uranium precipitate was calcined to UO₂ in a continuous muffle furnace under a hydrogen atmosphere. The resultant oxide was stored in birdcages to await blending.

2.1.4.6 **Blending**

To ensure homogeneity of the oxide, the precipitation batches were accumulated and blended before addition of the diluents (ThO₂ or ZrO₂). After a homogenous UO₂ blend was obtained, it was sorted in birdcages to await blending with the diluents.

2.1.4.7 Diluent Addition and Blending

Safe batches of UO₂ and diluents were weighed out and blended together in a twin-shell blender. To ensure thorough mixing, each batch was also ball milled.

2.1.4.8 Binder Addition

A binder was mixed with the oxide blend, and the wet mixture was granulated and dried. After drying, the granules were broken up by screening.

2.1.4.9 **Lubricant Addition and Pressing**

To improve pressability, a die lubricant was added to the granules and blended in. The oxides were then compacted into small pellets on a 40-ton press.

Binder Removal and Sintering 2.1.4.10

The resultant pellets were loaded into trays and heated in an oven to drive off the binder. They were then sintered in a continuous muffle furnace under a hydrogen atmosphere.

2.1.4.11 **Physical Measurement and Grinding**

The sintered pellets were inspected and measured to see that they met customer's requirements. Before packaging, the sintered pellets could have required grinding to the correct diameter If grinding were necessary, the pellets were cleaned by washing them in water, drying them, and re-measuring them.

2.1.4.12 **Tube Loading**

The finished pellets were stacked, weighed, and loaded into zircalloy tubes. The tubes were welded closed and loaded into shielded 55-gallon drum birdcages for temporary storage and eventual shipment to the customer.

2.1.5 **Production of Mixed Oxide Fuel**

Documentation of the process steps for the production of MOX fuel (combination of PuO₂) with low-enriched UO₂) for the SAFOR reactor at the facility was not available. However, other DOE sites have performed similar operations. From these documents, production of MOX fuel would have followed similar process steps for the production of ²³³UO₂/ThO₂ and ²³³UO₂/ZrO₂ fuel from the blending step on to tube loading. The process steps were completely within shielded and filtered gloveboxes for worker health and safety. It is assumed that the plutonium sent to the facility was in approved shipping packages as PuO₂ powder in sealed cans. The final product would have had approximately 5% ²³⁹Pu in fuel pin assemblies loaded into shielded, 55-gallon drum birdcages for temporary storage and shipment to the customer.

2.1.6 **Uranium Hexafluoride Cylinder Washing**

UF₆ cylinder washing was performed in the Building 200 complex to recover uranium in a ventilated glovebox using water or steam. The removed wash solution was transferred to the HEU scrap recovery process to recover the uranium.

3.0 **ESTIMATION OF INTERNAL EXPOSURE**

The primary sources of internal radiation exposure at W.R. Grace were uranium, plutonium, or thorium dust produced from the manipulation and chemical processing of those materials during recovery and fuel fabrication processes. Uranium enrichment levels included depleted, natural, low (3.5%), and high (93%). There was one indication of recycled uranium (RU) processing in the case of a recycled ²³³U pellet (Housholder 1963b). Table 3-1 lists the various enrichments and chemical forms of the processed radionuclides.

Document No. ORAUT-TKBS-0043	Revision No. 00	Effective Date: 02/14/2006	Page 15 of 47
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Table 3-1. Fuel types, chemical form, isotope and enrichment of W.R. Grace process material.

	Chemical form and		
Radionuclide or fuel	solubility type(s)	Isotope	Enrichment
Uranium	UF_6 , UO_2F_2 , & $UO_2(NO_3)_2$	²³³ U	DU, Natural,
	(F)	²³⁴ U	LEU (3.5%),
	$\dot{U}O_3 \& UF_4 (M)$	235	HEU (93%)
	$U_3O_8 \& UO_2, (S)$	²³⁶ U	,
		²³⁸ Ú	
Thorium	ThO ₂ (S)	²³⁰ Th (²³⁴ U)	Natural
		231 Th (235 U)	
		232 Th (236 U)	
		²³² Th (²³⁶ U) ²³⁴ Th (²³⁸ U)	
		²²⁹ Th (²³³ U)	
Plutonium	PuO ₂ (S)	²³⁸ Pu 0.64%, ²³⁹ Pu 2.06%, ²⁴⁰ Pu 1.07%, ²⁴¹ Pu 95.4%,	Fuel Grade
	, ,	²⁴⁰ Pu 1.07%, ²⁴¹ Pu 95.4%,	Aged 10 yr
		²⁴¹ Am 0.86% (% Activity)	
Technetium or other	Same as the Th, U or Pu	⁹⁹ Tc, ²³⁷ Np	NA
transuranic elements	matrix		
MOX	PuO ₂ /UO ₂ (S)	20% PuO ₂ and 80% UO ₂	About 3.5% ²³⁵ U
	, ,	by weight	
²³³ U fuel	ThO_2 / ²³³ UO ₂ or ZrO_2	20% UO ₂ and 80% ThO ₂ or	HEU (Likely > 20%)
	/ ²³³ UO ₂ (S)	ZrO ₂ by weight	, , , , , , ,

3.1 URANIUM EXPOSURES

For a given uranium process, the mass of (long-lived) uranium released to air does not change because of enrichment (ORAUT 2005a).

Uranium air sampling is further discussed in Section 3.1.1.

In general, it is not known how much of any specific enrichment versus another to which a worker could have been exposed. The uranium source term information in Table 3-2 can be used only if the information on the uranium enrichments that the worker processed is known. It is most claimant-favorable to assume the highest known processed enrichment.

Human and animal studies have indicated that oxides of uranium can be very insoluble (ICRP 1995), which indicates absorption type S. Other *in vitro* dissolution studies of compounds found at uranium facilities have shown that the oxides of uranium have exhibited moderate solubility (ORAUT 2005a), which suggests absorption type M. Because there was no specific information on the solubility of aerosols produced at W.R. Grace, this analysis assumed that both types M and S were available. The selection of absorption type should depend on the organ of interest. For example, absorption type S should be selected for respiratory tract dose calculations. Dose reconstructions should assume International Commission on Radiological Protection (ICRP) Publication 66 default parameters for particle deposition (ICRP 1994a).

ICRP (1994) lists UF₆, UO₂F₂, and UO₂(NO₃)₂ (uranyl nitrate) as type F; UF₄ and UO₃ as type M; and U₃O₈ and UO₂ as type S. The chemical form and the enrichment varied over time at W.R. Grace. The manufacture of uranium occurred in most of the buildings in W.R. Grace with the exception of Buildings 234A, 234B, and 234C, where plutonium was processed.

	Document No. ORAUT-TKBS-0043	Revision No. 00	Effective Date: 02/14/2006	Page 16 of 47
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Table 3-2. Uranium source term information.

		Specific activity			fractions	
Uranium source term	Reference	pCi/μg	²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U
Natural uranium	IMBA ^a	0.683	0.489	0.023	-	0.489
93.%	IMBA ^a	68.1	0.968	0.030	0.002	0.0003
3.5%	IMBA ^a	2.20	0.818	0.034	-	0.147
2%	HPS⁵	1.616	0.648	0.041	0.0009	0.311
Typical DU	IMBA ^a	0.402	0.155	0.011	0.0005	0.834
RU (1% ²³⁵ U)	Hanford ^c	0.910	0.563	0.023	0.048	0.365
		Specific activity	Specific	constituer	nt activity in	n mixture
Uranium source term	Reference	pCi/µg	()	ıCi/g, nCi/n	ng, or pCi/μ	ıg)
Natural Uranium	IMBA ^a	0.683	0.334	0.016	-	0.334
93.%	IMBA ^a	68.1	65.9	2.04	0.136	0.020
3.5%	IMBA ^a	2.20	1.80	0.075	-	0.323
2%	HPS⁵	1.616	0.778	0.049	0.001	0.373
Typical DU	IMBA ^a	0.402	0.062	0.004	0.0002	0.335
RU (1% ²³⁵ U)	Hanford ^c	0.910	0.563	0.023	0.048	0.365

a. IMBA = Integrated Modules for Bioassay Analysis software

3.1.1 <u>Uranium Air Sampling</u>

This discussion focuses on documented air-sampling data from two separate Health and Safety Laboratory (HASL) reports by the AEC. The first air sampling was performed at W.R. Grace during the recovery of 93% HEU from uranium-aluminum alloy scrap in the storage, the ceramics, and chemical buildings (AEC 1959). The second air sampling was performed during the recovery of 3.6% LEU from slag scrap (AEC 1961). Table 3-3 to 3-8 lists the results of the sampling analyses.

Table 3-3. Occupational exposures for 93% uranium-aluminum alloy recovery. a

	Number of	Average daily weighted
Operator	persons	exposures in dpm/m ³
Column operator	6	31
Ceramic building operator	3	45
Charge makeup	3	14
Accountability	3	15
Chemist and Technicians	8	3.1
Spectrographic Operator and Technician	3	2.5
Machine shop	5	12
Laundry	1	7
Geometric mean ^b		10.84
Geometric standard deviation ^b		2.76
95% confidence level value ^b		57.63
90% confidence level value ^b		39.84
50% confidence level value ^b		10.84

a. Data from AEC (1959).

The air samples consisted of collection on filters of radioactive particulates from breathing zones and general areas during processing. The alpha activity measured on the filter was used to determine the airborne alpha activity concentrations. When multiple samples at a location were collected, the AEC used the mean air concentration in subsequent calculations. The AEC matched air concentration

b. HPS N13.22 (ANSI 1995).

c. ORAUT (2004a).

b. Statistics calculated using a log normal distribution and the program Crystal Ball.

Table 3-4. Average breathing-zone samples for 93% uranium-aluminum alloy recovery.^a

Table 6 1. Average breathing 20116 dampies for 6070 dramam	Number of	,
Operation		Average concentration dpm/m ³
Operation	samples	
Shearing U-Al alloy for charge makeup and weighing	3	43
Charging digester with batch of U-Al alloy	2	170
Running material from digester thru filter press and column	3	50
Cleaning residue from filter press	2	19
Securing O.K. Liquor from columns	1	65
Precipitation of O.K. Liquor	2	16
Filtering ppt. on Buchner funnel	2	15
Removing filter cake-placing in tray	1	1
Transferring tray from furnace to cooling area	2	65
Transferring tray from furnace to dry box, weighing and unloading		
tray in dry box, cleaning residue tray	3	280
Digestion of organic ashes in hood	1	1
Filtration of digested organic ashes	1	65
Geometric mean ^b		26.65
Geometric standard deviation ^b		5.84
95% confidence level value ^b		485.13
90% confidence level value ^b		255.57
50% confidence level value ^b		26.65

a. Data from (AEC 1959).

Table 3-5. Average general air-sampling concentrations for 93% uranium-aluminum alloy recovery.^a

Area	Number of samples	Average concentration dpm/m ³
Shearing and Weighing	2	9
Solvent extraction area	6	19
Chemical building - oven area	3	23
Ceramics building	9	2
Machine shop	4	16
Wet chemistry lab	3	3
Spectrographic lab	3	2
Laundry	3	6
Lunch room	3	7
Clean locker room	3	2
Contaminated locker room	3	4
Geometric mean ^b		5.81
Geometric standard deviation ^b		2.52
95% confidence level value ^b		26.55
90% confidence level value ^b		18.98
50% confidence level value ^b		5.81

a. Data from AEC (1959).

determinations with information about worker categories, locations, tasks, and time at each location or task.

Although it is unlikely that workers would have been exposed to the same air concentrations from the many other processes that were conducted at NFS, this is the only air sample information currently available. In addition, changes in the processes and the safety controls could have resulted in both increased and decreased exposure for any given period. In this document an estimate of the intake

b. Statistics calculated using a log normal distribution and the program Crystal Ball.

b. Statistics calculated using a log normal distribution and the program Crystal Ball.

Table 3-6. Occupational exposures for 3.6% uranium recovery from slag.^a

Operator	Number of persons	Average daily weighted exposures in dpm/m ³
Williams Roll Mill	3	170
Digestion	3	45
Solvent extraction	3	10
ADU precipitation	3	45
ADU oxide (ceramics)	3	17
Micropulverizer-drifter	3	71
Accountability-shipping and receiving	2	22
Laundry	1	9.4
Janitor	1	37
Health physics technician	2	20
Geometric mean ^b		30.72
Geometric standard deviation ^b		2.46
95% confidence level value ^b		132.11
90% confidence level value ^b		95.55
50% confidence level value ^b		30.22

a. Data from AEC (1961).

Table 3-7. Average breathing-zone samples for 3.6% uranium recovery from slag.^a

Operation	Number of samples	Average concentration dpm/m ³
Operating jaw crusher	4	230
Loading top of Williams roll mill with crushed slag	3	500
Removing drum of 325 mesh MgF2 from hopper	2	70
Loading and unloading pot (oxidation furnace) with slag metal		
heavies	2	140
Charging digester with slag	3	150
Cleaning ADU from plate and frame filter press	3	130
Placing tray of ADU inside of oven	2	84
Removing tray of oxide from furnace-placing inside of glove		
box and transferring U ₃ O ₈	4	97
Micropulverizing U ₃ O ₈ (bag not working properly)	3	590
Geometric mean ^b		167.83
Geometric standard deviation ^b		2.12
95% confidence level value ^b	·	578.38
90% confidence level value ^b		440.08
50% confidence level value ^b		167.83

a. Data from AEC)1961).

was made by calculating the 95th percentile of the maximum average breathing-zone concentration for the 1961 AEC air sample results (from Table 3-7 later in this section) by assuming a lognormal distribution of the data in the AEC report (AEC 1961). Using Crystal ball, a lognormal distribution calculation yielded a value of 578 dpm/m³ with a geometric mean of 167 dpm/m³ and a geometric standard deviation (GSD) of 2.12. The calculated 95th-percentile air concentration was used to calculate upper estimates of internal exposures. The most conservative air concentrations were generated from the radiological task-oriented information in Table 3-7. If actual operator information from Table 3-3 and 3-6 were to be used, the calculated intakes would be about a factor of 4 to 10 less. Task-oriented weighted average samples involve the highest air concentrations encountered because of the closest proximity to the actual work and no accounting of the time away from the

b. Statistics calculated using a log normal distribution and the program Crystal Ball.

b. Statistics calculated using a log normal distribution and the program Crystal Ball.

Table 3-8. Average general air-sampling concentrations for 3.6% uranium recovery from slag.^a

Area	Number of samples	Average concentration dpm/m ³
Crushing room	5	110
Digester area	6	11
Leach area	3	20
Solvent extraction area - all levels	8	6.4
Boil-down feed tank area	3	17
O. K. liquor storage	3	9
Raffinate storage area	3	9
Neutralizer tank storage	4	10
ADU precipitation tanks	3	16
ADU filter area	4	17
U ₃ O ₈ transfer hood area	3	11
Micropulverizer area	4	62
Accountability room	5	21
Chemistry lab	4	4.3
Spectrographic lab	2	9.3
Research and development lab	4	6.6
Laundry	4	2.3
Locker room - clean side	2	17
Locker room - dirty side	2	45
Lunch room	4	21
Health physics room	2	1.5
Geometric mean ^b		12.57
Geometric standard deviation ^b		2.7
95% confidence level value ^b		64.52
90% confidence level value ^b		44.95
50% confidence level value ^b		12.57

- a. Data from AEC (1961).
- Statistics calculated using a log normal distribution and the program Crystal Ball.

actual or lower exposure rate activities. The use of the task-oriented time-weighted average air concentrations is claimant favorable.

As seen in Table 3-3, the uranium exposure by occupation differed with the column and ceramic building operators with the largest daily weighted exposures. This exposure variability between occupations is also seen in Table 3-6, where the Williams roll mill operators were clearly the highest exposed group, with the micropulverizer-drifter, digestion and ammonium diuranate (ADU) precipitation operators being the next highest groups.

The general air sample results in Tables 3-5 and 3-8 result in the lowest calculated intakes. This is to be expected because general air samples are typically lower than breathing-zone samples. Bioassay results are expected to be at levels between breathing-zone and general air sample results.

The breathing rate is based on the default for light work, 1.2 m³/hr, as indicated in ICRP Publication 66 (ICRP 1994a). This category assumes an activity distribution of one-third sitting and two-thirds light exercise. The intakes in picocuries are calculated by dividing the 95th percentile of the air concentration (578 dpm/m³) by 2.22 dpm/pCi and multiplying this result by the breathing rate and the assumed number of hours exposed at a given concentration. The organ doses are assumed to be a constant distribution. Several assumptions included in the intake and dose reconstruction are likely to be overestimating assumptions. This includes the use of a lognormal distribution, the 95% confidence

level concentration, the task-related as opposed to the occupation time-weighted average, and the assumption of constant work activity and worker exposure during the entire work period.

Air sampling for 233 U has also been identified. Airborne concentrations for enriched UO₂ decladding and dissolution of 233 U₃O₈ pellets from the immediate work area resulted in an average concentration of less than 1.8% of the maximum air concentration (1 × 10⁻¹⁰ μ Ci/ml) (Householder 1963b). This would result in calculations of intake less than that calculated above.

In the case where inhalation intakes are calculated from air concentrations, ingestion intakes are also to be considered. NIOSH (2004) indicates that the ingestion rate, in terms of pCi for an 8-hour workday, can be estimated by multiplying the air concentrations in pCi per cubic meter by a factor of 0.2. The uranium ingestion rate based on air concentration of 260 pCi/m³ would be 52 pCi/workday. The daily inhalation and ingestion intake rate from LEU recovery is estimated from the 95 % confidence level air concentration listed in Table 3-7. Table 3-9 lists the inhalation intake rate per year and AEC operational time period. Table 3-10 lists the ingestion intake rate by year and AEC operational time period. Note, bioassay data should be used first and not the intakes calculated based on air sampling results unless the bioassay data is deficient. Most potentially exposed workers at WR Grace/NFS were monitored. Some reports in the 1970s exist that indicate skin contamination was a recurrent issue at W.R. Grace/NFS. The contamination levels indicated in the claimant records should be bounded by the assumption of 10 % of the skin contamination equilibrium activity levels being ingested as per NIOSH 2004, "Estimation of Ingestion Intakes". A study was done at the Oak Ridge Gaseous Diffusion Plant to determine the intake from hand contamination. The study indicated that the amount of uranium transferred from the hand to the cigarette while smoking was approximately 1% of the material on the surface of the hand. (Bailey 1958) The ingestion calculation included both contamination of food or drink from contaminated air settling and the transfer from contaminated surfaces to the hands to food or drink on a chronic basis.

Table 3-9. Estimated uranium inhalation intake rates based on time weighted air concentrations measured during uranium recovery operations.

Work period	Number of years	Number of potential AEC work hours per work period	Air concentration (pCi/m³)	Breathing rate (m³/hr)	Intake (pCi)
1/1957-12/31/1970	14	28,000	260	1.2	8.73x10 ⁶
1 yr	1	2,000	260	1.2	6.24x10 ⁵

Table 3-10. Estimated uranium ingestion rates based on time weighted air concentrations measured during uranium recovery operations.

Work period	Number of years	Number of potential AEC work days per work period	Ingestion rate pCi/workday	Intake (pCi)
1/1957-12/31/1970	14	3,500	52	1.82×10^{5}
1 yr	1	250	52	1.30×10^4

a. Ingestion values were calculated according to NIOSH 2004. Choose the same f₁-value as used for inhalation in accordance with NIOSH (2004).

3.1.2 <u>Enriched and Recycled Uranium</u>

Because the AEC air samples were counted with alpha detectors, which detect radioactivity rather than mass, there is no need to adjust measured air concentration results for assumed uranium enrichment. AEC air sample results taken during the recovery of HEU were below the concentrations measured during the recovery of LEU. Because various levels of enrichment occurred during the

processing and recovery operations of uranium, this document assumes that intakes are U²³⁴ for the purpose of internal dose calculation.

Only the uranium intake should be assigned for facilities with exposure only to uranium (or natural thorium), except when exposure was to recycled uranium (RU). For RU, the dose reconstructor should add the intakes from Table 3-11 (ORAUT 2005b).

Table 3-11. Intakes of contaminants in recycled uranium as fractions of uranium intake.^a

Radionuclide	Activity fraction of contaminant (e.g., pCi X/pCi U)
²³⁴ U	1.0
²³⁸ Pu	0.06
²³⁷ Np	0.005
⁹⁹ Tc	0.4
²³² Th	0.02
²²⁸ Th	0.02
106Ru	0.04

a. Source: Based on ORAUT 2005b.

3.1.3 <u>Uranium Bioassay</u>

The highest recorded uranium in urine result for W.R. Grace/NFS from claimant records was 6229 dpm/L recorded in 1967. Urinalysis for uranium started in October of 1964. Samples were sent to Eberline until about 1974. From about that time, on-site analysis was performed. Details of the analysis are not known. The recording level was 1 dpm/L at the 2-sigma or 95% confidence level as indicated from the review of employee documents as indicated from the NFS Health Physics department. (Tester 2005) This is likely the assumed decision level. The minimum detectable level should be twice the critical level or 2 dpm/L. Some random samples were sent off the site for fecal analysis of uranium and urine analysis for enriched uranium. Some bioassay records received from W.R. Grace/NFS have some of the radiometric uranium bioassay results with a minimum recording level of <10 dpm/L. Other records have bioassay results reported on two forms, one hand written form indicating laboratory results with actual results recorded down to 1.0 dpm/L, and another typed report in which the lowest values are recorded as <10 dpm/L, i.e., all values less than 10 dpm/L were recorded as <10 dpm/L on the typed form. The 10 dpm/L recording limit should be considered the MDA for results for which the actual lab recorded result is not available. When bioassay data is available, dose reconstructors should use bioassay data for estimating worker intakes. Intakes of the contaminants listed in Table 3-11 should be added based on the given ratios.

A summary table of the highest recorded values compiled from a review of claimant records is presented below in Table 3-12. Most of the highest recorded doses occurred during the AEC contract years of 1957 through 1970. Most of the urine results that greatly exceeded 100 dpm/L decreased to less than 100 dpm/L within several days. This included the highest values listed in Table 3-12. The highest values recorded from 1964 though 1970 were used to estimate the intake of uranium.

Lung counts were performed from 1970 to the present for uranium as well as plutonium and neptunium. Lung counts are in general not as reliable as urinalysis for routine monitoring. However, this monitoring was routine and was used to assess routine exposures to transuranic and fission products and to further analyze results from accidental intakes. The MDA for ²³⁵U was about 120 µg as indicated from the cursory review of claimant documents, which is a reasonable default MDA value. Actual MDAs from workers records should be used when available.

3.2 **PLUTONIUM EXPOSURES**

Many forms of plutonium were possible over the years including metal and oxide. Because not enough information for the recovery or manufacture of plutonium was found, the exact chemical forms are not known. It is possible that MOX fuel of a plutonium and thorium mixture was processed at some point because UO₂ and ThO₂ mixtures were produced on the site and plutonium production capacity existed at the same time. Most of the manufacture of plutonium occurred in Buildings 234A, 234B, and 234C.

> Table 3-12. Uranium urinalysis recorded results for claims submitted as of 10/01/05 in dpm/L

_	Highest	Second highest	
Sample year ^a	result	result	Comment
1964	334	238	
1965	725	467	Rate < 100 in 2 to 8 weeks
1966	1,480	122	Rate < 100 in < 1 week
1967	80	47	
1968	80	76	
1969	135	134	Rate < 100 in < 1 week
1970	234	112	Rate < 100 in < 1 week
1971	56	53	
1972	133	63	Rate < 100 in < 1 week
1973	435	46	Rate < 100 in < 1 week
1974	66	30	
1975	1,420	60	Rate < 100 in < 1 week
1976	75	35	
1977	332	89	Rate < 100 in < 1 week
1978	55	21	
1979	99	77	
1980	2,427	2,129	Rate < 100 in < 1 week
1981	70	67	
1982	6,229	5,423	Rate < 100 in < 1 week
1983	221	150	Rate < 100 in < 1 week
1984	94	68	
1985	105	52	Rate < 100 in < 1 week
1986	229	127	Rate < 100 in < 1 week
1987	60	58	
1988	15	10	
1989	89	28	
1990	19	20	
1991	17	10	
1992	67	63	

a. All data from claims filed as of 11/30/05.

In general, plutonium oxides, carbides, and hydroxides are absorption type S; nitrates and other compounds are type M (ICRP 1995). Older materials, even when starting out as soluble, can have a tendency to oxidize when left in contact with air. Oxides, metals, and old contamination should be treated as type S. If nothing is known about the chemical form of plutonium, then either type M or S can be used to maximize the dose to the organ of concern. Americium-241 is a component of plutonium contamination and should be modeled in the lung the same as the plutonium matrix in which it has grown. In other words, the americium should be treated as absorption type S if the plutonium is type S (ORAUT 2004a).

There are essentially three types of plutonium-based material. Reactor grade, fuel grade and weapons grade with fuel grade falling in between reactor and weapons grade. For this section, lacking any specific information on the actual composition of the processed plutonium, an assumption of 10-yr-old fuel-grade plutonium is claimant-favorable and reasonable (Table 3-16). This is noted in the Hanford site profile: Hanford processed much of the DOE complex plutonium.

3.2.1 **Plutonium Organ Dose Calculations**

The isotopic breakdown of plutonium mixtures in this TBD assumes the composition as listed in Table 3-13 which is from the Hanford reference fuel grade plutonium aged for 10 years. The assumption of 10 year aged fuel should be claimant favorable. If the intake level for plutonium was set at the uranium value of 6.24 x 10⁵ pCi/yr, as estimated from uranium time weighted air samples, the annual dose from the above 10-yr fuel-grade plutonium of a 12% mixture absorption type M would result in the organ doses listed in Table 3-14. Five-yr fuel-grade plutonium is included as a comparison. Table 3-14 is for illustrative purposes only and indicates the possible maximally exposed organs.

Table 3-13. Activity composition of Hanford reference fuel-grade

plutonium mixture (12%).a

Mixture designation	Specific activity ^a	Fraction of activity	Type S⁵ pCi/yr	Type M ^c pCi/yr
Total activity/yr			1.50E+06	6.24E+05
²³⁸ Pu	1.58E-02	7.73E-03	1.16E+04	4.82E+03
²³⁹ Pu	5.26E-02	2.57E-02	3.86E+04	1.60E+04
²⁴⁰ Pu	2.72E-02	1.33E-02	2.00E+04	8.30E+03
²⁴¹ Pu	1.91E+00	9.34E-01	1.40E+06	5.83E+05
²⁴² Pu	3.93E-06	1.92E-06	2.88E+00	1.20E+00
²⁴¹ Am	3.89E-02	1.90E-02	2.85E+04	1.19E+04

- Source: ORAUT (2004a).
- b. Calculated form urinalysis data.
- c. Calculated form Uranium air sample data.

3.2.2 **Plutonium Bioassay**

Plutonium-239 was analyzed in urine from about 1967 to 1980. Some Pu bioassay results are also available from the 1980s and 1990s. Some bioassay records indicate that the later results are from decommissioning work. The minimum detectable concentration (MDC), analytical methodology, or frequency is not known. The lowest positive reported level was 0.03 dpm/L (0.014 pCi/L) on February 5, 1967, by NFS. The highest reported level was 2.04 dpm/L (0.92 pCi/L) on October 03, 1967, by NFS. Forty six of the 81 recorded values were zero. Thirty five of the 81 recorded values are listed in Table 3-15. From the worker internal dose records, the frequency of the bioassay could have been about twice per year but no pattern could be determined. The years of plutonium exposures likely occurred from 1967 though February of 1973. (Claimant records and Congress 1986) Most radiation worker records had extensive uranium urinalysis from 1964 to 1989 that indicated intermittent and rare plutonium urinalysis, in comparison.

Since historical detection limits for W.R. Grace are not available, a review of detection limits for the 1960s and 1970s at other AEC sites was made. The internal dosimetry section of the DOE Hanford site profile (ORAUT 2004a) reports an MDA of 0.05 dpm/sample. The Savannah River site profile (ORAUT 2005e) has a plutonium recording level and MDA of 0.1 dpm/1.5 L. These values are reasonably consistent with the observed reporting limits from W.R. Grace. From a review of bioassay records, the lowest observed nonzero recorded plutonium results at WR Grace was 0.03 dpm/L.

although some results are reported in units of dpm/sample, with a given sample volume. If additional information on detection capabilities is not available from the records, dose reconstructors should assume a decision level of 0.03 dpm/L and an MDA of 0.06 dpm/L.

Lung counting for ²³⁹Pu started at W.R. Grace in 1990. A germanium detector system had an ability to detect ²³⁹Pu but had a variable minimum detectable activity (MDA) of about 168 nCi (July 6, 1989) to 481 nCi (October 22, 1987). The earlier Helgesson counting system likely could have been used for plutonium lung counting but was used only for uranium lung counting from about 1970. The detection limits are based on a cursory review of claimant files. Most results were non-detections or perhaps near the MDA. Urinalysis records are more useful for dose reconstruction purposes than lung counting because of the high MDAs of the lung counters.

Table 3-14. Organ dose for 1 yr for Hanford reference 10-yr-aged fuel-grade plutonium mixture (12%) and Hanford reference 5-yr-aged fuel-grade plutonium mixture (6%) for an annual chronic intake of 6.24×10^5 pCi/yr and absorption type M.^{a,b}

	1-yr organ dose in rem for	1-yr organ dose in rem for
Description	10-yr plutonium fuel grade	5-yr plutonium fuel grade
Adrenals	7.37E-03	5.63E-03
Bladder wall	7.37E-03	5.63E-03
Bone surface	3.66E+00	2.65E+00
Brain	7.37E-03	5.63E-03
Breast	7.37E-03	5.63E-03
Esophagus	7.37E-03	5.63E-03
ST wall	7.51E-03	5.73E-03
SI wall	7.58E-03	5.77E-03
ULI wall	8.71E-03	6.58E-03
LLI wall	1.11E-02	8.31E-03
Colon	9.76E-03	7.33E-03
Kidneys	8.10E-02	5.76E-02
Liver	7.70E-01	5.15E-01
Muscle	7.37E-03	5.63E-03
Ovaries	4.23E-02	2.91E-02
Pancreas	7.37E-03	5.63E-03
Red marrow	3.49E-01	2.57E-01
ET airways	1.84E+00	1.30E+00
Lungs	3.08E+00	2.14E+00
Skin	7.37E-03	5.63E-03
Spleen	7.37E-03	5.63E-03
Testes	4.28E-02	2.96E-02
Thymus	7.37E-03	5.63E-03
Thyroid	7.37E-03	5.63E-03
Uterus	7.37E-03	5.63E-03
Remainder	9.05E-03	6.82E-03
Effective dose	4.94E-01	3.46E-01

a. The table is for illustrative purposes only. It indicates the possible maximally exposed organs.

Major plutonium processing sites that had ample air and bioassay sampling data can demonstrate the plutonium exposures that occurred in the DOE/AWE sites. In Attachment B of ORAUT (2005b), plutonium urine samples from Rocky Flats and Hanford for 1953 to 1969 were evaluated. Even though the median sample varied for most years, the value was less than 0.023 pCi/d. The Rocky

b. ICRP 2000, ICRP DCF database code.

Flats/Hanford site analysis concluded that the assumption of 3,300 pCi/yr to which an individual could be exposed to type M or S characteristics is valid because these are the limiting absorption types for systemic organs. However, for non-systemic organs such as the lungs or respiratory tract for type S, this may not be the case. Table 3-15 lists all of the positive plutonium urinalysis results obtained from claimant files submitted by October 2005.

Table 3-15. Plutonium urinalysis results for claims.^a

Ciairis.		Urinalysis in
Sample number ^a	Date	pCi/day
1	7/28/1968	0.08
2	8/23/1970	1.29
3 ^b	10/3/1967	0.19
4 ^b	10/11/1967	0.20
5 ^b	11/10/1967	0.12
6 ^b	11/19/1967	0.04
7 ^b	12/13/1967	0.21
8 ^b	3/3/1968	0.03
9 ^b	3/29/1968	0.03
10	2/15/1973	0.02
11	12/5/1967	0.29
12	2/19/1969	0.02
13	12/5/1967	0.12
14	2/21/1969	0.16
15	12/13/1967	0.03
16	10/7/1971	0.10
17	10/16/1968	0.02
18	2/4/1967	0.06
19	5/20/1969	0.12
20	8/1/1969	0.04
21	12/7/1967	0.06
22	8/6/1968	0.44
23	8/6/1968	0.16
24	10/8/1967	0.04
25	10/15/1968	0.18
26	10/5/1967	0.03
27	11/3/1967	0.08
28	2/20/1969	0.04
29	12/7/1967	0.11
30	2/5/1968	0.73
31	8/5/1968	0.07
32	9/7/1967	0.10
33	11/10/1967	0.08
34	12/28/1967	0.06
35	12/5/1967	0.08

a. All data from claims filed as of 11/30/05.

b. These urinalysis results are from one claimant which also had the highest recorded excretion rate.

3.3 **THORIUM EXPOSURES**

Thorium oxides, carbides, and hydroxides are absorption type S; nitrates and other compounds are type M. In the absence of specific information, the dose reconstructor should assume either type M or S whichever is more claimant-favorable. There is no air-sampling or urinalysis information available for the W.R. Grace facility for thorium. Therefore, either the gross alpha count data for early uranium air sampling from HASL reports 1959 and 1961, the urinalysis for uranium or the lung counts for uranium were used to estimate the annual intake. The most claimant favorable results will be used to estimate the thorium intake. Assuming equilibrium between ²³²Th and ²²⁸Th would be claimant favorable.

There were no dedicated facilities for thorium processing thorium operations utilized the same facilities as uranium operations. The low enriched uranium operations air sampling data is bounding for natural thorium. To account for unmonitored thorium exposures at the W.R. Grace/NFS plant it is assumed that the thorium intake is equal to the uranium intake by mass. Natural uranium has a lower specific activity than enriched uranium, so it is claimant favorable to assume natural uranium when determining the relative activity of thorium. To determine the relative activities of uranium to thorium, the specific activity of 232 Th (1.1 x 10 $^{-7}$ Ci/g) is divided by the specific activity of natural uranium (6.83 \times 10⁻⁷ Ci/g). This results in a relative ²³²Th-to-uranium intake fraction by activity of 0.161. Exposure from ²²⁶Ra (half-life of 5.75 years) is assumed to be insignificant because the thorium was likely to have been recently produced and because the dose conversion factor is small compared to thorium.

Thorium oxide as ThO_2 and uranium oxide as $^{233}UO_2$ were the finished products for the production of $^{233}U/^{232/228}Th$ MOX fuel. The ^{233}U was received from Oak Ridge in the form of uranyl nitrate, then precipitated to (NH₄)₂-U₂O₇ (ADU), then calcined to UO₂. Blending, pressing, and grinding were also a part of the process in addition to chemical processing. Particle sizes did vary. Because of criticality concerns. 4 kg of material could only be processed at one time.

Usually tens of kilograms were processed per feasibility report or campaign. No enrichment was stated in the feasibility report, but the ZrO₂-²³³UO₂ pellet feasibility report (Housholder 1963b) mentioned 93% enrichment.

The combination of thorium and uranium oxides ($^{232/228}$ ThO₂ and 233 UO₂) follows the following matrix: 1% by weight 233 UO₂ / 99% by weight, $^{232/228}$ ThO₂ with a breakdown of 50% by activity 232 Th and 50% by activity 228 Th, and assuming equilibrium. It is not known if this material had aged substantially but ^{232,/228}Th/²³³U fuel can have considerable in-growth of many radionuclides because of the short half lives of their daughters.

There may have been several $^{232/228}$ Th lung counts performed. These counts were likely below the MDAs of the time. A reasonable MDA for 232 Th is 30 nCi. (ORNL 2002)

Radon-220 from the decay of natural thorium is a potential source of dose to the lungs.

3.4 **URANIUM AND PLUTONIUM DIOXIDE- MOX, MIXED OXIDE FUEL EXPOSURES**

In this document, the proposed MOX fuel consists of 20% by weight of 3.5%-enriched ²³⁵UO₂ and 80% by weight of Hanford reference 10-yr-aged fuel-grade plutonium mixture (12%) PuO₂. There is currently no specific process or operational information on MOX fuel recycling at W.R. Grace.

3.5 AMERICIUM

Americium is type M for all compounds. Note, the IMBA code can calculate type S intakes and thus the recommendation stated in section 3.2 of assigning ²⁴¹Am as intake type S if the primary radionuclide is type S can be followed if deemed necessary.

There may have been several ²⁴¹Am lung counts performed. These counts were likely below the MDAs of the time. For example, a lung count performed in 1981 yielded a recorded result of 0.11 nCi but the reasonable MDA at the time was between 0.3 to 0.9 nCi. The current (2005) MDA would be about 0.1 nCi (ORAUT 2004f)

3.6 OCCUPATIONAL INTERNAL DOSE RECONSTRUCTION SUMMARY AND ASSUMPTIONS - RESERVED

4.0 ESTIMATION OF EXTERNAL EXPOSURE

During operations at the facility, occupational exposure occurred from handling received material, standing near stored fissile material (either as feed or product), and airborne radioactivity with the resultant buildup of surface contamination. The fissile material was enriched UF₆ arriving in approved shipping and storage cylinders, solid fissile material in various forms (liquid, powder, or metal) to be converted into or made into nuclear fuel, or scrap material containing LEU or HEU uranium.

4.1 RECORDED OCCUPATIONAL EXPOSURE AND EXTERNAL DOSIMETRY

Beginning in 1957, Chicago Nuclear supplied all dosimetry badges and performed the necessary calibrations. Landauer supplied all dosimetry beginning in 1961 using control film. The personnel monitoring reporting were normally in net exposure, and the control film reading was deducted from the personnel film reading. If the control film appeared to have been exposed differently from the personnel packets, the densities on the personnel film were normalized to Landauer controls only, and a non- control reading was reported. Minimal beta or soft X-ray skin dose readings were not reported until after a positive skin dose exposure was recorded (Koperski 2004). In 1989 Landauer supplied W.R. Grace/NFS with thermal luminescent dosimetry (TLD).

External radiation exposure recording at WR Grace/NFS utilized the U.S. AEC form AEC 5 similar to the NRC form 5 of today. Gamma, beta and neutron exposures are listed in separate columns. In 1961, Landauer started to report the external dose in the same format as the AEC form 5. In 1964 Landauer still reported the gamma, beta and neutron exposures in separate columns but had the same forms filled out separately for whole body, skin and extremities. Code 1 was for the whole body, code 2 for the skin of the whole body and code 3 for the hands and forearms. Up to December 31, 1969, the reported skin dose represents only the shallow dose component (i.e., it does not include the penetrating gamma component). From January 1 1970, the reported skin exposure included the penetrating gamma component. For this reason, the shallow component should be calculated as the difference between the reported skin and total body dose.

In addition, there was forearm and hand (extremity) monitoring at W.R. Grace/NFS from at least the 1960s to the 1970s. The MDL information in Table 4-1 for wrist (forearm) and finger (hand or extremity) is from Landauer. Ring and wrist badges were calibrated only for high-energy gamma (for ¹³⁷Cs at 0.662 MeV) and high-energy beta (1.5 MeV) unless special arrangements were made with the Plant (Koperski 2004) MDL information for photon and beta whole body monitoring are also listed

in Table 4-1. The missed dose listed in Table 4-1 are considered default values and are to used if the available claimant dosimetry records are incomplete. The dosimetry used is further described in Table 4-2.

Table 4-1. Minimum detectable level (MDL) and maximum potential missed photon or beta dose.^a

Dosimeter	Period of use	MDL (rem)	Max annual missed dose (rem)
Nuclear Chicago film - whole body	1957-5/1959 ^b	0.01 photons 0.01 beta	0.26 beta -photons (every 2 weeks)
Nuclear Chicago film - whole body	6/1959-12/1960 ^b	0.01 photons 0.01 beta	0.12 beta -photons (monthly)
Landauer film- whole body	1/1961 – 12/1963 ^b	0.01 photons 0.01 beta	0.12 beta -photons (monthly)
Landauer film- whole body	1/1964 – 12/1988 ^c	0.01 photons 0.01 beta	0.12 beta -photons (monthly)
Landauer TLD- whole body	1/1989 – 12/1998 ^c	0.01 photons 0.01 beta	0.04 beta -photons (quarterly)
Landauer OSL- whole body	1/1/1999-12/2004 ^c	0.001 photons 0.001 beta	0.004 beta -photons (quarterly)
Landauer TLD- whole body	1/2005 – present ^c	0.01 photons 0.01 beta	0.04 beta -photons (quarterly)
Film-badge- wrist	1957- June 1974 ^d	0.04 photons 0.04 beta	0.24 beta-photons (monthly)
Landauer Type M - wrist	July 1974-About 1983 ^c	0.02 photons 0.04 beta	0.12 photons (monthly) 0.24 beta (monthly)
G5 wrist film badge – wrist	About 1983 – 1990 ^c	0.02 photons 0.04 beta	0.12 photons (monthly) 0.04 photons (quarterly) 0.24 beta (monthly) 0.08 beta (quarterly)
K5 TLD wrist	1991- present ^c	0.01 photons 0.04 beta	0.06 photons (monthly) 0.24 beta (monthly)
Film badge- finger	1957-1982°	0.04 photons 0.04 beta	0.24 beta-photons (monthly)
U3 TLD (LiF) - finger ring	About 1983 – present ^c	0.03 photons 0.04 beta	0.18 photons (monthly) 0.08 photons (quarterly) 0.24 beta (monthly) 0.08 beta (quarterly)

a. TLD = thermoluminescent dosimeter.

4.1.1 NRC Annual Reports

The NRC has reported on the annual occupational radiation exposure for nuclear power reactors since 1974. For facilities like W.R. Grace, consistent annual reporting could not be found in NRC records until 1982. The only information from before 1982 was for whole-body exposures in excess of 1.25 rem for employees with employment of less than 90 d. The reporting format varied over the years. In the early years, only the number of monitored individuals and the measurable dose, collective dose, and average measurable doses in rem were provided. In later years, the annual whole-body doses were segregated into dose bins with the number of workers in each bin. Tables 4-3 and 4-4 summarize the results of the research into NRC records.

b. MDLs from Tester (2005).

c. Koperski (2004). MDL information as communicated by Landauer Inc.

Table 4-2. Dosimetry for external whole-body, wrist, and extremity exposures.

Period	Monitoring technique	Dosimeter description
	Beta/pho	oton dosimeters
1957 – June 1974 whole body	Photographic film badge	Nuclear-Chicago or similar film badges. Nuclear-Chicago film badge contained single film packet. Three filters (front and back) were incorporated into film badge for energy dependence: cadmium, aluminum, and lead.
July 1974 – About 1983* whole body	Landauer J (beta and gamma)	Type J dosimetry were film badges. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.
About 1974* – April 1990 whole body	Landauer G1	Film emulsion packaged placed in standard Gardray holder/badge for monitoring beta, X-ray, and gamma exposure. Insensitive to neutron radiation. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.
May 1990 – present whole body	Landauer Z1 dosimeter	Comprised of 3 TLD-700 chips for monitoring beta, X-ray and gamma exposure. Insensitive to neutron radiation. Replaced Landauer G1.
July 1974 – About 1983* wrist	Landauer Type M (wrist beta-gamma) badges.	Type M dosimetry was a film badge. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.
About 1983* – 1990 wrist	G5 wrist film badge	Responded to beta, X-ray, and gamma exposure to provide data on extremity dose. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.
1991 – present wrist	K5 TLD wrist badge	Comprised of 3 TLD-100 chips.
1957-1974	Film badge- <i>finger</i>	Nuclear-Chicago or similar film badges. Nuclear-Chicago film badge contained single film packet. Three filters (front and back) were incorporated into film badge for energy dependence: cadmium, aluminum, and lead.
About 1983* – present finger ring	U3 TLD (LiF)	Responded to beta, X-ray, and gamma exposure to provide data on extremity dose. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.

^{*} This analysis found no documentation that shows the start of G1, U3, G5, I8, and E1 dosimeter use and the end of Type K, J, and M badges use.

Table 4-3. Annual occupational radiation exposures for 1982 to 1994.

	Total workers	Workers with measurable	Collective dose	Average measurable	
Year	monitored	dose	(person-rem)	dose (rem)	Reference
1982	1161	1014	59.77	0.06	NUREG-0714 Vol. 4 and 5 ^a
1983	994	591	35.37	0.06	
1984	904	626	37	0.06	NUREG-0713 Vol. 6 ^b
1985	871	562	28	0.05	NUREG-0713 Vol. 7 ^b
1986	996	389	22	0.06	NUREG-0713 Vol. 8 ^b
1987	3559	367	20	0.05	NUREG-0713 Vol. 9 ^b
1988	5516	627	32	0.05	NUREG-0713 Vol. 10 ^b
1989	6136	698	40.525	0.06	NUREG-0713 Vol. 11 ^b
1990	7373	739	46.025	0.06	NUREG-0713 Vol. 12 ^b
1991	4347	830	41.750	0.05	NUREG-0713 Vol. 13 ^b
1992	1,423	1,312	177.825	0.14	NUREG-0713 Vol. 14 ^b
1993	3,373	323	78.650	0.24	NUREG-0713 Vol. 15 ^b
1994	610	484	31.945	0.07	NUREG-0713 Vol. 16 ^b

a. NRC (1985).

Document No. ORAUT-TKBS-0043	Revision No. 00	Effective Date: 02/14/2006	Page 30 of 47

b. NRC (1985-2003).

Effective Date: 02/14/2006

Table 4-4. Annual occupational TEDE for 1995 to 2002.^a

	N	umber	of Indi	viduals	with v	whole b	ody d	oses i	n the	range	s (rem)		No.	Total	Ave.	
														with	collective	meas.	
	No.		0.1-	0.25 -	0.5 –	0.75 –	1.0 –	2.0 -	3.0 –	4.0 –	5.0 -		Total no.	meas.	TEDE	TEDE	NUREG-0713 ^b
Year	meas.	<0.1	0.25	0.5	0.75	1.0	2.0	3.0	4.0	5.0	12.0	>12.0	monitored	dose	(rem)	(rem)	volume no.
1995	192	166	12	16	8	0	0	0	0	0	0	0	392	200	15.185	0.08	Vol. 17
1996	178	195	26	26	7	3	0	0	0	0	0	0	435	257	24.937	0.097	Vol. 18
1997	180	323	38	27	8	0	4	0	0	0	0	0	578	396	33.656	0.065	Vol. 19
1998	126	387	66	45	16	13	7	0	0	0	0	0	660	534	64.951	0.122	Vol. 20
1999	58	559	68	38	15	2	0	0	0	0	0	0	740	682	46.227	0.068	Vol. 21
2000	155	377	62	55	15	4	8	0	0	0	0	0	676	521	60.703	0.177	Vol. 22
2001	70	498	104	64	21	10	12	2	0	0	0	0	781	711	95.599	0.134	Vol. 23
2002	92	628	159	92	20	8	11	3	0	0	0	0	1,013	921	115.848	0.126	Vol. 24

a. TEDE = total effective dose equivalent.b. NRC (1985–2003).

4.1.2 **Penetrating Dose Determination and Deep Dose Calculations**

The majority of photons from natural uranium metals and solutions are in the 30 to 250 keV energy range. As uranium becomes more enriched or less enriched, the photon spectrum increases. Also the spectrum can become hardened since solid or liquid uranium objects provide considerable shielding. While it is recognized that solid or liquid uranium sources will have a hardened photon spectrum, exposure to a thin layer of uranium on a surface will result in a larger fraction of exposure to lower energy photons. (ORAUT-OTIB-0004 2005a) Considering the organ dose conversion factors and radiation effectiveness factors it would be claimant-favorable to assume that the workers were exposed to photon energies in the 30 to 250 keV range.

Thorium and its progeny create an increase in photon energies in the range of 30-250 keV; therefore it would be appropriate to assign energy that workers are exposed to in the 30-250 keV energy range.

W. R. Grace/NFS distribution of low energy photons is dependent on the amounts, separation and enrichments of uranium and the age and type of plutonium used at the site.

To ensure claimant favorability, a photon energy range of 100% 30 to 250 keV should be applied. Table 4-2 lists a more detailed description of the Nuclear Chicago or Landauer dosimetry used at W.R. Grace/NFS.

4.1.3 Non-penetrating Dose Determination and Skin Dose Calculations

Non-penetrating dose can be considered for this TBD as electrons greater than 15 keV and photons less than 30 keV if the employee worked at a plutonium facility. Actual assignment of the electron or low energy photon distribution is dependent upon the cancer location and radiation work environment of the claimant. The general approach to skin dose reconstruction is as follows: (ORAUT-OTIB-0017 2005):

- 1. Translate the reported doses into non-penetrating and penetrating doses.
- 2. Assign the penetrating dose as photons in the energy range of 30-250 keV.
- 3. Assign the non-penetrating dose as electrons > 15 keV (corrected to account for attenuation, if applicable), or photons < 30 keV if the employee worked in a plutonium facility.
- 4. Add missed electron and/or photon dose.
- 5. Include missed neutron dose, if applicable.
- 6. All dose conversion factors for the skin should be assumed to be 1.

In general, non-penetrating radiation dose should be assigned as < 30 keV photons if the employee worked with or around plutonium. Otherwise, > 15 keV electrons should be assigned. If unknown, consider the following guidance:

- 1. For a likely non-compensable case, it is acceptable to assume the non-penetrating dose is associated with < 30 keV photons, as this is claimant favorable.
- 2. For a likely compensable case, it is acceptable to assume the non-penetrating dose is associated with > 15 keV electrons.

3. If the compensability decision may hinge on this issue, and if the partitioning of the non-penetrating dose cannot be based on the available information, additional research may be required.

As noted earlier, prior to 1969, the reported skin dose represents only the shallow dose component and after 1969 the reported skin exposure includes the penetrating gamma component. For this reason, the shallow component should be calculated as the difference between the reported skin and total body dose.

For film dosimetry, there is an over-response to 16 keV x-rays and 59 keV photons by factors of 8.5 to 12 and 14 to 19 respectively. (Wilson, 1990) This would have bound response to lower energy uranium photons as well. The general approach to skin dose calculation for W.R. Grace/NFS is as follows:

Measured dose

- Subtract the reported whole body or total body (deep) dose from the skin of the whole body or skin of the total body dose starting in 1969. This will result in giving the nonpenetrating/shallow dose component. Before1969, the skin dose was assigned only as the measured beta component.
- Assign the calculated non-penetrating dose as electrons > 15 keV. A correction factor should be provided for clothing, if applicable, depending on likely clothing thickness and beta energy. Assuming two pairs of coveralls including the paper lining (0.80) and one Dacron cotton lab coat (0.91) the midpoint correction factor would be about 0.86 for uranium betas. Use Table 6-11 on page 6-23 of DOE 2000.
- 3. Assign the reported whole body or deep dose to the energies in the 30 to 250 keV range.
- 4. For Chemical/Production Operators only assign the calculated neutron dose partitioned in the energy range of 0.1 to 2 MeV and use an organ DCF of 1.

Missed Dose

- 1. For any badge cycle with a zero or minimal result in either the whole body or beta reading or both, assign a single missed dose as explained in items 6-8 below.
- 2. If only the whole body or deep dose was reported as zero or minimal (designated with a "M" (minimal) in the records for results at or below the minimal detectable level of 10 mrem), the missed dose assigned should be the appropriate MDL (gamma/x-ray) for that era (divided by 2, treated as lognormal with a GSD of 1.52) and considered to be photons in the energy range form 30 to 250 keV Use Table 4-1.
- 3. For any badge cycle with a zero result in either the skin or beta recorded dose, the missed dose can be assigned based on the appropriate MDL of the beta of that era (divided by 2, treated as lognormal) and considered to be electrons corrected by a factor 0.86 or low energy photons. Use Table 4-1.
- 4. If both the whole body or deep dose and the skin or beta readings were reported as zero, the missed dose assigned should be the appropriate MDL (gamma/x-ray) for that era (divided by 2, treated as lognormal) and considered to be 30-250 keV photons.

5. For Chemical/Production Operators only assign neutron dose according to section 4.1.4, Neutron dose.

4.1.4 <u>Neutron Dose Assessment</u>

In order to estimate neutron dose to unmonitored workers a 0.20 to 1 neutron to photon ratio based upon their photon exposure is recommended. This should be applied to "Chemical/Production Operators" for which dosimetry information is available or based upon their missed dose photon calculation. See also Table 4-16.

4.2 ENRICHED URANIUM SOURCE TERM INFORMATION

ORAUT (2004c) assessed the potential deep dose to a worker from fissile material in a 5-by-5 double-stacked birdcage array for 2,000 hr/yr used the same methods for nuclear criticality control (geometry and mass control). The calculation showed a dose result of 0.13 R/yr for a worker 1 m from the birdcage array and the 95th-percentile exposure of 0.76 R/yr for a worker 1 foot from the array. Table 4-5 lists the annual organ doses due to the potential exposure to the stacked birdcage array.

4.3 SURFACE CONTAMINATION

Facility operations created areas of surface contamination at W.R. Grace. A component of external exposure is from the settling of airborne radioactivity particles onto surfaces in the work area as described in the guidance for estimating the maximum plausible dose at atomic weapons employer

Table 4-5. Annual organ doses due to exposure to a 5-by-5 stacked array of birdcages.

	Annual	organ dose (rem)	
Organ	Photons E=30-250 keV	Photons E>250 keV	Total
Bladder	8.09E-02	5.74E-02	1.38E-01
Red bone marrow	4.07E-02	4.68E-02	8.75E-02
Bone surface	7.99E-02	4.97E-02	1.30E-01
Breast	8.23E-02	6.05E-02	1.43E-01
Colon	6.89E-02	5.49E-02	1.24E-01
Esophagus	4.47E-02	4.84E-02	9.31E-02
Eye	8.03E-02	5.72E-02	1.38E-01
Ovaries	6.21E-02	5.32E-02	1.15E-01
Testes	9.32E-02	6.12E-02	1.54E-01
Liver	6.92E-02	5.49E-02	1.24E-01
Lung	6.41E-02	5.47E-02	1.19E-01
Remainder organs	5.71E-02	5.12E-02	1.08E-01
Skin	5.80E-02	5.43E-02	1.12E-01
Stomach	8.13E-02	5.75E-02	1.39E-01
Thymus	9.15E-02	5.80E-02	1.50E-01
Thyroid	9.36E-02	6.32E-02	1.57E-01
Uterus	6.57E-02	5.11E-02	1.17E-01

Source: ORAUT (2004c).

facilities (ORAUT 2005a). The settling of radioactive particles on surfaces establishes an area-wide exposure source term and an external exposure pathway. Dose coefficients by radionuclide for assessing the external exposure to organs from surface contamination (i.e., a ground plane) are provided in Federal Guidance Report No. 13 (EPA 2002).

The available documents are not clear as to the resultant surface contamination levels in each of the buildings where radioactive material operations occurred during the time the site was an AWE facility. Therefore, to provide the component of external exposure for dose estimation, the surface contamination levels were assumed at certain values that should encompass surface contamination levels found at this facility.

Four surface contamination levels were selected for dose estimation. The lowest surface contamination level is the facility's administrative control limit of 5,000 dpm/100 cm² (2.25E+07 pCi/m²), above which surface contamination cleanup would be required (AEC 1959, 1961; Congress 1986). This level should be applied to building areas (office spaces, cafeteria, etc.) that would not have processed radioactive material but could have been contaminated by the nearby process operations. The exposure period should encompass the entire work year (i.e., 2,000 hr).

The second surface contamination level is based on the guidance for AWE facilities for the maximum air concentration from 365 d of constant disposition (ORAUT 2005a). The contamination level for this case would be approximately 8.1 E+07 pCi/m². This level is higher than the maximum surface contamination levels provided in the two AEC survey documents (AEC 1959, 1961). Because this level of contamination is only for process areas, the time of exposure is the annual work time minus 1 hr for lunch each workday.

The last two surface contamination cases consider the highest stated contamination level found in any publicly available document for this facility. Based on worker testimony given to the U.S. Congress, the highest value stated is approximately 100,000 dpm/100 cm² with an average of approximately 45,000 dpm/100 cm² from all of the values in Congress (1986). These values translate into approximately 4.50E+08 pCi/m² and 2.00E+08 pCi/m², respectively. Again, the time of exposure is only when a worker is in a process area, which is the annual work time minus 1 hr for lunch each workday.

The annual organ dose due to exposure to contaminated surfaces was estimated by multiplying the surface contamination level by the dose coefficients for contaminated ground surfaces for the applicable radionuclides. Using ground contamination dose conversion coefficients would be claimant favorable when used for surface contamination near the organ of interest. This is because the conversion factors for the ground would have an inverse square component built into them. So, for example, dose to the stomach would be different for equivalent contamination levels if on the ground versus a work surface near the abdomen. Because the facility operations varied by building, the annual organ dose estimates were determined for the combination of the radionuclides for a specific operation. Based on the process descriptions, seven radionuclide groups were assessed for external exposure, thorium, ²³³U/thorium, 93%-enriched uranium, 3.5%-enriched uranium, DU, plutonium, and MOX fuel.

For the 233 U/thorium annual organ doses, the percentage of 234 U and thorium in the surface contamination is necessary to determine the total annual dose. Documentation on the manufacture of 233 U/thorium fuel for Shippingport is not available. However, historical information implies that the fuel mixture would be 20% 233 U and 80% ThO₂ by weight (WNA 2005). This mixture was applied to the four contamination levels. The fraction of specific activity for thorium isotopes is associated with 230 Th and 232 Th. Based on a ORAUT (2005c), weight fractions of 0.116 for 230 Th and 0.884 for 232 Th are applied in this assessment.

To assess the annual external organ dose from uranium operations, the isotopic fractions for HEU, EU, and DU must be set. Based on information for Hanford (ORAUT 2004a), Table 4-6 shows the activity fractions of these forms of uranium, which were applied for the annual organ doses.

Table 4-6. Uranium activity fractions.

	Activity fractions					
Uranium form	²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U		
93% enriched ^a	0.968	0.030	0.002	0.0003		
3.5% enriched	0.818	0.034		0.147		
DU	0.155	0.011	0.0005	0.834		

Source: ORAUT (2005d).

For the production of MOX fuel for SEFOR, the mixture was 20% reactor-grade PuO_2 and 80%-depleted UO_2 (Sharma 2002). This same mixture was assumed for the W.R. Grace facility during the times of MOX fuel production. The isotopic activity fractions for DU were previously presented. For the isotopic activity fractions for plutonium radionuclides, the time since separation must be known because 241 Am will build up in the PuO_2 . It was assumed that the reactor-grade plutonium used to produce MOX fuel by the facility was not kept very long in storage before being mixed with DU for the MOX fuel. For this analysis, a storage time of 5 yr was assumed with the isotopic activity fractions from DOE Standard 1128 (DOE 2005). Therefore, the isotopic activity fractions for 5-yr-aged reactor-grade plutonium would be 0.0064 for 238 Pu, 0.0206 for 239 Pu, 0.0107 for 240 Pu, 0.9537 for 241 Pu, and 0.0086 for 241 Am.

Tables 4-7 to 4-13 show the annual organ dose estimates for the radionuclide combinations.

Table 4-7. External exposure from thorium surface contamination.

Organ	Annual dose for 2.25E+07 pCi/m ² (rem)	Annual dose for 7.5E+07 pCi/m² (rem)	Annual dose for 2.0E+08 pCi/m ² (rem)	Annual dose for 4.5E+08 pCi/m ² (rem)
R_marrow	1.37E-04	3.99E-04	1.22E-03	2.73E-03
Adrenals	9.24E-05	2.69E-04	8.21E-04	1.85E-03
B_surface	6.76E-04	1.97E-03	6.01E-03	1.35E-02
Brain	9.10E-05	2.65E-04	8.09E-04	1.82E-03
Breasts	6.86E-04	2.00E-03	6.10E-03	1.37E-02
GB_wall	8.97E-05	2.61E-04	7.97E-04	1.79E-03
Esophagus	7.45E-05	2.17E-04	6.62E-04	1.49E-03
ST_wall	1.07E-04	3.13E-04	9.54E-04	2.15E-03
SI_wall	8.78E-05	2.56E-04	7.81E-04	1.76E-03
ULI_wall	9.31E-05	2.72E-04	8.27E-04	1.86E-03
LLI_wall	9.24E-05	2.69E-04	8.21E-04	1.85E-03
HT_wall	1.02E-04	2.96E-04	9.03E-04	2.03E-03
Kidneys	1.21E-04	3.53E-04	1.08E-03	2.42E-03
Liver	1.06E-04	3.10E-04	9.45E-04	2.13E-03
Lung	1.17E-04	3.40E-04	1.04E-03	2.33E-03
Ovaries	1.09E-04	3.19E-04	9.71E-04	2.19E-03
Pancreas	8.33E-05	2.43E-04	7.41E-04	1.67E-03
Skin	4.13E-03	1.21E-02	3.67E-02	8.27E-02
Spleen	1.03E-04	3.02E-04	9.19E-04	2.07E-03
Testes	5.22E-04	1.52E-03	4.64E-03	1.04E-02
Thymus	1.20E-04	3.50E-04	1.07E-03	2.40E-03
Thyroid	1.73E-04	5.05E-04	1.54E-03	3.46E-03
UB_wall	1.08E-04	3.16E-04	9.64E-04	2.17E-03
Uterus	8.72E-05	2.54E-04	7.75E-04	1.74E-03
Muscle	4.59E-04	1.34E-03	4.08E-03	9.18E-03
H_remaind	4.26E-04	1.24E-03	3.79E-03	8.53E-03
H_E	2.85E-04	8.32E-04	2.54E-03	5.71E-03

These values also correspond to the specific activities provided in Table C.1 of ANSI 1995.

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Table 4-8. External exposure from ²³³U/thorium surface contamination.

Organ	Annual dose for 2.25E+07 pCi/m² (rem)	Annual dose for 7.5E+07 pCi/m² (rem)	Annual dose for 2.0E+08 pCi/m ² (rem)	Annual dose for 4.5E+08 pCi/m² (rem)
R marrow	1.53E-04	4.45E-04	1.36E-03	3.05E-03
Adrena	1.06E-04	3.08E-04	9.39E-04	2.11E-03
B surface	7.00E-04	2.04E-03	6.23E-03	1.40E-02
Brain	1.04E-04	3.05E-04	9.29E-04	2.09E-03
Breasts	7.02E-04	2.05E-03	6.24E-03	1.40E-02
GB_wall	1.03E-04	3.00E-04	9.13E-04	2.06E-03
Esophagus	8.67E-05	2.53E-04	7.71E-04	1.73E-03
ST wall	1.22E-04	3.55E-04	1.08E-03	2.43E-03
SI wall	1.01E-04	2.94E-04	8.96E-04	2.02E-03
ULI wall	1.07E-04	3.11E-04	9.48E-04	2.13E-03
LLI wall	1.06E-04	3.08E-04	9.40E-04	2.12E-03
HT wall	1.15E-04	3.36E-04	1.02E-03	2.31E-03
Kidneys	1.37E-04	3.99E-04	1.22E-03	2.74E-03
Liver	1.21E-04	3.52E-04	1.07E-03	2.41E-03
Lung	1.32E-04	3.84E-04	1.17E-03	2.63E-03
Ovaries	1.22E-04	3.57E-04	1.09E-03	2.45E-03
Pancreas	9.57E-05	2.79E-04	8.50E-04	1.91E-03
Skin	4.02E-03	1.17E-02	3.57E-02	8.04E-02
Spleen	1.17E-04	3.43E-04	1.04E-03	2.35E-03
Testes	5.42E-04	1.58E-03	4.82E-03	1.08E-02
Thymus	1.34E-04	3.91E-04	1.19E-03	2.68E-03
Thyroid	1.91E-04	5.58E-04	1.70E-03	3.83E-03
UB_wall	1.23E-04	3.59E-04	1.10E-03	2.46E-03
Uterus	1.00E-04	2.93E-04	8.93E-04	2.01E-03
Muscle	4.74E-04	1.38E-03	4.21E-03	9.48E-03
H_remaind	4.41E-04	1.29E-03	3.92E-03	8.83E-03
H_E	3.00E-04	8.76E-04	2.67E-03	6.00E-03

Table 4-9. External exposure from 93%-enriched uranium surface contamination.

0	Annual dose for	Annual dose for	Annual dose for	Annual dose for
Organ	2.25E+07 pCi/m² (rem)	7.5E+07 pCi/m² (rem)	2.0E+08 pCi/m² (rem)	4.5E+08 pCi/m² (rem)
R_marrow	2.57E-03	7.51E-03	2.29E-02	5.15E-02
Adrenals	2.26E-03	6.58E-03	2.00E-02	4.51E-02
B_surface	6.35E-03	1.85E-02	5.65E-02	1.27E-01
Brain	2.32E-03	6.76E-03	2.06E-02	4.64E-02
Breasts	3.67E-03	1.07E-02	3.26E-02	7.34E-02
GB_wall	2.23E-03	6.51E-03	1.98E-02	4.46E-02
Esophagus	2.06E-03	6.00E-03	1.83E-02	4.11E-02
ST_wall	2.43E-03	7.09E-03	2.16E-02	4.86E-02
SI_wall	2.26E-03	6.60E-03	2.01E-02	4.53E-02
ULI_wall	2.34E-03	6.82E-03	2.08E-02	4.68E-02
LLI_wall	2.34E-03	6.82E-03	2.08E-02	4.68E-02
Twill	2.34E-03	6.81E-03	2.08E-02	4.67E-02
Kidneys	2.44E-03	7.12E-03	2.17E-02	4.88E-02
Liver	2.43E-03	7.08E-03	2.16E-02	4.86E-02
Lung	2.56E-03	7.47E-03	2.28E-02	5.13E-02
Ovaries	2.29E-03	6.69E-03	2.04E-02	4.59E-02
Pancreas	2.19E-03	6.38E-03	1.94E-02	4.37E-02
Skin	8.77E-03	2.56E-02	7.80E-02	1.75E-01
Spleen	2.44E-03	7.11E-03	2.17E-02	4.88E-02
Testes	3.48E-03	1.01E-02	3.09E-02	6.96E-02
Thymus	2.34E-03	6.84E-03	2.08E-02	4.69E-02
Thyroid	2.69E-03	7.83E-03	2.39E-02	5.37E-02
UB_wall	2.42E-03	7.05E-03	2.15E-02	4.83E-02
Uterus	2.28E-03	6.65E-03	2.03E-02	4.56E-02
Muscle	3.30E-03	9.61E-03	2.93E-02	6.59E-02
H_remaind	3.21E-03	9.37E-03	2.85E-02	6.42E-02
H_E	2.86E-03	8.34E-03	2.54E-02	5.72E-02

	Document No. ORAUT-TKBS-0043	Revision No. 00	Effective Date: 02/14/2006	Page 38 of 47
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Table 4-10. External exposure from 3.5%-enriched uranium surface contamination.

	Annual dose for	Annual dose for	Annual dose for	Annual dose for
Organ	2.25E+07 pCi/m ² (rem)	7.5E+07 pCi/m ² (rem)	2.0E+08 pCi/m ² (rem)	4.5E+08 pCi/m ² (rem)
R_marrow	2.89E-03	8.43E-03	2.57E-02	5.78E-02
Adrenals	2.54E-03	7.41E-03	2.26E-02	5.08E-02
B_surface	7.07E-03	2.06E-02	6.28E-02	1.41E-01
Brain	2.61E-03	7.63E-03	2.32E-02	5.23E-02
Breasts	3.99E-03	1.17E-02	3.55E-02	7.99E-02
GB_wall	2.52E-03	7.34E-03	2.24E-02	5.03E-02
Esophagus	2.32E-03	6.77E-03	2.06E-02	4.64E-02
ST_wall	2.74E-03	7.99E-03	2.43E-02	5.48E-02
SI_wall	2.55E-03	7.44E-03	2.27E-02	5.10E-02
ULI_wall	2.64E-03	7.69E-03	2.34E-02	5.27E-02
LLI_wall	2.64E-03	7.69E-03	2.34E-02	5.27E-02
HT_wall	2.63E-03	7.67E-03	2.34E-02	5.26E-02
Kidneys	2.74E-03	8.01E-03	2.44E-02	5.49E-02
Liver	2.73E-03	7.98E-03	2.43E-02	5.47E-02
Lung	2.89E-03	8.42E-03	2.57E-02	5.77E-02
Ovaries	2.58E-03	7.53E-03	2.30E-02	5.17E-02
Pancreas	2.47E-03	7.19E-03	2.19E-02	4.93E-02
Skin	9.06E-03	2.64E-02	8.06E-02	1.81E-01
Spleen	2.75E-03	8.02E-03	2.44E-02	5.50E-02
Testes	3.82E-03	1.11E-02	3.39E-02	7.63E-02
Thymus	2.64E-03	7.69E-03	2.34E-02	5.27E-02
Thyroid	3.01E-03	8.77E-03	2.67E-02	6.02E-02
UB_wall	2.72E-03	7.94E-03	2.42E-02	5.44E-02
Uterus	2.57E-03	7.50E-03	2.29E-02	5.14E-02
Muscle	3.63E-03	1.06E-02	3.23E-02	7.26E-02
H_remaind	3.54E-03	1.03E-02	3.15E-02	7.09E-02
H_E	3.18E-03	9.27E-03	2.82E-02	6.35E-02

Table 4-11. External exposure from DU surface contamination.

Organ	Annual dose for 2.25E+07 pCi/m ² (rem)	Annual dose for 7.5E+07 pCi/m² (rem)	Annual dose for 2.0E+08 pCi/m ² (rem)	Annual dose for 4.5E+08 pCi/m ² (rem)
	9.81E-04	2.86E-03	8.72E-03	1.96E-02
R_marrow				
Adrena	8.34E-04	2.43E-03	7.41E-03	1.67E-02
B_surface	2.59E-03	7.55E-03	2.30E-02	5.18E-02
Brain	8.51E-04	2.48E-03	7.57E-03	1.70E-02
Breasts	1.79E-03	5.23E-03	1.59E-02	3.59E-02
GB wall	8.21E-04	2.39E-03	7.30E-03	1.64E-02
Esophagus	7.53E-04	2.20E-03	6.70E-03	1.51E-02
ST_wall	9.02E-04	2.63E-03	8.02E-03	1.80E-02
SI_wall	8.31E-04	2.42E-03	7.39E-03	1.66E-02
ULI_wall	8.60E-04	2.51E-03	7.64E-03	1.72E-02
LLI_wall	8.60E-04	2.51E-03	7.64E-03	1.72E-02
HT_wall	8.65E-04	2.52E-03	7.69E-03	1.73E-02
Kidneys	9.20E-04	2.68E-03	8.18E-03	1.84E-02
Liver	8.99E-04	2.62E-03	7.99E-03	1.80E-02
Lung	9.51E-04	2.77E-03	8.45E-03	1.90E-02
Ovaries	8.59E-04	2.50E-03	7.63E-03	1.72E-02
Pancreas	8.02E-04	2.34E-03	7.13E-03	1.60E-02
Skin	5.83E-03	1.70E-02	5.19E-02	1.17E-01
Spleen	8.99E-04	2.62E-03	7.99E-03	1.80E-02
Testes	1.61E-03	4.69E-03	1.43E-02	3.21E-02
Thymus	8.81E-04	2.57E-03	7.83E-03	1.76E-02
Thyroid	1.05E-03	3.06E-03	9.32E-03	2.10E-02
UB_wall	8.98E-04	2.62E-03	7.99E-03	1.80E-02
Uterus	8.37E-04	2.44E-03	7.44E-03	1.67E-02
Muscle	1.48E-03	4.31E-03	1.31E-02	2.95E-02
H_remaind	1.42E-03	4.15E-03	1.26E-02	2.84E-02
H_E	1.19E-03	3.47E-03	1.06E-02	2.38E-02

Document No. ORAUT-TKBS-0043	Revision No. 00	Effective Date: 02/14/2006	Page 39 of 47
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Table 4-12. External exposure from plutonium surface contamination.

Organ	Annual dose for 2.25E+07 pCi/m² (rem)	Annual dose for 7.5E+07 pCi/m² (rem)	Annual dose for 2.0E+08 pCi/m ² (rem)	Annual dose for 4.5E+08 pCi/m ² (rem)	
R_marrow	9.08E-05	2.65E-04	8.07E-04	1.82E-03	
Adrenals	7.77E-05	2.27E-04	6.90E-04	1.55E-03	
B_surface	5.02E-04	1.46E-03	4.46E-03	1.00E-02	
Brain	8.15E-05	2.38E-04	7.24E-04	1.63E-03	
Breasts	2.12E-04	6.18E-04	1.88E-03	4.23E-03	
GB_wall	7.64E-05	2.23E-04	6.79E-04	1.53E-03	
Esophagus	6.26E-05	1.83E-04	5.57E-04	1.25E-03	
ST_wall	9.52E-05	2.78E-04	8.46E-04	1.90E-03	
SI_wall	7.73E-05	2.26E-04	6.87E-04	1.55E-03	
ULL_wall	8.31E-05	2.42E-04	7.39E-04	1.66E-03	
LLI_wall	8.05E-05	2.35E-04	7.16E-04	1.61E-03	
HT_wall	8.83E-05	2.57E-04	7.85E-04	1.77E-03	
Kidneys	1.02E-04	2.98E-04	9.07E-04	2.04E-03	
Liver	9.56E-05	2.79E-04	8.50E-04	1.91E-03	
Lung	1.06E-04	3.10E-04	9.44E-04	2.12E-03	
Ovaries	7.89E-05	2.30E-04	7.01E-04	1.58E-03	
Pancreas	7.25E-05	2.12E-04	6.45E-04	1.45E-03	
Skin	5.74E-04	1.67E-03	5.10E-03	1.15E-02	
Spleen	9.53E-05	2.78E-04	8.47E-04	1.91E-03	
Testes	1.91E-04	5.58E-04	1.70E-03	3.83E-03	
Thymus	1.04E-04	3.04E-04	9.27E-04	2.09E-03	
Thyroid	1.20E-04	3.51E-04	1.07E-03	2.41E-03	
UB_wall	9.39E-05	2.74E-04	8.34E-04	1.88E-03	
Uterus	7.53E-05	2.19E-04	6.69E-04	1.51E-03	
Muscle	1.64E-04	4.78E-04	1.46E-03	3.28E-03	
H_remaind	1.57E-04	4.57E-04	1.39E-03	3.14E-03	
H_E	1.31E-04	3.82E-04	1.16E-03	2.62E-03	

Table 4-13. External exposure from MOX surface contamination.

Organ	Annual dose for 2.25E+07 pCi/m ² (rem)	Annual dose for 7.5E+07 pCi/m² (rem)	Annual dose for 2.0E+08 pCi/m² (rem)	Annual dose for 4.5E+08 pCi/m ² (rem)
Organ R marrow	8.03E-04	2.34E-03	7.14E-03	1.61E-02
Adrenals	6.82E-04	1.99E-03	6.07E-03	1.36E-02
B_surface	2.17E-03	6.33E-03	1.93E-02	4.34E-02
Brain	6.97E-04	2.03E-03	6.20E-03	1.39E-02
Breasts	1.48E-03	4.31E-03	1.31E-02	2.95E-02
GB_wall	6.72E-04	1.96E-03	5.97E-03	1.34E-02
Esophagus	6.15E-04	1.79E-03	5.47E-03	1.23E-02
ST_wall	7.41E-04	2.16E-03	6.59E-03	1.48E-02
SI_wall	6.80E-04	1.98E-03	6.05E-03	1.36E-02
ULI_wall	7.04E-04	2.05E-03	6.26E-03	1.41E-02
LLI_wall	7.04E-04	2.05E-03	6.26E-03	1.41E-02
HT_wall	7.09E-04	2.07E-03	6.31E-03	1.42E-02
Kidneys	7.56E-04	2.21E-03	6.72E-03	1.51E-02
Liver	7.38E-04	2.15E-03	6.56E-03	1.48E-02
Lung	7.82E-04	2.28E-03	6.95E-03	1.56E-02
Ovaries	7.03E-04	2.05E-03	6.25E-03	1.41E-02
Pancreas	6.56E-04	1.91E-03	5.83E-03	1.31E-02
Skin	4.78E-03	1.39E-02	4.25E-02	9.57E-02
Spleen	7.38E-04	2.15E-03	6.56E-03	1.48E-02
Testes	1.32E-03	3.86E-03	1.18E-02	2.65E-02
Thymus	7.25E-04	2.12E-03	6.45E-03	1.45E-02
Thyroid	8.63E-04	2.52E-03	7.67E-03	1.73E-02
UB_wall	7.38E-04	2.15E-03	6.56E-03	1.48E-02
Uterus	6.85E-04	2.00E-03	6.09E-03	1.37E-02
Muscle	1.21E-03	3.54E-03	1.08E-02	2.43E-02
H remainder	1.17E-03	3.41E-03	1.04E-02	2.34E-02

4.4 PLUTONIUM OPERATIONS

Due to the lack of information about plutonium operations at the W.R. Grace facility, certain assumptions were necessary for estimating the external exposure from MOX fuel production. The assumptions relate to the classification of the PuO₂, the buildup of ²⁴¹Am, and the extent of contamination outside of gloveboxes.

The classification or grade of plutonium is based on the source of the material because this sets the isotopic content of the PuO_2 . In general, there are two classifications of plutonium, namely weapons grade and reactor grade (DOE 2005). Weapons-grade plutonium has a relatively short reactor exposure time. Reactor-grade plutonium is from reprocessing DOE spent nuclear fuel or from research reactors. For radiological exposures, reactor-grade plutonium has higher radiation levels than weapons-grade plutonium due to the buildup of ²⁴¹Am from the beta decay of ²⁴¹Pu (Carbaugh 2003; DOE 2005). Therefore, it was assumed that reactor-grade plutonium was used in the production of MOX fuel at the facility.

A storage time for buildup of 241 Am was assumed based on the isotopic content in Table 8.4 of Carbaugh (2003); it is reasonable to assume that the storage time for PuO₂ at the facility would be no greater than 5 yr. Table 4-14 lists the isotopic content and specific activity of plutonium based on these assumptions.

Table 4-14. Plutonium isotopic content and specific activity.

Isotope	Decay mode ^a	Content ^a (wt%)	Specific activity ^a (Ci/g)	Content ^b (wt%)	Specific activity after 5 yr of aging ^b (Ci/g)
²³⁶ Pu	Alpha	3.0E-06	53.4	NA	NA
²³⁸ Pu	Alpha	0.58	17.1	0.10	1.64E-02
²³⁹ Pu	Alpha	72.10	6.22E-02	84.8	5.26E-02
²⁴⁰ Pu	Alpha	19.15	0.229	12.0	2.72E-02
²⁴¹ Pu	Alpha	6.29	2.52E-03	3.0	2.43E+00
	Beta		103		NA
²⁴² Pu	Alpha	1.88	3.93E-03	0.1	3.93E-06
²⁴¹ Am	Alpha	0.02	3.43	0.0	2.19E-02

a. DOE 2005.

Section 6.2.2 of DOE (2005) provided results for measured extremity photon dose rates from plutonium glovebox operations. Two glovebox cases with reactor-grade plutonium based on the isotopic concentration are shown in the third column of Table 4-14.

As discussed on pages 6-7 of DOE 2005:

Doses to the extremities are usually dominated by gamma rays in typical glovebox operations. [...] The extremity dose is more limiting than a whole body dose if the dose gradient is greater than 10:1 over a distance of 1 meter, the maximum distance from the fingers to the trunk of the body. In most cases, the source is not at arm's length and the dose gradient needs to be 10:1 or 20:1 for the extremity dose to be limiting (NUREG/CR-4297; Reece et al. 1985). But in highly shielded gloveboxes, it is possible to have a very high extremity dose from dust layer on gloves; the dose to the torso can be much lower because of shielding applied to the glovebox.

b. Carbaugh 2003.

4.5 MEDICAL X-RAYS

Site workers, whether under W.R. Grace or NFS, received an annual occupationally related diagnostic medial X-ray and a pre-employment X-ray. The exposure geometry was posterior-anterior. The air kerma at skin entrance for the diagnostic chest X-ray was estimated to be 0.2 R for pre-1970 X-rays, 0.1 R for 1970 to 1985, and 0.05 R for 1986 to the present. To date no site-specific information is available for W.R. Grace. Photofluorography (PFG) could also have been possible but unless there is evidence presented that PFG was performed in the claimant files it is reasonable to assume that PFGs were not performed. Table 4-19 shows the annual organ doses due to the assumed annual diagnostic chest X-ray (ORAUT 2005d). The values in Table 4-15 should be entered into the Interactive RadioEpidemiology Program (IREP) software as the annual dose to an acute exposure to photons with energy from 30 to 250 keV. The distribution is assumed to be normal with a standard deviation of 30%.

4-15. Annual organ doses due to the assumed annual diagnostic chest X-ray.

Organ	Annual dose (rem)	Annual dose (rem)	Annual dose (rem)
Period	1957-1969	1970-1985	1985-present
Bladder	0.025	0.0001	0.00026
Red bone marrow	0.0184 m	0.0092	0.0089
	0.0172 f		
Bone surface	0.0902	0.0451	0.0337
Breast	0.0098	0.0049	0.0058
Colon/rectum	0.025	0.0001	0.00026
Esophagus	0.0902	0.0451	0.0337
Eye	0.00640	0.0032	0.0039
Ovaries	0.025	0.0001	0.00026
Testes	0.005	0.000001	0.000005
Liver/gall bladder/spleen	0.0902	0.0451	0.0337
Lung	0.0838 m	0.0451	0.0337
	0.0902 f		
Remainder organs	0.0902	0.0451	0.0337
Skin	0.270	0.135	0.070
Stomach	0.0902	0.0451	0.0337
Thymus	0.0902	0.0451	0.0337
Thyroid	0.0348	0.0032	0.0039
Uterus	0.025	0.00013	0.00026

4.6 URANIUM-233

In the spring of 1961 NFS processed 28 kg of 233 U in used pellets. The radiation levels reached 3,000 mR/hr at 1 inch for 15 g of U_3O_8 pellets. Twenty operators were exposed to these operations with none exceeding the 10 C.F.R. pt. 20 limits; the average exposure for the operation was 377 mrem. Rubber gloves were worn during the operations in shielded gloveboxes with filtration. Birdcages were used for storage of the materials. The exposure rate for the surface of each container did not exceed 200 mR/hr, and the contamination level did not exceed 500 dpm/100 cm 2 . The reading from each birdcage did not exceed 1 mR/hr at 1 m (Householder 1963b).

4.7 OCCUPATIONAL EXTERNAL DOSE RECONSTRUCTION ASSUMPTIONS AND SUMMARY

Limited film and TLD badge results from the operational era of 1957-1970 are available to develop coworker data in order to determine doses. The minimum detectable limits MDLs of the external

Effective Date: 02/14/2006 Page 42 of 4

dosimetry may be best to use to estimate unmonitored or missed doses. (Table 4-1) The annual exposures as determined by the various source terms are estimated and include:

Revision No. 00

- Neutron exposure from the (α, n) reaction, (thermal to 2 MeV penetrating)
- Penetrating extremity exposure from plutonium glove box work, (< 30 keV photons)
- Penetrating and non-penetrating exposure from Uranium metal.
- Penetrating and non-penetrating exposure from contamination.
- Penetrating and non-penetrating Medical x-ray exposure

Table 4-16 summarizes operational annual external doses to be assigned for monitored and unmonitored workers. The annual exposures can be used to estimate doses for unmonitored workers or unmonitored periods.

Table 4-16 is to be applied to "Chemical/Production Operators" only for those claims without dosimetry information. If dosimetry information is available, the 0.20:1 neutron to photon ratio can be applied. For all other radiation workers 10 % of the highest single value in Table 4-16 can be applied.

Table 4-16 Operational external exposure summary.

Document No. ORAUT-TKBS-0043

Source	Period of exposure	Exposure category	Exposure type	Basis	Annual exposure	IREP distribution
Course	охросито	<u> </u>	red Workers		охросито	diotribution
	1	T	Γ-	T	1	Γ-
Uranium metal	1/1/1957- 12/31/1970	Photons, 30-250 keV, AP acute	Penetrating	Monte Carlo Rectangular ingot	4.16 rem	Constant
Uranium metal	1/1/1957- 12/31/1970	Photons, < 30 keV or electrons > 15 keV, AP, acute	Non- penetrating	10 × penetrating	41.6 rem	Constant
Plutonium Extremity	1/1/1966- 12/31/1973	Photons, < 30 keV, AP, acute (skin on hands and forearms)	Non- penetrating	DOE 2000	19.125 rem ring See Table 4-18	Constant
Contamination ^a	1/1/1957- 12/31/1970	Photons, 30-250 keV, AP acute	Penetrating	Table 4-12 (Tables 4-9 to15)	0.064 rem	Constant
Contamination ^a	1/1/1957- 12/31/1970	Photons, < 30 keV or electrons > 15 keV, AP, acute	Non- penetrating	10 × penetrating	0.640 rem	Constant
Neutron	1/1/1957- 12/31/1970	Neutrons, 0.1-2 MeV, AP acute	Penetrating	Neutron to photon & ratio of 0.20:1	Neutron to photon& ratio of 0.20:1 ^b	Constant
Medical x-ray	1/1/1957- 12/31/1970	PA radiographic chest exam Photons, 30-250 keV, AP acute & Photons, < 30 keV or electrons > 15 keV, AP, acute	Penetrating	Initial plus one examination per year		See ORAUT- OTIB-0006 and Table 4- 19
		Monitore	ed Workers			
Neutron	1/1/1957- 12/31/1970	Neutrons, 0.1-2 MeV, AP acute	Penetrating	Neutron to photon& ratio of 0.20:1	Neutron to photon& ratio of 0.20:1 ^b	Constant

	Period of		Exposure		Annual	IREP
Source	exposure	Exposure category	type	Basis	exposure	distribution
Medical x-ray	1/1/1957- 12/31/1970	PA radiographic chest exam Photons, 30-250 keV, AP acute & Photons, < 30 keV or electrons > 15 keV, AP, acute	Penetrating	Initial plus one examination per year		See ORAUT- OTIB-0006 and Table 4- 19
Photon Missed Dose	1/1/1957- 12/31/1970	Photons, 30-250 keV, AP acute	Penetrating	Table 4-1	0.02-0.12 rem (MDL/2) Dependent on wear period	Lognormal GSD 1.52
Photon Missed Dose	1/1/1957- 12/31/1970	Photons, < 30 keV or electrons > 15 keV, AP, acute	Non- penetrating	Table 4-1	0.04-0.24 rem (MDL/2) Dependent on wear period	Lognormal GSD 1.52

a. Uranium -233 from 1961-1969.

5.0 <u>ESTIMATION OF EXPOSURE TO RESIDUAL ACTIVITY – RESERVED</u>

6.0 ENVIRONMENTAL DOSE - RESERVED

b. Only to be applied to chemical/production operators.

REFERENCES

- AEC (U.S. Atomic Energy Commission), 1959, Davison Chemical Company, Nuclear Reactor Materials Plant, Erwin, Tennessee: Occupational Exposure to Radioactive Dust, HASL-75, Health and Safety Laboratory, Environmental Sciences Division, Field Services Branch, November. [part of PDN 240000707]
- AEC (U.S. Atomic Energy Commission), 1961, *Davison Chemical Company, Nuclear Reactor Materials Plant, Erwin, Tennessee: Occupational Exposure to Radioactive Dust*, HASL Technical Memorandum 61-9, Health and Safety Laboratory, Health Protection Engineering Division, August. [part of PDN 240000707]
- ANSI (American National Standards Institute), 1995, *Bioassay Programs for Uranium*, N13.22-1995, Health Physics Society, McClean, Virginia, October.
- Bailey, J.C.,1958, "Personnel Contamination as a Uranium Hazard," in Symposium on Occupational Health Experience and Practices in the Uranium Industry, HASL-58, Health and Safety Laboratory, U.S. Atomic Energy Commission, New York Operations Office, October 15-17.
- Carbaugh, E. H., 2003, *Methods and Models of the Hanford Internal Dosimetry Program*, PNNL-MA-860, Radiation and Health Technology, Internal Dosimetry Program, Pacific Northwest National Laboratory, Richland, Washington, January.
- Congress (U.S. Congress), 1986, Erwin Nuclear Fuel Services Hearing before the Subcommittee on Energy Conservation and Power of the Committee on Energy and Commerce House of Representatives, Ninety-ninth Congress, Second Session, Serial No. 99-178, September 18, Washington, D.C.
- DOE (U.S. Department of Energy), 2005, *Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*, DOE-STD-1128-98 Change Notice 1, Washington, D.C., February.
- DOE (U.S. Department of Energy), 2000, *Guide of Good Practices for Occupational Radiological Protection in Uranium Facilities*, DOE-STD-1136-2000 Change Notice 3, Washington, D.C., December.
- EPA (U.S. Environmental Protection Agency), 2002, "Cancer Risk Coefficients for Environmental Exposure to Radionuclides," CD Supplement in Federal Guidance Report No. 13, EPA-402-C-99-001, Rev. 1, Washington, D.C.
- Glauberman, H., 1962, "Feasibility Report for the New Highly Enriched Scrap Recovery Facilities at NRMP (Davison Chemical Company)," internal memorandum to H. Werner (Technical Services Division), June 27. [part of PDN 240000711]
- Housholder, W. R., 1963a, *Feasibility Report for the Production of Uranium Metal and Uranium Metal Alloys Enriched Up To 12%* ²³⁵*U*, W.R. Grace, Davison Chemical Division, Erwin, TN, October. [part of PDN 240000711]
- Housholder, W. R., 1963b, *Revised Feasibility Report for the Production of* ²³³UO₂/ThO₂ and ²³³UO₂/ZrO₂ Pellets, W.R. Grace, Davison Chemical Division, Erwin, TN, November. [part of PDN 240000708]

- Housholder, W. R., and Runion, H. L., 1962, Feasibility Report for New Highly Enriched Scrap Recovery Facilities at NRMP, W.R. Grace, Davison Chemical Division, Erwin, TN. [part of PDN 240000711]
- ICRP (International Commission on Radiological Protection), 1959, Report of Committee II on Permissible Dose for Internal Radiation. ICRP Publication 2, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection), 1994a, *Human Respiratory Tract Model for Radiological Protection, Publication 66*, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection), 1994b, Dose Coefficients for Intakes of Radionuclides by Workers, Publication 68, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection), 1995, Age Dependent Doses to Members of the Public from Intake of Radionuclides: Part 4 Inhalation Dose Coefficients, Publication 71, Pergamon Press, Oxford, England.
- ICRP (International Commission on Radiological Protection), 2000, *ICRP Database of Dose Coefficients: Workers and Members of the Public*, version 2.01, Pergamon Press, Oxford, England.
- Katine, E., 1960, "Davison Chemical Company Scrap Recovery Process under AEC Contract AT(30-1)2659," memorandum to H. V. Werner (Nuclear Safety Review Board), December 21, Davison Chemical Company, Erwin, TN. [part of PDN 240000703]
- Koperski, B., 2004, "Copy of the key to a 1970s era dosimetry report," facsimile, October 27, R. S. Landauer, Jr. and Company, Glenwood, IL.
- NIOSH (National Institute for Occupational Safety and Health), 2002, *External Dose Reconstruction Implementation Guideline*, OCAS-IG-001, Rev. 1, Office of Compensation Analysis and Support, Cincinnati, Ohio, August.
- NIOSH (National Institute for Occupational Safety and Health), 2004, *Estimation of Ingestion Intakes. Technical Information Bulletin*, OCAS-TIB-009, Rev. 0, Office of Compensation Analysis and Support, Cincinnati, Ohio, April.
- NIOSH (National Institute for Occupational Safety and Health), 2005, *Special External Dose Reconstruction Considerations for Glovebox Workers*, OCAS-TIB-0010, Rev. 1, Office of Compensation Analysis and Support, Cincinnati, Ohio, April.
- NFS (Nuclear Fuel Services), 2005, NFS Timeline Table, http://www.nuclearfuelservices.com/timeline-more.htm, accessed May 17, 2005.
- NRC (U.S. Nuclear Regulatory Commission), 1985, "Occupational Radiation Exposure, Fifteenth and Sixteenth Annual Reports, 1982 and 1983," NUREG-0714, Volumes 4 and 5, Washington, D.C., October.
- NRC (U.S. Nuclear Regulatory Commission), 1985–2003, "Occupational Radiation Exposure at Commercial Nuclear Power Reactors and Other Facilities," NUREG-0713, Volumes 6 through 24, Washington, D.C.

- NRC (U.S. Nuclear Regulatory Commission), 2002, Re-evaluation of the Indoor Resuspension Factor for the Screening Analysis of the Building Occupancy Scenario for NRC's License Termination Rule, NUREG-1720, Draft Report for Comment, Washington, D.C.
- ORAUT (Oak Ridge Associated Universities Team), 2004a, *Technical Basis Document for the Hanford Site Occupational Internal Dose*, ORAUT-TKBS-0006-5, Rev. 01, Oak Ridge, Tennessee, November.
- ORAUT (Oak Ridge Associated Universities Team), 2004c, *Technical Basis Document Basis for Development of an Exposure Matrix for Huntington Pilot Plant*, ORAUT-TKBS-0004, Rev. 01, Oak Ridge, Tennessee, January.
- ORAUT (Oak Ridge Associated Universities Team), 2004e, *Estimation o Neutron Dose Rates from Alpha-Neutron Reactions in Uranium and Thorium Compounds*, ORAUT-OTIB-0024, Rev. 00, Oak Ridge, Tennessee, April.
- ORAUT (Oak Ridge Associated Universities Team), 2005a, *Estimating the Maximum Plausible Dose to Workers at Atomic Weapons Employer Facilities*, ORAUT-OTIB-0004, Rev. 03, Oak Ridge, Tennessee, August.
- ORAUT (Oak Ridge Associated Universities Team), 2005b, *Internal Dose Overestimates for Facilities with Air Sampling Programs*, ORAUT-OTIB-0018, Rev. 01, Oak Ridge, Tennessee, August.
- ORAUT (Oak Ridge Associated Universities Team), 2005c, Basis for the Development of an Exposure Matrix for the Mallinckrodt Chemical Company St. Louis Downtown Site and the St Louis Airport Site, St. Louis, Missouri, Period of Operation: 1942-1958, ORAUT-TKBS-0005, Rev. 01, Oak Ridge, Tennessee, March.
- ORAUT (Oak Ridge Associated Universities Team), 2005d, *Dose Reconstruction form Occupationally Related Diagnostic X-Ray Procedures,* ORAUT-OTIB-0006, Rev. 03, Oak Ridge, Tennessee, August.
- ORAUT (Oak Ridge Associated Universities Team), 2005e, *Technical Basis Document for the Savannah River Site Occupational Internal Dose*, ORAUT-TKBS-0003, Rev. 03, Oak Ridge, Tennessee, April.
- ORNL (Oak Ridge National Laboratory), 2002, *Internal Dosimetry Program Technical Basis Document*, Oak Ridge, Tennessee, January.
- Sharma A. R. (2002), *Integral Validation and Use of Improved Nuclear Data in Fusion Blanket Studies*, Final Report, International Atomic Energy Commission Contract 302-F4-IND-11566 (B5-IND- 29641), Institute for Plasma Research, Gujrat, India, February.
- Tester, M., 2005, "OCAS conference call," documented communication with NFS, August 11, Oak Ridge Associated Universities Team, Oak Ridge, Tennessee.
- USEC (United States Enrichment Corporation), 1995, *Uranium Hexafluoride: A Manual of Good Handling Practices*, USEC-651, Rev. 7, January.

Wilson, R. H., J. J. Fix, W. V. Baumgartner, and L. L. Nichols, 1990, *Description and Evaluation of the Hanford Personnel Dosimeter Program form 1944 Through 1989*, PNL-7447, Pacific Northwest Laboratory, Richland Washington.

WNA (World Nuclear Association), 2005, *Information and Issue Briefs – Thorium*. http://www.world-nuclear.org/info/inf62.htm (accessed June 29, 2005)