

GUIDE TO RADIOLOGICAL PROTECTION IN PLUTONIUM FACILITIES - VOL 1 OF 3

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Exam Preview:

- 1. In the past, most plutonium in DOE facilities was produced for nuclear weapons and was composed of greater than _____ wt% ²³⁹Pu and about 6 to 8 wt% ²⁴⁰Pu. This material has been referred to as "weapons grade" or "low exposure" plutonium.
 - a. 78
 - b. 85
 - **c.** 90
 - d. 92
- 2. The 87.7-year half-life of ²³⁸Pu makes it an excellent heat source for space applications. Unfortunately, the same nuclear properties of plutonium that make it attractive to science also make this element hazardous to human beings.
 - a. True
 - b. False
- 3. Using Table 2.1. Isotopic Composition of Three Grades of Plutonium: Heat Source, Weapons, and Reactor, which of the following isotopes is the highest reactor grade isotope of Plutonium?
 - a. ²⁴²Pu
 - b. ²⁴¹Pu
 - c. ²⁴⁰Pu
 - d. ²³⁹Pu
- 4. Of the 15 plutonium isotopes, the two that have proven most useful are masses 239 and 241. Plutonium-239 is fissile, i.e., atoms of plutonium split upon exposure to thermal or fast neutrons.
 - a. True
 - b. False

- 5. Using Table 2.2 Uses and Availabilities of Plutonium Isotopes, which of the following Plutonium isotopes matches the description: Popular environmental and biological chemical tracers?
 - a. ²³⁶Pu
 - b. ²³⁹Pu
 - c. ²⁴⁴Pu
 - d. ²³¹Pu
- 6. The primary control for contamination in a plutonium plant is the facility design. Contamination is confined primarily by enclosing the process areas and using controlled ventilation systems.
 - a. True
 - b. False
- 7. According to the reference material, at least one professional staff member at the plutonium facility should have a minimum of _____ years of health physics experience in the operation of plutonium facilities.
 - a. 10
 - b. 8
 - c. 5
 - d. 3
- 8. The decontamination factor is the ratio of the initial contamination level to the contamination level after decontamination methods are applied, as determined by survey instrument readings. Nonabrasive methods should be repeated until the decontamination factor between washes drops below 5 or 6 with significant contamination still remaining.
 - a. True
 - b. False
- 9. Using Table 2.5. Allotropic Forms of Plutonium Metal, what is the stability range for the β phase of plutonium metal?
 - a. $\sim \! 115$ to 200 °C
 - b. ~ 200 to $310 \ ^{\circ}C$
 - c. 310 to 452 °C
 - d. Stable below 115 °C
- 10. According to the reference material, Radioactive material can enter the body by four different pathways: by inhalation, through a wound (including an accidental injection), by ingestion, or by absorption through intact skin. Which of the following pathway matches the description: is probably the most prevalent mode for occupational intake of plutonium. It also provides a generally conservative assumption of intake for designing bioassay programs?
 - a. By ingestion
 - b. Through a wound
 - c. Inhalation
 - d. By absorption through intact skin

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Foreword

This Technical Standard does not contain any new requirements. Its purpose is to provide information on good practices, update existing reference material, and discuss practical lessons learned relevant to the safe handling of plutonium. U.S. Department of Energy (DOE) health physicists may adapt the recommendations in this Technical Standard to similar situations throughout the DOE complex. The Standard provides information to assist plutonium facilities in complying with Title 10 of the Code of Federal Regulations (CFR), Part 835, <u>Occupational Radiation Protection</u>. The Standard also supplements the DOE 10 CFR 835 Implementation Guide, DOE Orders, and DOE standard, DOE-STD-1098-2008, <u>Radiological Control</u>, (RCS) and has as its sole purpose the protection of workers and the public from the radiological hazards that are inherent in plutonium storage and handling.

This Standard uses the word "shall" to identify a required practice or the minimum acceptable level of performance. The word "should" is used to identify good practices (preferred practices) recommended by this Standard. The word "may" is used to identify permitted practice (neither a requirement nor a recommendation).

This Standard includes provisions in the 2007 amendment to 10 CFR 835. This amendment updated the dosimetric terms and models for assessing radiation doses, both internal and external. Of particular interest for this Standard, the biological transportability of material is now classified in terms of absorption types; F (fast), M (medium) and S (slow). Previously this was classified in terms of material class; D (days), W (weeks) and Y (years). Throughout this Standard, discussions of previous studies describing the biological transportation of material in the body will continue to use D, W and Y, as appropriate. Discussions of other requirements which have not amended their dosimetric terms and models continue to use the older terminology.

This Standard does not include every requirement applicable to every plutonium facility. Individuals responsible for implementing Radiation Protection Programs at plutonium facilities need to be knowledgeable of which requirements (contractual or regulatory) are applicable to their facility.

Copies of electronic files of this Technical Standard may be obtained from either the DOE Radiation Safety Home Page Internet site

(http://www.hss.energy.gov/HealthSafety/WSHP/radiation/ts.html) or the DOE Technical Standards Program Internet site (http://www.hss.doe.gov/nuclearsafety/techstds/standard.html).

1.0 INTRODUCTION

1.1 PURPOSE AND APPLICABILITY

This Technical Standard (TS) does not contain any new requirements. Its purpose is to provide information on good practices, update existing reference material, and discuss practical lessons learned relevant to the safe handling of plutonium. U.S. Department of Energy (DOE) health physicists may adapt the recommendations from this TS to similar situations throughout the DOE complex. Generally, DOE contractor health physicists will be responsible to implement radiation protection activities at DOE facilities and DOE health physicists will be responsible for oversight of those activities. This guidance is meant to be useful for both efforts. The TS provides information to assist plutonium facilities in complying with Title 10 of the Code of Federal Regulations (CFR), Part 835, Occupational Radiation Protection (DOE, 2011); hereinafter referred to as 10 CFR 835. The TS also supplements the DOE 10 CFR 835 Implementation Guide, G 441.1-1C, Ch 1 (DOE, 2011a), DOE Orders, and DOE's standard Radiological Control, (DOE, 2017) and has as its sole purpose the protection of workers and the public from the radiological hazards that are inherent in plutonium storage and handling. This Standard does not include every requirement applicable to every plutonium facility. Individuals responsible for implementing Radiation Protection Programs at plutonium facilities need to be knowledgeable of which requirements (contractual or regulatory) are applicable to their facility.

This TS was originally based upon the data in PNL-6534, <u>Health Physics Manual of</u> <u>Good Practices for Plutonium Facilities</u> (PNL, 1988), which provided information of situations that were typical of DOE's plutonium operations; safe storage, handling and inspection, decontamination, and decommissioning (environmental restoration); and weapons disassembly. This 2013 revision updates the 2008 revision and discusses requirements for DOE accident investigations of particular applicability for plutonium facilities and updates information on chelation therapy.

The technical information presented here represents the best technical information available from within the DOE complex. Except to the extent that the guidance presented here duplicates mandatory regulations or contract requirements, it is not binding or mandatory. Any DOE Orders, manuals or guides, referred to in this TS are not binding unless they have been incorporated into the applicable contract to assist in identifying applicable requirements, "shall" statements are followed by a reference. Should and may statements are provided for consideration. However, judicious use of this TS, along with the regulatory documents discussed above, will help assure a comprehensive and technically defensible radiological protection program.

References are current as of December 2011.

1.2 DEFINITIONS

A glossary is provided (see Appendix) to ensure uniform understanding of words in this document. In all cases, the definitions given here are consistent with those used in the Implementation Guide (DOE, 2011a).

1.3 ACRONYMS

AC	Alternating Current
AMAD	Activity Median Aerodynamic Diameter
ANSI	American National Standards Institute
ALI	Annual Limit on Intake
ALARA	As Low As Reasonably Achievable
BNL	Brookhaven National Laboratory
CDC	Centers for Disease Control and Prevention
CAS	Chemical Abstracts Service
CAM	Continuous Air Monitor
CFR	Code of Federal Regulations
CED	Committed Effective Dose
D	Day
D&D	Decontamination and Decommissioning
DTPA	Diethylenetriamine Pentaacetic Acid
DAC	Derived Air Concentration
DPM	Disintegrations Per Minute
DOE	U.S. Department of Energy
DCF	Dose Conversion Factor
EA	Environmental Assessment
EIS	Environmental Impact Statement
EMG	Emergency Management Guide
EMS	Emergency Management System
EOC	Emergency Operations Center
EPA	U S Environmental Protection Agency
FPHA	Emergency Planning Hazards Assessment
FRO	Emergency Response Organization
F	Fast
FDA	U.S. Food and Drug Administration
GI	Gastrointestinal
GM	Geiger-Mueller
HPS	Health Physics Society
ΗΕΡΔ	High Efficiency Particulate Air
HLW	High-Level Waste
HO	Headquarters
IMBA	Integrated Modules for Bioassay Analysis
IG	Implementation Guide
IRF	Intake Retention Function
ISMS	Integrated Safety Management System
ICRP	International Commission on Radiological Protection
IEC	International Electrotechnical Commission
ISO	International Organization for Standardization
I A NI	Los Alamos National Laboratory
LANL	Los Alamos National Laboratory
LLI	Low-Level Waste
M	Medium
MDA	Minimum Detectable Amount/Activity
MDD	Minimum Detectable Dose
MARSAME	Multi-Agency Radiation Survey and Assassment of
	Materials and Equipment
	·

MARSSIM	Multi-Agency Radiation Survey and Site Investigation
	Manual
MW	Mixed Waste
NAD	Nuclear Accident Dosimeters
NCRP	National Council on Radiation Protection and
	Measurements
NEPA	National Environmental Policy Act
NIOSH	National Institute for Occupational Safety and Health
NNSA	National Nuclear Security Administration
NIST	National Institute of Standards and Technology
NRC	U. S. Nuclear Regulatory Commission
NVLAP	National Voluntary Laboratory Accreditation Program
ORNL	Oak Ridge National Laboratory
PAGs	Protective Action Guides
PNAD	Personnel Nuclear Accident Dosimeter
PSO	Program Secretarial Office
QC	Quality Control
RCRA	Resource Conservation and Recovery Act
RCT	Radiological Control Technician
RWP	Radiological Work Permit
RCS	DOE Radiological Control Standard
RESL	Radiological and Environmental Sciences Laboratory
S	Slow
SNAP	Space Nuclear Auxiliary Power
TEPC	Tissue Equivalent Proportional Counter
TS	Technical Standard
TLD	Thermoluminescent Dosimeter
TED	Total Effective Dose
TRU	Transuranic
TSCA	Toxic Substances Control Act
USC	United States Code
WIPP	Waste Isolation Pilot Plant
W	Week
Y	Year

1.4 DISCUSSION

Chapters 2 through 10 provide technical information to assist in safely managing plutonium operations. The topics covered are those considered by representatives of many of DOE's plutonium facilities to be most beneficial: Manufacture, Properties and Hazards, Radiation Protection, Contamination Control, Internal Dosimetry, External Dose Control, Nuclear Criticality Safety, Waste Management, Emergency Management, and Decontamination and Decommissioning.

2.0 MANUFACTURE, PROPERTIES, AND HAZARDS

This chapter briefly describes the manufacture of plutonium and presents the nuclear, physical, chemical, and radiobiological properties of plutonium (and/or sources for these data) that form the basis for radiological and toxic control limits. The data and discussion are intended to provide a basis for understanding the changes in hazards as a function of such parameters as isotopic composition, age since chemical processing, physical form, and chemical form. Data are presented to facilitate the calculation of radiation effects, which occur from a variety of plutonium sources.

Plutonium is the first man-made element produced on an industrial scale. The special nuclear properties of ²³⁹Pu and ²³⁸Pu have led scientists to focus their efforts on these two isotopes. The fission cross-section of ²³⁹Pu makes it a useful energy source for atomic weapons and nuclear power reactors. The 87.7-year half-life of ²³⁸Pu makes it an excellent heat source for space applications. Unfortunately, the same nuclear properties of plutonium that make it attractive to science also make this element hazardous to human beings. All 15 plutonium isotopes are radioactive, with half-lives ranging from 26 minutes for ²³⁵Pu to 7.6 x 10⁷ years for ²⁴⁴Pu.

2.1 MANUFACTURE OF PLUTONIUM

Because of its high specific alpha activity and high decay heat, ²³⁸Pu has been used as an isotopic heat source for devices that generate thermoelectric power, such as the Space Nuclear Auxiliary Power (SNAP) systems used in lunar and deep space missions. Small amounts of ²³⁸Pu with low ²³⁶Pu content were used as a power source for medical prosthetic devices such as cardiac pacemakers and a prototype artificial heart, but lithium batteries have replaced these plutonium power sources. ²³⁸Pu containing a few parts per million of ²³⁶Pu is produced by irradiating ²³⁷Np with slow neutrons. It can also be produced by irradiating ²⁴¹Am to form ²⁴²Cm, which quickly decays to ²³⁸Pu.

In the past, most plutonium in DOE facilities was produced for nuclear weapons and was composed of greater than 90 wt% ²³⁹Pu and about 6 to 8 wt% ²⁴⁰Pu. This material has been referred to as "weapons grade" or "low exposure" plutonium. It is produced on a large scale by irradiating ²³⁸U in moderated production reactors (see Figure 2.1). Plutonium has also been produced as a byproduct in the operation of research reactors, and commercial nuclear power plants. It is recovered and purified by solvent extraction and ion exchange processes. The resulting highly concentrated Pu(NO₃) ₄ product solution is converted to a nonhygroscopic PuF₄ intermediate by one of the several processes before being reduced to metal with calcium. Plutonium is also produced from the waste streams of the conversion processes and scrap recovery operations, which include material from research and development efforts. Other processes for reduction to metal include direct reduction of the oxide and electrolytic reduction. Typical isotopic compositions of three common grades of plutonium are given in Table 2.1.

$${}^{238}_{92}\text{U} + {}^{1}_{0}\text{n} \longrightarrow {}^{239}_{92}\text{U} \xrightarrow{\beta^{-}}_{23.6 \text{ min}} {}^{239}_{62}\text{Np} \xrightarrow{\beta^{-}}_{2.3566 \text{ d}} {}^{239}_{94}\text{Pu}$$

Figure 2.1. Principal Mode of Plutonium Production by Neutron Irradiation of Uranium

 Table 2.1. Isotopic Composition of Three Grades of Plutonium: Heat Source, Weapons, and Reactor (PNL, 1988)

Isotope Heat Source		Weapons Grade	Reactor Grade	
²³⁸ Pu	90.0	<0.05	1.5	
²³⁹ Pu	9.1	93.6	58.1	
²⁴⁰ Pu	0.6	6.0	24.1	
²⁴¹ Pu	0.03	0.4	11.4	
²⁴² Pu	< 0.01	<0.05	4.9	

Overviews of plutonium process chemistry at DOE's Hanford, Los Alamos, Rocky Flats, and Savannah River sites are given by Christensen et al. (1983), Baldwin and Navratil (1983), Coops et al. (1983), and Christensen and Mullins (1983). In each case, solutions for recovery, purification, and waste treatment operations are emphasized.

2.1.1 Future Sources of Plutonium

High-exposure plutonium, i.e., plutonium containing significant fractions of ²⁴⁰Pu, ²⁴¹Pu, and ²⁴²Pu and generating external dose rate fields greater than the other isotopes of plutonium, is produced in power reactor fuels. Currently, this form of plutonium is in the irradiated fuel in spent-fuel storage basins and other sources resulting from development work performed to demonstrate plutonium fuel cycles. Because recycling of commercial reactor fuel is not anticipated, future supplies of plutonium will be primarily from DOE production facilities and from reprocessing of current material. In the more distant future, Space Nuclear Auxiliary Power may be a potential source of plutonium.

Special isotopes of reasonably high purity are also available, which can be useful to health physicists for calibration purposes. These isotopes and their sources are listed in Table 2.2.

New sources of plutonium include the return of atomic weapon components and plutonium recovered from decontamination and decommissioning (D&D) operations. Foreign plutonium from states of the former Soviet Union may become an additional

source. Their weapons-grade plutonium is believed to contain 5% ²⁴⁰Pu. Americium is not periodically removed from their stockpile material.

Isotopes	Uses	Availability
²³⁶ Pu, ²³⁷ Pu	Popular environmental and biological chemical tracers.	Both available in microcurie quantities. ^(a)
²³⁸ Pu	Small thermal and electric-power generators.	Available in various isotopic enrichments, ranging from 78% to 99+%. ^(a)
²³⁹ Pu	Nuclear weapons and as a fast reactor fuel. Also, frequently used in chemical research where production-grade material of mixed isotopic content is suitable.	Available enrichments range from 97% to 99.99+%. ^(a)
²⁴⁰ Pu	Principally in flux monitors for fast reactors.	Available enrichments range from 93% to 99+%. ^(a)
²⁴¹ Pu	The parent from which high-assay ²⁴¹ Am can be isolated for industrial purposes.	Samples available in enrichments of 93% ^(a)
²⁴² Pu	For study of the physical properties of plutonium; also as a mass spectroscopy tracer and standard.	Samples available in enrichments ranging from 95% to 99.9+%; enrichments of production-grade material range from 85% to 95%. ^(a)
²⁴⁴ Pu	Available as a National Institute of Standards and Technology (NIST) Standard Reference Material (SRM)	DOE's New Brunswick Laboratory.

Table 2.2 Uses and Availabilities of Plutonium Isotopes

(a) Available in small quantities from the Oak Ridge National Laboratory (ORNL): ORNL Isotopes Sales Office, Oak Ridge National Laboratory, P.O. Box X, Oak Ridge, Tennessee 37830.

2.2 NUCLEAR PROPERTIES

Of the 15 plutonium isotopes, the two that have proven most useful are masses 239 and 238. Plutonium-239 is fissile, i.e., atoms of plutonium split upon exposure to thermal or fast neutrons. Chemical reactions can release a few electron volts of energy per atom; however, when a plutonium nucleus splits, it releases about 200 MeV of energy and two or three neutrons. This release of energy makes ²³⁹Pu useful for nuclear weapons and reactor fuel. In fact, in light water reactors much of the power originates from the fission of ²³⁹Pu, which is produced by neutron capture in ²³⁸U. Because of its higher specific activity, ²³⁸Pu is used as long-lived heat sources for powering planetary space missions where adequate solar energy is not available.

As mentioned before, all plutonium isotopes are radioactive. Isotopes with even mass numbers (except mass number 246) are primarily alpha emitters. Isotopes of mass numbers 232, 233, 234, 235, and 237 also decay by electron capture; isotopes of

mass numbers 241, 243, 245, and 246 decay by beta emission. Many of the alphaemitting isotopes, such as ²³⁸Pu and ²⁴⁰Pu, also fission spontaneously and emit neutrons. All of the particle emissions are accompanied by X-ray and gamma-ray emissions over a wide range of energies.

A review of the nuclear properties of plutonium (e.g., cross-sections, nuclear levels, half-lives, and fission yields) can be found in Volume 1 of the <u>Plutonium Handbook:</u> <u>A Guide to the Technology</u> (Wick, 1967) and in American National Standards Institute (ANSI) Standard N317, <u>Performance Criteria for Instrumentation Used for</u> <u>In-Plant Plutonium Monitoring</u> (ANSI, 1980a). Plutonium decay schemes, neutron yields, and neutron energy spectra are described in the following sections.

2.2.1 Decay Schemes

The decay modes of some important plutonium and other isotopes and decay products are shown in Table 2.3. For brevity, only the most abundant radiations have been included in the table; more detailed information can be found in papers by Gunnink and Morrow (1967) and Clein (1971), in International Commission on Radiological Protection (ICRP) Publication 107 (ICRP, 2007), and from the National Nuclear Data Center. Most of the isotopes are strong alpha-emitters, making alpha heating a problem for the storage and handling of large amounts of plutonium. The specific activities and decay heats for selected isotopes and decay products are given in Table 2.4. Kilogram quantities of ²³⁹Pu or gram quantities of ²³⁸Pu can generate enough heat to melt plastic bags. Sources of ²³⁸Pu shall be handled with insulated gloves, and special precautions shall be taken to ensure a good thermal heat sink during shipping and storage. (See also Section 2.5.1, "Self-Heating.")

The plutonium isotopes emit relatively few high-energy gamma rays, so even kilogram quantities can be processed without serious gamma-exposure problems. Because of the high density of plutonium, many gamma rays are self-absorbed. In some instances, the decay products may become significant in radiation protection and metallurgy. For instance, the isotope ²³⁶Pu often constitutes less than 1% of plutonium and is often ignored in dose calculations. However, if the plutonium is shielded by greater than 1 cm of lead or steel, the decay products of ²³⁶Pu may be the largest contributors to exposure. The decay product ²⁰⁸Tl emits a highly penetrating gamma ray with an energy of 2.615 MeV. Although ²⁴¹Pu is a beta emitter and not as great an inhalation hazard as other isotopes of plutonium, in plutonium that contains a few weight percent ²⁴¹Pu the ²⁴¹Am decay product is important because it emits a large number of 60-keV photons, which can be a significant source of exposure to the hands and forearms when handling plutonium in gloveboxes (See Section 6.3.3 for more information). Also, ²⁴¹Am can contribute to neutron dose. Americium-241 contributes to increased alpha emission which affects the neutron dose as well as radiolysis and helium retention and release. Because of its importance to radiation exposure, the fractional amount of ²⁴¹Am produced by beta decay from ²⁴¹Pu is given as a function of time since chemical separation (see Figure 2.2).

Isotope	Half-Life	Mode of			X-ray ^(b)		Gamma R	ay
1		Decay	Energy	Yield	Energy	Yield	Energy	Yield
		Particle	Mev	%	MeV	%	MeV	%
236 P 11	2 851 v	α	5 77	693	L's 0 011-0 021	13(c)	0.0476	6.6x10 ⁻²
1 u	2.001 y	a	5 72	30.6	1 5 0.011 0.02	1.15	0 109	1.2×10^{-2}
238 D.	977	u a	5.50	71.0	$I_{2} = 0.011 + 0.021$	1 5 (c)	0.0425	2.05×10^{-2}
²³⁰ Fu	o/./ y	u a	5.50	/1.0 20.0	L \$ 0.011-0.02	1 13(*)	0.0000	7.25×10^{-3}
220	2 41 104	ά	5.40	28.8	1, 0,0116	5 O(c)	0.0999	7.55×10^{-3}
²³⁹ Pu	2.41 x 10 ⁺ y	α	5.157	15.0	L's 0.0116-	5.0(0)	0.077	1.22x10
		ά	5.144	13.0	0.0215		0.129	0.41×10^{-3}
		α	5.106	11.8			0.375	1.55x10
							0.414	1.46x10 ⁻³
²⁴⁰ Pu	6564 y	α	5.168	72.8	L's 0.0115-	10.8(c)	0.0452	4.50x10 ⁻²
		α	5.124	27.1	0.0215		0.104	7.08x10 ⁻³
²⁴¹ Pu	14.35 y	β	0.0052 ^(d)	100			0.077	2.20x10-5
		α	4.896	2.04	x 10 - 3		0.1037	1.01x10-4
							0.114	6.0x10-6
							0.149	1.9x10-4
	E						0.160	6.71x10-6
²⁴² Pu	3.73 x 10 ⁵ y	α	4.901	77.5	L's 0.0116-	9.1 ^(c)	0.0449	3.6x10 ⁻²
		α	4.857	22.4	0.0215		0.104	7.8x10 ⁻³
²⁴¹ Am	432.2 y	α	5.486	85.2	L's 0.0119-	42 ^(c)	0.0263	2.4
		α	5.443	12.8	0.0222		0.0332	1.2×10^{-1}
		α	5.388	1.4			0.0595	35.7
²³⁷ U	6.75 d	β	0.039 ^(e)	0.8	L's 0.0119-	70(c)	0.0263	2.43
		β	0.050 ^(e)	3.4	0.0206		0.0595	34.5
		β	$0.065^{(e)}$	51			0.0648	1.28
		β	0.069 ^(e)	42	K's 0.097-0.11	4 53	0.165	1.85
							0.208	21.1
							0.268	7.1x10 ⁻¹
							0.332	1.2
							0.335	9.5×10^{-2}
							0.369	4.0×10^{-2}

Table 2.3.	Radioactive Decay Properties of Selected Isotopes and Decay
Products, Exe	cluding Spontaneous Fission ^(a)

Data from Dunford and Burrows (1993). (a)

L's = L X-rays; K's = K X-rays. (b)

Total for all X-rays. The value represents an average obtained from data at Pacific Northwest Laboratory, (c) Lawrence Berkeley Laboratory, and Lawrence Livermore Laboratory. Average beta energy given. The maximum beta average for ²⁴¹Pu is 0.0208 MeV. Average beta energy. The maximum beta energy for ²³⁷U is 0.248 MeV.

(d)

(e)

Isotope	Half-Life Y	Spe	cific Activity, Ci/G	Averag per Di	ge Particle Energy sintegration, MeV ^(b)	Decay Heat W/g ^(b)
²³⁶ Pu	2.851	α	53.4	α	5.75	18.2
238Pu	87.7	α	17.1	α	5.49	0.567
²³⁹ Pu	2.407 x 10	$^{4}\alpha$	6.22 x 10 ⁻²	α	5.14	1.93 x 10 ⁻³
²⁴⁰ Pu	6564	α	0.229	α	5.16	7.13 x 10 ⁻³
²⁴¹ Pu	14.35	α	2.52 x 10 ⁻³	α+β	5.27 x 10 ⁻³	3.29 x 10 ⁻³
		β	103			
²⁴² Pu	3.733 x 10	5α	3.93 x 10 ⁻³	α	4.90	1.16 x 10-4
232U	72.0	α	21.5	α	5.31	0.690
233 U J	1.59 x 10 ⁵	α	9.75 x 10 ⁻³	α	4.72	2.84 x 10 ⁻⁴
234 []	2.45 x 10 ⁵	α	6.29 x 10 ⁻³	α	4.76	1.81 x 10 ⁻⁴
235 []	7.04 x 10 ⁸	α	2.17x10 ⁻⁶	α	4.24	6.02 x 10 ⁻⁸
236 []	2.34 x 10 ⁷	α	6.5 x 10 ⁻⁵	α	4.48	1.77 x 10 ⁻⁶
238 []	4.47 x 10 ⁹	α	3.38 x 10 ⁻⁷	α	4.18	8.58 x 10 ⁻⁹
²³⁷ Nn	2.14 x 10 ⁶	α	7.08 x 10 ⁻⁴	α	4.76	2.08 x 10 ⁻⁵
²⁴¹ Am	432.2	α	3.43	α	5.37	0.115

Table 2.4. Specific Activity Decay Heats of Selected Isotopes^(a)

(a) Data from ICRP 38 (1983).

(b) Includes atomic recoil and low-energy X-ray production.

2.2.2 Neutron Yields and Spectra

Plutonium and plutonium compounds also emit neutrons from spontaneous fission and from alpha-neutron reactions with light elements. The spontaneous fission half-life and the neutron yields from spontaneous fission and alphaneutron reactions for plutonium metal and plutonium compounds are provided in Section 6.0 of this TS. The approximate neutron yield from a substance with a known isotopic composition can be determined by adding the contributions from each component. This procedure and its limitations are described in detail in Section 6.0, which also discusses neutron equivalent dose rates.

Energy spectra from Pu-Be and Pu-B neutron sources are shown in Figure 2.3 Because of licensing restrictions on plutonium, these sources have been replaced with source fabricated from americium. Metallic plutonium emits neutrons having a Maxwellian energy distribution, with an average energy of about 1.9 MeV. Plutonium compounds and alloys also emit neutrons from alpha-neutron reactions, and these neutrons have significantly different energies:

-- PuF4, about 1.3 MeV

-- 10% plutonium-aluminum alloys, 1.6 MeV

-- PuO₂, slightly more than 2 MeV

-- PuBe, 4.3 MeV.

Plutonium compounds or alloys containing sodium, magnesium, silicon, chlorine, carbon, or oxygen have significant alpha-neutron yields, but little information is available about their neutron energy spectra.



Figure 2.2. Atom Ratio of ²⁴¹Am to ²⁴¹Pu (t=0) Produced by the Beta Decay of ²⁴¹Pu as a Function of Time Since Chemical Separation

2.3 PHYSICAL AND CHEMICAL PROPERTIES

This discussion of plutonium's physical and chemical properties begins with plutonium metal, followed by its alloys and compounds. Knowledge of the physical properties of these classes of materials and how the plutonium was produced is the key to understanding and predicting the hazards of working with this challenging element. According to Healy (1993), "Nature does not decide what happens to any material based on its radioactivity but rather on its form and mass." Form and mass are determined by the engineering application and the kinds of processes needed to achieve both intermediate and final products. Thus, to prevent nature from taking its course, there can be no shortcuts in good practices for plutonium facilities.

2.3.1 Plutonium Metal

The metallic state of plutonium is undoubtedly the most complicated of all the elements. Plutonium is a silvery-white metal, much like nickel in appearance. It has a low melting point (640°C) and an unusually high boiling point (3327°C). The metal exists in six allotropic forms, as indicated in Table 2.5. Two of the allotropic forms, σ and σ ', contract upon heating; the other forms expand upon heating.



Figure 2.3 Neutron Energy Spectra of Plutonium-Beryllium and Plutonium-Boron Neutron Sources Compared with a Fission Source

At room temperature, pure plutonium exists in the α phase, which has a triclinic structure with a theoretical density of about 19.86 g/cm³. The dimensional stability of this phase is aggravated by its high linear thermal expansion coefficient and its low $\alpha \rightarrow \beta$ transition temperature. This transformation takes place at approximately 115°C, resulting in a 10% volume change. The combination of a high specific activity and low thermal conductivity can result in significant dimensional distortion during metal-forming operations. For this reason, a σ -stabilized dilute gallium alloy, which has a density of about 15.75 g/cm³, is used when a more dimensionally stable plutonium is desired (Merz, 1971).

Phase	Stability Range °C	Density g/cm ^{3(b)}	
α	Stable below 115	19.86	
β	~115 to 200	17.70	
λ	~200 to 310	17.14	
σ	310 to 452	15.92	
ο'	452 to 480	16	
€	480 to 640	16.51	

Table 2.5. Allotropic Forms of Plutonium Metal^(a)

(a) Wick, 1967, p. 34.

(b) Theoretical X-ray density. The actual density is slightly lower due to crystal lattice imperfection.

Plutonium is an active metal. In moist air or moist argon, the metal oxidizes rapidly, producing a mixture of oxides and hydrides (Haschke, 1992). If the metal is exposed long enough, an olive-green powdery surface coating of PuO₂ is formed. With this coating, the metal is pyrophoric, so plutonium metal is usually handled in an inert, dry atmosphere of nitrogen or argon. Oxygen retards the effects of moisture and acts as a passivating agent (Raynor and Sackman, 1963). For a description of the storage hazards that the oxidation of plutonium metal creates, see Section 2.6.3.1, "Oxidation of Plutonium." A comprehensive treatment of the oxidation of plutonium, the properties of its oxides, oxide growth, and oxidation kinetics was reviewed by Colmenares (1975).

Plutonium metal also reacts with most common gases at elevated temperatures. Plutonium metal is rapidly dissolved by HCl, HBr, 72% HCl04, 85% H₃PO4, concentrated CCl₃COOH (trichloroacetic acid), sulfamic acid, and boiling concentrated HNO₃ in the presence of 0.005M HF. The metal reacts slowly with water, dilute sulfuric acid, and dilute acetic acid. There is no reaction with the metal in pure HNO₃ at any concentration, with concentrated acetic acid, nor with dilute sodium hydroxide.

2.3.2 Plutonium Alloys

Alloying plutonium gives rise to a host of materials with a wide range of physical, chemical, and nuclear properties.¹ The search for and development of new alloys has been focused mainly on the manufacture of atomic weapons, reactor fuels, heat sources, and neutron sources. The challenge of alloy development is how to maximize the desired properties without adding undesired ones. Unfortunately, some properties mutually exclude others (e.g., a gain in hardness usually results in a loss of ductility), so users may be forced to rethink their needs.

The radiological hazards of a plutonium alloy taken through its product life cycle differ from those of the pure metal isotope by virtue of the alloy's properties, which affect its form (i.e., its chemical composition, density, and geometric shape). Because form can be radically changed by external conditions (e.g., heat, pressure, and chemical atmosphere), a knowledge of the following properties will aid in evaluating the radioactive hazard:

melting point	diffusivity
viscosity	strength
vapor pressure	ductility
corrosion resistance	 pyrophoricity.

In nuclear fuel applications, the neutron absorption cross-section of the alloying elements and impurities shall also be considered for its effect on radiation exposure.

2.3.3 Plutonium Compounds

Much of what was said in Section 2.3.2 about the properties of plutonium alloys also applies to plutonium compounds because both are mixtures of plutonium and other elements.

Plutonium is the fifth element in the actinide series, which consists of elements with properties that stem from partial vacancies in the 5th electron shell. These elements form the seventh row in the periodic table. In general, there are four oxidation states: III, IV, V, and VI. In aqueous solutions, plutonium (III) is oxidized into plutonium (IV), which is the most stable state. The compounds PuF_4 , $Pu(I0_3)_4$, $Pu(OH)_4$, and $Pu(C_2O_4)_2 6H_2O$

¹ See Volume 1 (Section 2) and Volume 2 (Section 5) of the <u>Plutonium Handbook: A Guide to the Technology</u> (Wick, 1967); Plutonium (Taube, 1964); and Chapter 11 of the "Reactor Handbook" in <u>Materials</u>, vol. 1 (Tipton, 1960). Beginning in 1957, a series of international conferences were held whose proceedings contain a wealth of information on plutonium alloys. From 1960 through 1975, the conferences were held every five years and produced a proceedings for each conference: Refer to <u>The Metal Plutonium</u> (Coffinberry and Miner, 1961); <u>Plutonium 1960</u> (Grison et al., 1961); "Plutonium 1965" (Kay and Waldron, 1966); "Plutonium 1970 and Other Actinides," Parts I and II (Miner, 1971); and "Plutonium 1975 and Other Actinides" (Blank and Lindner, 1976).

(plutonium oxalate) are insoluble in water. The chlorides, nitrates, perchlorates, and sulfates are soluble in water. Plutonium (IV) ions complex readily with organic and inorganic compounds. Of particular importance for radiological safety considerations are the solubility, particle size, and surface area of plutonium compounds. These properties play an important part in the transportability of plutonium in the environment and in the body. All plutonium compounds, except the oxides, were assumed in ICRP 30, Part 1 (ICRP, 1979) to behave as class W compounds in the ICRP lung model. Plutonium oxides were assumed to be class Y. The 2007 amendment to 10 CFR 835 adopted models from ICRP 60, 61, 66 and 68 (ICRP, 1991a, 1191b, 1994a, and 1994b). These models assumed plutonium compounds had an absorption type of either M or S (medium or slow). The solubility of plutonium compounds is an important parameter in avoiding "unintentional" homogeneous reactions. Knowledge of this property for both aqueous and organic solvents plays a key role in criticality safety and deserves a high priority.

Unfortunately, little data on particle-size are available, and those that have been generated focus on the reactivity of the materials in the separation and conversion processes. Much of the data are reported as crystallite size, which relates to surface area and solubility but not necessarily to the way the particles would be dispersed in the air. Surface area plays a role in the ability of materials to adsorb gases and vapors that can affect the long-term storage behavior of plutonium compounds. Pressure buildup in storage containers, either from out gassing due to self-heating or radiolytic effects, will depend on the stability of the compound and the amounts of chemisorbed or physisorbed water or other substances.

The following sections discuss the essential compounds of plutonium: plutonium nitrate and associated compounds, plutonium dioxide, plutonium hydride, plutonium sulfate, plutonium chlorides, and plutonium fuel mixtures.

2.3.3.1 Plutonium Nitrate, Oxalate, Peroxide, and Fluorides

Plutonium (IV) nitrate is the most used of all plutonium compounds. Essentially all chemical processing of plutonium has been conducted in nitrate solutions. These solutions of appropriate acidities range from concentrations of 10g to 250g of Pu/L for efficient precipitation processes. Intermediate compounds are also used in the processing of plutonium prepared from the nitrate: plutonium (III) fluoride, plutonium (II or IV) oxalate, and plutonium peroxide. Plutonium (IV) fluoride can be prepared from any of the preceding solids by hydrofluorination. Plutonium fluoride has been the compound of choice for reduction to the metal with calcium, principally because it is nonhygroscopic. The solubilities in various media, bulk densities, and particle sizes of these compounds are given in Table 2.6.

	Measured Solubility		Bulk	-	
Compound	Medium	g/PuL	Filter Cake	Dry Compound	Sintered Media Porosity, um ^(a)
					, , , , , , , , , , , , , ,
Flouride (III)	1M HF – 1M HCl	0.03		1-2.5	15-20
Fluoride (IV)	$2M HF - 2M HNO_{2}$	0.70	0.6-0.8	0.5-2.0	15-20
	$0.31 \times 10^{-2} \times 10^{-2}$	3 0.01	0.6-0.8	-	15-20
Oxalate (IV)	$0.1M C_2 O_{42} - 4M HNO_{3}$	3 0.003	0.5-0.6	0.6	15-20
Peroxide (IV)	$3M H_2O_2 - 1M HNO_3$	0.10	0.10-0.6	-	30-80

Table 2.6. Solubilities and Properties of Selected Compounds

(a) Sintered media porosity required to remain precipitate.

Plutonium hexafluoride is the only volatile plutonium compound (bp 62° C) and is marginally stable. It can be prepared by oxidizing PuF₄with F₂ at an elevated temperature (Weinstock and Malm, 1956). It can also be prepared at low temperatures by a fluorinating agent, fluorine dioxide (Malm et al., 1984). Plutonium waste treatment and decontamination may benefit from processes using photolysis or microwave discharge to produce active fluorine species from FOOF or CF₄/O₂ mixtures, which will react with plutonium or plutonium dioxide to form PuF₆(Martz et al., 1991).

2.3.3.2 Plutonium Dioxide

Plutonium dioxide may now be the most important and most thoroughly studied of all plutonium compounds. Due to its chemical stability and relative inertness, it is the preferred form for shipping and storing plutonium at the present time. Direct oxide reduction (DOR) of PuO_2 is part of the integrated pyrochemical system used at the Los Alamos National Laboratory (LANL) (Christensen and Mullins, 1983; Mullins et al., 1982). Plutonium dioxide is formed when plutonium or its compounds (except the phosphates) are ignited in air, and often results when oxygen-containing compounds are heated in vacuum or in an inert atmosphere to $1000^{\circ}C$ (Cleveland, 1970). The properties of PuO_2 are reported by Moseley and Wing (1965).

Loose PuO₂ powder, as formed by calcination, usually has a density of about 2 g/cm³. If the oxide is pressed and sintered into pellets, it may have a density of about 10.3 to 11.0 g/cm³. Surface measurements of typical oxides prepared from the calcination of plutonium (IV) oxalate at various temperatures range from 10 to 60 m₂/g. Caldwell (1961) found that the surface area decreased with increasing temperatures. Plutonium oxide fired at temperatures >600°C is difficult to rapidly or completely dissolve in common

acids or molten salts. The best solvents are 12-16M HNO₃ with 0.10–0.1M HF, 5–6M HI, and 9M HBr (Cleveland, 1964; Holley et al., 1958). Processes were developed to correct this deficiency using a superacid, HF/SbF₅ (Olaha et al., 1985) and CEPOD, a fluoride-free electrochemical dissolver that used the silver anion as a redox catalyst (Bray et al., 1987).

2.3.3.3 Plutonium Hydride

Plutonium hydride is a compound of interest for separating plutonium scrap from other materials that do not readily unite with hydrogen.² The reaction between plutonium and hydrogen apparently proceeds by the initial formation of PuH2. As more hydrogen is added, the dihydride becomes PuH_{2+x}. The hexagonal PuH₃ begins to form when the H/Pu ratio becomes about 2.75; when the H/Pu ratio reaches 2.9 to 3.0, only the hexagonal form remains. A wide spread is reported in the measured induction period for the first reaction (Haschke, 1991). Because the hydriding reaction is fully reversible, plutonium metal can be recovered by pumping off the hydrogen in a suitable vacuum furnace. This metal typically contains significant amounts of plutonium oxide but is suitable for feed to either molten salt extraction or electrorefining processes. The hydride can also be converted to the oxide. The advantage of the hydride recovery process is its ability to recover a large fraction of the scrap in metallic form. This method, therefore, has a major economic advantage over chemical recycling and subsequent reduction to metal. It is being used as a production aid for metallic scrap recovery.

2.3.3.4 Plutonium Sulfates

Plutonium sulfate tetrahydrate, Pu(SO₄) $_2$ • 4H₂O, has not been of any process importance but has been of interest as a primary standard for plutonium. It is a good example of a stable compound that could be suitable as an interim storage form. Samples stored at relative humidities of up to 75% showed no evidence of alpha radiolysis of the water of crystallization after 28 months. The compound is hygroscopic in air of 95% relative humidity, and stable up to 650°C, at which point it quickly decomposes to PuO₂ (Cleveland, 1970). The potassium salt, K₄Pu(SO₄) ₄- 1H₂O, was under study as a possible primary standard for ²³⁸Pu. Crystals stored in an air-tight steel container, which also functioned as a heat sink, proved to be stable. The solubility product of this compound was determined to be 10⁻¹⁸.

² The properties of plutonium hydrides may be found in Volume 3 of the <u>Handbook of Physics and Chemistry of the Actinides</u> (Ward, 1985). Kinetics of the plutonium hydrogen reaction are reviewed by Haschke (1991).

2.3.3.5 Plutonium Chlorides

Chloride salts, which are a very important category of residues, are byproducts of pyrochemical operations. Pyrochemical chloride-based operations currently in use include:

- -- DOR
- -- electrorefining
- -- molten salt extraction
- -- pyroredox.

Treatment of chloride-based residues is especially challenging for aqueous recovery techniques because of corrosion problems with stainless steel equipment. At the LANL site, Kynar-lined gloveboxes were installed to evaluate their behavior in production-scale operations. The Rocky Flats Plant (RFP) also had extensive experience in aqueous recovery of plutonium from chloride-based residues (Muscatello et al., 1986a, 1986b, 1987). Cesium chloroplutonate, Cs_2PuCl_6 , was a primary analytical standard due to its stability to alpha radiolysis and may now have application as a storage form. It was first prepared by Anderson (1949). There is no evidence of water absorption at relative humidities as high as 53% (Miner et al., 1963). After 64 days at 90% relative humidity, Cs_2PuCl_6 forms a paste.

2.3.3.6 Plutonium Fuels

Plutonium and plutonium-uranium fuel mixtures were developed and tested in experimental reactors to prove the feasibility of operating power reactors. These fuels included both liquids and solids consisting of alloys and ceramic mixtures. Wick (1967) and Schneider and Roepenack (1986) provide comprehensive lists of fuels. Because of their pyrophoric nature, some of these alloys and compounds require special care and handling when exposed to reactive liquids or gases.

2.4 RADIOLOGICAL EFFECTS ON HUMANS

The radiobiological properties of plutonium and other transuranic (TRU) elements are known primarily from experiments performed on rats, dogs, baboons, and rabbits. Human data on plutonium are limited. Reviews of the vast literature on plutonium include Hodge et al. (1973); ICRP 19 (1972); ICRP 30, Part 1 (1979); ICRP 48 (1986); ICRP 30, Part 4 (1988b); and Liverman et al. (1974). Factors affecting radiobiological effects include the mode of entry of plutonium into the body, its distribution in the body, and its transfer to a fetus.

2.4.1 Modes of Entry into the Body

Radioactive material can enter the body by four different pathways: by inhalation, through a wound (including an accidental injection), by ingestion, or by absorption through intact skin. These pathways may occur singly or in any combination.

- -- Inhalation is probably the most prevalent mode for occupational intake of plutonium. It also provides a generally conservative assumption of intake for designing bioassay programs.
- -- Wounds are potentially the most serious mode of intake because of the high dose-per-unit uptake of plutonium. Wounds can result from direct penetration by an object (i.e., a puncture or cut), from abrasion, or from burning by an acid, caustic, or thermal source.
- -- Occupational ingestion of plutonium poses a relatively small risk because the uptake factor from the GI (gastrointestinal) tract to the blood is quite small and because most of the alpha energy from transformations within the GI tract is absorbed by the contents of the GI tract, rather than by the target tissues of the tract itself.
- -- Absorption of plutonium through intact skin is, for practical purposes, almost nonexistent. However, when removing skin contamination, care shall be taken to ensure that the skin integrity is not damaged by rough or extensive decontamination procedures. If the skin integrity is damaged, the result can be considered a wound, regardless of how it occurred.

2.4.2 Distribution Within the Body

Three commonly encountered biokinetic models were promulgated by the ICRP for the internal distribution and retention of plutonium. These models are identified by the ICRP publications in which they were first reported: ICRP 30, Part 1 (1979), ICRP 48 (1986), and ICRP 30, Part 4 (1988b). These models were later updated with models derived from ICRP 60, 67 and 68 values. The models are all similar with regard to the organs of significance, but differ with regard to the fraction of uptake deposited in the organ and its respective retention (or clearance) half-time in the organ.

In all the ICRP models, once plutonium has reached the bloodstream, it is translocated primarily to the liver and skeleton. In the skeleton, it is deposited primarily on the endosteal surfaces of mineral bone, from which it is gradually redistributed throughout the bone volume by resorption and burial. Because of the extremely slow nature of this redistribution, plutonium is considered to be uniformly distributed over bone surfaces at all times following skeleton deposition. A small fraction of the translocated plutonium reaches the gonads. Although the gonadal fraction is different for males and females, the calculated gonadal doses are the same regardless of gender because the plutonium concentration in the tissues is assumed to

be the same. The ICRP assumes that the remainder goes directly to excretion.

Metabolic distribution and retention parameters for the ICRP models are shown in Table 2.7. The table also includes the absorption factors from the GI tract to the bloodstream, as well as the inhalation class of common forms of plutonium.

Table 2.7 includes values from ICRP 60, 67 and 68 models which includes data from the Human Respiratory Tract Model for Radiological Protection Publication 66 (ICRP, 1994) and Age- Dependent Dose to Members of the Public from Intakes of Radionuclides: Part 2 Ingestion Dose Coefficients (ICRP, 1993). The Human Respiratory Tract Model constitutes an updating of the model used in Publication 30 for workers. The new model takes into account extensive data on the behavior of inhaled materials that has become available since the Publication 30 model was developed.

Americium, as an ingrown impurity from the decay of ²⁴¹Pu, can behave the same way as the plutonium host matrix in which it is contained. This implies that the ²⁴¹Am associated with an absorption type S inhalation of plutonium might exhibit absorption type S behavior, rather than the absorption type M behavior assigned by the ICRP. This type of observation was made in ICRP 48 (1996) and by Eidson (1980).

Experience has shown that the biokinetic models in Table 2.7 are subject to some significant variations. A Hanford plutonium-oxide-exposure case described by Carbaugh et al. (1991) has demonstrated lung retention far greater than that expected for a class Y material, leading to the suggestion of a tenaciously retained "super class Y" form. This phenomenon had been informally verified by dosimetry personnel at the Rocky Flats, Savannah River, and Los Alamos sites, and is supported in the literature by Foster (1991). At the other extreme, La Bone et al. (1992) have identified a circumstance in which a ²³⁸Pu oxide inhalation class appeared to exhibit biokinetic behavior more characteristic of an inhalation class D material. These extremes emphasize the importance of addressing the uniqueness of individual workers and exposure circumstances when dealing with known intakes, rather than relying on the assumed standard models.

2.4.3 Transfer to the Fetus

In its most recent review of the metabolism of plutonium and related actinides, it was noted in ICRP 48 (1986) that there is no strong evidence for preferential deposition of plutonium in the fetus and that the concentration of plutonium in the bone of the embryo or fetus is rapidly diluted by growth. However, experimental animal studies have shown that plutonium crosses the placenta after injection in pregnant animals (Green et al., 1979). For fallout plutonium, it has been qualitatively confirmed in humans that plutonium crosses the placenta (Okabayashi and Watanabe, 1973). However, placental and fetal membranes appear to effectively trap a portion of the plutonium that might otherwise reach the fetus.

The behavior of plutonium in the embryo/fetus changes with the development of the embryo/fetus (Sikov, 1987; Sikov et al., 1992). Liver and bone surfaces are the principal sites of plutonium deposition in the embryo/fetus, accounting for approximately 80% of the deposited plutonium (ICRP 48, 1986). Plutonium that deposits on bone surfaces following prenatal or neonatal exposure gradually moves into the bone matrix during subsequent bone-remodeling processes.

The radiation doses produced in the embryonic stage are assumed to be relatively homogeneous and represent a small fraction of the doses received by the pregnant woman when averaged over all tissues. The dose to the fetus would constitute an even smaller fraction of the maternal dose to any tissue in which there was specific deposition (Sikov et al., 1992). As gestation progresses, there is an increase in the relative plutonium concentration in specific fetal tissues, namely the bone and liver (Sikov et al., 1992). Although limited information is available, experimental animal and human data suggest that the average concentration is higher in the fetus during the second or third trimesters than in soft tissues of the pregnant woman, exclusive of the liver, yet significantly less than in maternal tissues of primary deposition, i.e., the bone and liver.

Because placental structures, including the yolk sac, effectively trap plutonium, progenitor cells of the gametes and hematopoietic lines that appear initially in the blood islands of the yolk sac are irradiated while they are primitive stem cells. However, the dose received by the early embryonic cells and the detriment produced is not currently known.

Model Parameter	ICRP 30, Pt 1		ICRP 48		ICRP 30, Pt 4		ICRP 66/67/68
Metabolic	F	Т	F	Т	F	Т	
Distribution ^(a)							
Bone Surfaces	0.45	100 y	0.50	50 y	0.45	50 y	n.a.(c)
Liver	0.45	40 y	0.30	20 y	0.45	20 y	n.a.
Gonads ^(b)							
Male	3.5 x 10-4	ŀ	3.5 x 10-4	ŀ	3.5 x 10-4	ŀ	3.5 x 10-4
Female	1.1 x 10-4	Ļ	1.1 x 10-4	Ļ	1.1 x 10-4	ŀ	1.1 x 10-4
GI Tract							
Absorption							
Factor							
Pu oxides	10-5		10-5		10-5		10-5
Pu nitrates	n.a.		10-4		10-4		10-4
Pu others	10-4		10-3		10-3		5 x 10 ⁻⁴
Am (any)	5 x 10 ⁻⁴		10-3		10-3		5 x 10 ⁻⁴
Inhalation							
Class/Absorption Type							
Pu oxides	Y		Y		Y		S
Pu others	W		W		W		М
Am (any)	W		W		W		М

Table 2.7. Common Biokinetic Models for Plutonium and Americium

(a) F is the fraction of plutonium reaching the bloodstream that is translocated to the organ of concern. T is the retention (or clearance) half-time in the organ of concern.

(b) Plutonium is assumed to be uniformly concentrated in male and female gonadal tissue where it is permanently retained. The deposition fractions are derived based on the relative mass of the reference male and female tissues.

(c) n.a. = not specifically addressed, ICRP 67 lists a relative deposition for adults of 5/3 for skeleton/liver.

2.5 RADIATION EFFECTS ON MATERIALS

The following sections discuss, in order, self-heating and the various effects of radiolysis. Radioactive decay, particularly alpha decay, can and does affect operations in plutonium purification processes. The change in emphasis from plutonium production to waste cleanup, environmental restoration, and the retirement of nuclear weapons will present favorable circumstances for cumulative radiolytic effects, especially in the stabilization processes and the final storage form.

Self-heating and helium retention and release are also included in this section since they too are part of the end result of the alpha decay process. Neutron production from the alpha-neutron reaction is discussed in Section 6.0. The degree of all these effects depends

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2.5.1 Self-Heating

Heat generated by radioactive decay in plutonium, its alloys, or its compounds can be calculated from data provided in Table 2.4, together with the isotopic composition and plutonium fraction. The power output of reactor-produced ²³⁹Pu metal is usually in the range of 2 to 10 W/kg. According to Van Tuyl,³ the equilibrium surface temperature of a metal can that contains 1.2 kg of plutonium at the higher specific power would be 150°C. This calculation is complex because it depends on the thermal conductivities and configuration of all the materials in the shipping container. Thermal diffusivity measurements reported by Kruger and Robbins (1975) were combined with existing heat capacity values to derive a curve for the thermal conductivity of the Pu-1wt% Ga alloy from room temperature to 600°C. Gram quantities of ²³⁸Pu can melt from self-heating under poor heattransfer conditions. The major effects to be expected from self-heating are phase transformation, dimensional changes, chemical reactions (depending on the gaseous environment or other materials in contact with the plutonium), and desorption of previously sorbed gases or vapors.

2.5.2 Radiolysis

In gases, liquids, and covalently bonded solids, the chemical effects of alpha particles and the associated recoil nucleus can cause ionization, excitation, and dissociation of molecules. From the energy requirement for ion pair formation, only about half the energy causes ionization; the other half goes into molecular excitation. Radiation effects are commonly measured by a quantity called the G-value, i.e, the number of molecules destroyed for each 100 eV of energy absorbed. For free radical production, this quantity is expressed as the G_R -value. For organic liquids, G_R -values typically range from 0.85 for carbon disulfide to 70 for carbon tetrachloride (Prevost-Bérnas et al., 1952).

Although there is a considerable body of data on the radiolysis of aqueous solutions, organic liquids, and solids irradiated by gamma rays, X-rays, and fast electrons, little has been published on the radiolysis of plutonium compounds, solvents containing plutonium, or radiation-induced damage in materials that come in contact with plutonium. Nevertheless, radiation-induced damage can affect all aspects of plutonium handling.

³ Van Tuyl, H. H. 1981. "Packaging of Plutonium for Storage or Shipment." Unpublished report by the Pacific Northwest Laboratory task force chairperson to the U.S. Department of Energy.

Radiation-Induced Reaction	Potential Hazard or Damage Problem
Radiolysis of oxygen-contaminated	Production of ozone-damage to elastomers: gloves, seals, etc.
glovebox atmospheres	
Gaseous PuF ₆	Deposition of solid PuF ₄ on equipment
PuO ₂ exposed to hydrocarbons or humid environments	Production of hydrogen gas pressure buildup in nonvented containers.
Ion exchange resins	Damaged resin can react violently with HNO ₃ or other oxidizers. Also may result in hydrogen gas- pressure buildup.
CCl4 saturated with H2O	Production of Cl ₂ . C ₂ Cl ₆ HCl, and phosgene.
Polyethylene	Disintegrates with production of H ₂ .
Polyvinylchloride (PVC) plastics	Disintegrates with production of HCl-corrosion.
Tri-n-butylphosphate	Production of hydrogen and oxygen-pressure buildup in nonvented containers.
Aqueous plutonium solutions	Production of polymeric plutonium hydroxide (plutonium polymer), which plates out on vessel surfaces and piping, producing swelling, cracking, loss of ductility.
Low-acidity plutonium solutions	Increase in leachability.

Table 2.8. Potential	Hazards or D	Damage to I	Materials from	Exposure to	Radiation
	1100200100 01 2	, mininge te i		2	1.0000000000000000000000000000000000000

It would be futile and inappropriate to list, let alone discuss, all the possible radiolytic reactions affecting plutonium-handling. However, it is important to recognize the potential for and anticipate the consequences of these reactions. The following sections cover a broad range of the types of radiation-induced damage common to plutonium handling.

2.5.2.1 Hydrogen Production

The G-value for the production of H_2 by the alpha radiolysis of pure water is 1.9 ± 0.1 molecules of hydrogen per 100 eV (Prevost-Bérnas et al., 1952). Cleveland (1970) calculates that the energy released in 0.001M (0.24 g/L) of plutonium solution is on the order of 2 x 10^{14} eV per minute. Thus, the hydrogen evolution would be approximately 3.8×10^{15} molecules per liter per day for a 1M solution, or about 73 cm³ of hydrogen per year.

The G-values for H_2 in solids irradiated by gamma rays are lower: 0.1 for ice (Johnson, 1970) and 0.01 for the hydrates of a large number of sulfates (Huang and Johnson, 1964). Because the stability of $PuSO_4$. $4H_20$ was found to be remarkably high (Cleveland, 1970), one of the sulfates may well serve as an alternate interim waste form. Dole (1974) summarized the radiation chemistry of polyethylene, quoting G-values for hydrogen as 5 molecules per 100 eV. Destruction of plutonium hexafluoride as the solid phase amounts to about 1.5% of the material per day (Weinstock and Malm, 1956). Cleveland (1970) calculated the mean change in average oxidation number in 0.5–2M of perchloric acid to be 0.018 moles per day, corresponding to a G-value of 3.2 equivalents per 100 eV. The formation of hydrogen peroxide from the radiolysis of water is believed to be the mechanism for the reduction of plutonium (VI)

ions. Lower oxidation states are formed by the disproportionation of the plutonium (V) species.

Pressurization of storage containers holding TRU wastes is a potential hazard for both long and interim storage periods (Kazanjian et al., 1985). Sampling of TRU waste drums shows that hydrogen is usually created (Roggenthem et al., 1989). Waste drums with pinholes can "breathe" when the atmospheric pressure changes, thereby introducing water vapor. Water vapor adsorbed on plutonium compounds is radiolytically decomposed, thereby producing hydrogen. It may be possible to add pressure relief valves and appropriate in-line filters to waste drums. (See Section 2.7 for more information on storage and containment.)

2.5.2.2 Redox Reactions

In most chemical processes for purifying plutonium, it is essential to maintain its valence state. The formation of hydrogen peroxide from the radiolysis of water is believed to be the mechanism for the reduction of plutonium (VI) ions. Lower oxidation states are formed by the disproportionation of the plutonium (V) species. Cleveland (1970) calculated the mean change in average oxidation number in 0.5-2M of perchloric acid to be 0.018 moles per day, corresponding to a G-value of 3.2 equivalents per 100 eV.

In the radiolysis of solutions, the presence of other ionic species can accelerate or inhibit the disproportionation of plutonium valence states. For example, the presence of the chloride ion in plutonium (VI) solutions prevents reduction to plutonium (IV). Reactions may reverse after long irradiation periods, in which case a steady-state condition should ultimately be reached, resulting in a net decomposition rate of zero. An excellent review of the radiation chemistry of plutonium nitrate solutions may be found in Miner and Seed (1966). In dilute solution (0.1M), G_{H2} is about 0.5 and G_{02} increases to 1.45. Self-reduction of Plutonium hexafluoride as the solid phase amounts to about 1.5% of the material per day (Weinstock and Malm, 1956). See Cleveland (1970) Chapter 2, for more information.

2.5.2.3 Miscellaneous Radiolytic Reactions

A serious limitation to the use of organic ion exchange materials is their radiation stability. Brookhaven National Laboratory (BNL) reviewed the literature and summarized the effect of ionizing radiation on both organic and inorganic ion exchange materials (Gangwer et al., 1977). Extraction of plutonium (IV) from 3M HNO₃ into 30 vol% tributyl phosphate in kerosene at 5°C decreased the extraction coefficient by a factor of two when irradiated to a dose of 3.6 x 10⁷ R (Tsujino and Ishihara, 1966). The mechanical properties of thin plastic films such as polyethylene and polyvinyl chloride degrade with exposure to plutonium. Cellulose vacuum-cleaner bags

will disintegrate in less than a month if used for housekeeping purposes in plutonium-contaminated gloveboxes. Leachability of plutonium-containing wastes could be affected by the production of nitric acid for air-equilibrated dilute salt solutions (Rai et al., 1980).

2.5.2.4 Helium Retention and Release

Helium introduced by alpha-bombardment of plutonium and the alloys and compounds of plutonium can cause lattice expansion. This was first observed for plutonium oxides, carbides, and nitrides by Rand et al. (1962) and was later observed for two plutonium carbide phases. Helium is retained in vitrified compounds. The retention and release behavior of helium in plasma-torch-fused Pu0, microspheres for SNAP is an important parameter in the design of the heat source. Approximately 530 cm³ at standard temperature and pressure per year-kg are produced by ²³⁸Pu0₂ (Stark, 1970). Microspheres of 80% ²³⁸PuO₂and 20% ²³⁹PuO₂ that were approximately 50 mm in diameter, prepared by the sol-gel process, released 92.8% of the helium in 8 months at room temperature (Northrup et al., 1970). Metals at temperatures well below the melting point trap the insoluble helium gas in tiny bubbles, which are more or less evenly distributed through the matrix material (Stevens et al., 1988). Helium buildup in weapon-grade material is approximately 4 standard cm³ per year-kg.

2.6 OCCUPATIONAL HAZARDS

The major industrial hazard in plutonium facilities is the potential for loss of control of a highly toxic substance, resulting in either the inhalation or ingestion of plutonium or one of its compounds by personnel, or the exposure to excessive radiation from a criticality accident. The possibility of a fire or explosion in a plutonium facility is probably the most serious threat because the consequences of a fire could lead to loss of containment and subsequent dispersal of highly mobile plutonium particulates. In addition, fighting the fire with water to maintain containment could create the potential for a criticality accident and/or loss of containment in the immediate vicinity.

The day-to-day hazards for personnel in plutonium facilities involve exposure to gamma rays, X-rays, and neutrons, as well as possible accumulation of plutonium in the body. These hazards are described in more detail in Section 3.0, "Radiation Protection," and Section 7.0, "Nuclear Criticality Safety." The amount of plutonium needed to present potential hazards to personnel in plutonium-handling facilities is summarized in Figure 2.4. Hazards related to interim and long term storage of plutonium will be found in Section 2.7, "Storage and Containment."

2.6.1 Chemical Versus Radiological Hazards

The radiological toxicity of reactor-produced plutonium far exceeds the chemical toxicity of this heavy element. Furthermore, its low solubility in near-neutral or basic solutions reduces the uptake through ingestion by a factor >1000 for any plutonium compounds except certain complexes, such

as the citrate or ethylenediamine tetraacetic acid complex. (Refer to Sections 2.3, "Radiobiological Properties," and 6.0, "External Dose Control"). Tipton (1960) summarizes the differences in chemical hazards between plutonium and uranium: "In contrast to uranium, the chemical toxicity of plutonium is insignificant in comparison to the hazard arising from its natural radioactivity." Moreover, "the toxicity of plutonium and other transuranic elements," according to Voelz et al. (1985), "has only been studied in animals since acute toxicity has never been observed in man for these elements and epidemiologic studies have not produced positive results." However, recent evidence suggests that plutonium can catalyze reactions including oxidative stress in the absence of significant radioactive decay. These data presented by Claycamp and Luo (1994) suggest that plutonium complexes might contribute to long-term oxidative stress related to tumor promotion.

2.6.2 Associated Chemical Hazards

The main chemical hazard of plutonium is its vulnerability to oxidation and the pyrophoricity of some of its alloys and compounds (see Section 2.6.3).

The processing of plutonium, including separation from irradiated uranium, purification, conversion, waste disposal, environmental restoration, and D&D, necessarily requires the use of chemicals and reagents with varying degrees of toxicity and hazardous properties. A partial list of chemicals that have been used at DOE plutonium facilities is provided in Table 2.9. An abbreviated evaluation of the potential hazards of these substances is also provided. Table 2.9 is not meant to replace the Material Safety Data Sheet available from chemical manufacturers; rather, it is intended to help readers recognize the toxicity of these chemicals and identify any possible side effects from their use that could jeopardize radiation safety or plutonium containment.

2.6.3 Hazards Created by Oxidation and Pyrophoricity

This section describes the oxidation and burning characteristics of plutonium, summarizes the storage properties of the metal and oxides, and presents recommendations for their storage conditions. Waste remediation plans for TRU materials and the necessity for dealing with ton quantities of plutonium metal from the retirement of weapons require the identifying of long-term and intermediate-term waste forms with appropriate stability. Economic considerations make clear the importance of generating few, if any, new wastes in accomplishing this task.

2.6.3.1 Oxidation of Plutonium

The problems of oxidation of metallic plutonium were recognized shortly after the discovery of plutonium, and extensive studies of the low-temperature corrosion of plutonium and its alloys have been performed. Oxidation can produce fine loose plutonium oxide, which disperses easily in glovebox systems, complicating housekeeping chores. If not controlled, loss of accountability and

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increased radiation exposure to personnel is certain. The reactivity of plutonium metal is discussed in Section 2.3.1. The tendency for enhanced oxidation is promoted by the self-heating properties of plutonium isotopes (discussed in Section 2.5.1). A kilogram of ²³⁹Pu can easily reach an equilibrium temperature of 80°C in a glove-box environment (Raynor and Sackman, 1967). Thermally isolated ²³⁸Pu metal can easily melt from its own decay heat. The heat generated by oxidation may be sufficient to ignite nearby combustible materials. Metal turnings and scrap should be reprocessed or converted to stable alternatives as soon as practicable. Plutonium metal, its alloys, and its reactive compounds need to be excluded from both oxygen and water vapor, but especially the latter since it catalyzes and accelerates oxidation.





Chemical Name	CAS No.(b)	Hazard	Formula
Aluminum nitrate	7784-27-2		$AL(NO_3)_39H_2O$
Antimony pentafluoride	7783-70-2		SbF ₅
Beryllium (metal)	7740-41-7	Neutron	Be
Calcium (metal)	7740-70-2	Release H_2 when wet,	Ca
		flammable	
Calcium oxide	1305-78-8	Corrosive	CaO
Calcium chloride	10043-52-4		CaCl ₂
Ferrous ammonium sulfate	7783-85-9		$Fe(SO_4)(NH_4) _2 SO_4 6H_2O$
Fluorine	7782-41-4	Oxidizer, poison	F ₂
Fluorine dioxide		Oxidizer, poison	F ₂ O ₂
Carbon tetrachloride	56-23-5		CCl ₄
Ferrous sulfamate			Fe (SO ₃ NH ₂) ₂
Gallium (metal)	7440-55-3		Ga
Hydrogen	1333-74-0	Flammable, explosive	H ₂
Hydrogen Fluoride	7664-39-3	Corrosive	HF
Hydrochloric acid	7647-01-0	Corrosive	HCl
Hydrogen peroxide	7722-84-1	Oxidizer	H_2O_2
Iodine	7553-56-2	Poison	I ₂
Magnesium (metal)	7439-95-4	Water reactive, flammable	Mg
		when combined with Pu	
Magnesium chloride	7786-30-3	Neutron	MgCl ₂
Magnesium oxide	1309-48-4	Neutron	MgO
Mercuric nitrate	10045-94-0	Oxidizer, poison	$Hg(NO_3)_2$
Nitric acid	7697-37-2	Oxidizer, corrosive, poison	HNO 3
Oxalic acid	144-62-7	Poison	$H_2C_2O_4$
Potassium hydroxide	1310-58-3	Corrosive, Poison	КОН
Potassium chloride	7447-40-7		KCI
Sodium chloride	7647-40-7		NaCl
Sodium Hydroxide	1310-73-2	Corrosive, poison	NaOH
Sodium nitrate	7631-99-4	Oxidizer	NaNO ₃
Stannous chloride	7772-99-8		SnCl ₃
Sulfamic acid	5329-14-6	Corrosive	NH ₂ SO ₃ H
Sulfuric acid	7664-93-9	Corrosive, poison	H ₂ SO ₄
Tri-n-butyl phosphate	126-73-8	Flammable liquid	$(C_4H_7O)_3PO_4$
Urea	57-13-6		$CO(NH_2)$
Uranium (metal)		Flammable	U
Zinc chloride	7646-85-7		ZnCl ₂
Soltrol 170 Phillips 66	68551-19-9	Flammable liquid (isoparafins)	(Mixture C10–C14)
Carbon tetrafluoride	75-73-0		CF ₄

Table 2.9. Hazards of Chemicals Used in Processing Plutonium ^(a)

(a) Refer to Material Safety Data Sheets for complete discussion of hazards.

(b) Chemical Abstracts Service Registry number.

The corrosion or oxidation of plutonium does not always occur in a linear or predictable manner. The oxidation rate is a complex function of the surrounding atmosphere, the moisture content, and the alloys or impurities present in the metallic plutonium.⁴

2.6.3.2 Ignition Temperatures and Pyrophoricity of Plutonium, Its Alloys, and Its Compounds

Plutonium and some of its alloys and compounds are pyrophoric. Pyrophoric material is a liquid or solid that, even in small quantities and without an external ignition source, can ignite within 5 minutes after coming in contact with air (NFPA Fire Protection Handbook). Pyrophoric plutonium metal has been defined as "that metal which will ignite spontaneously in air at a temperature of 150°C (320°F) or below in the absence of external heat, shock, or friction" (Stakebake, 1992).⁵ Finely divided plutonium metal would be considered pyrophoric while massive plutonium would be nonpyrophoric. Martz et al. (1994) has proposed a mechanism for plutonium pyrophoricity that predicts the ignition temperature as a function of surface mass ratio and particle size. The most numerous forms of pyrophoric plutonium are chips, lathe turnings, and casting crucible skulls. Plutonium hydride and sesquioxide (Pu_2O_3) are probably the most commonly occurring pyrophoric compounds. Plutonium carbide, oxycarbide, nitride, and oxide phases with compositions between the sesquioxide and dioxide are potentially pyrophoric. Known pyrophoric alloys include Pu-U and Pu-Ce, Waber (1967) summarized much of the early work on plutonium corrosion and oxidation and is a good source for identifying other pryophoric alloys.

⁴ See Wick (1967), Coffinberry and Miner (1961), and Kay and Waldron (1966) for details on the oxidation of unalloyed plutonium and the stabilized alloy of plutonium.

⁵ Also in DOE/DP-0123T, Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities (DOE, 1994a).

The health physics aspects of an accidental plutonium fire can be serious. A fire can burn through containment structures, resulting in the dispersal of PuO₂ over a wide area, with the potential for inhalation exposure during the fire or during subsequent decontamination efforts. The conditions under which a plutonium fire can occur in a dry glovebox have been studied. With only 5% oxygen in nitrogen, the metal will burn easily. At the 1% level, however, a fire will not continue to burn unless heat is supplied (Rhude, 1962). Turnings shall be generated in a dry atmosphere and should be converted to the oxide as soon as convenient, preferably on the same day they are made. Some solvents and organic compounds form flammable mixtures with plutonium. In one incident, tetrachloroethane was inadvertently substituted for another lathe coolant in a metal-turning operation. Chips of plutonium aluminum alloys were ignited, resulting in the blowout of a glove-box panel. In a separate event, burning plutonium chips dropped into carbon tetrachloride resulted in an explosion (AEC, 1965).

2.6.3.3 Aerolization of Plutonium

The ignition of plutonium metal becomes a major hazard when enough plutonium has burned to produce a significant amount of dispersible material and a serious enough fire to damage the pertinent containment structures. The particle size of PuO₂ fired at a low temperature varies from 3% at <1 μ m to 97% at 1-5 μ m (Stakebake and Dringman, 1967). Sintered PuO₂ has a particle size <2 μ m. Haschke (1992) made an effort to define the maximum value of the source term for plutonium aerosolization during a fuel fire. He found the rate to be constant (0.2-g PuO₂/cm² of metal surface per minute) above 500°C. The mass distribution for products of all metal gas distributions are approximately 0.07 mass% of the oxide particles having geometric diameters ≤10 μ m.

2.7 STORAGE AND CONTAINMENT

The DOE mission for utilization and storage of nuclear materials has changed as a result of the end of the "Cold War" era. Past and current plutonium storage practices largely reflect a temporary, in-process, or in-use storage condition which shall now be changed to accommodate longer-term storage.

The DOE has sponsored a number of workshops on disposing of plutonium. Two of the objectives of these workshops have been to make recommendations for near-term and long-term storage forms and to identify possible alternatives. At the Hanford Plutonium Disposition Workshop held in Richland, Washington, from June 16 to 18, 1992, the two highest ranking stabilization processes were, first, denitration of plutonium nitrate, and, second, thermal stabilization. The third-ranked process included the precipitation of Cs_2PuC1_6 or $K_4Pu(SO_4)_4$ followed by thermal stabilization (Hoyt, 1993). At the workshop on plutonium storage sponsored by DOE Albuquerque, on May 26 and 27, 1993, both metal and oxide were considered suitable storage forms. A report has been issued summarizing information presented here and resulting from this workshop (DOE, 1994a). This important report includes sections on:

- -- materials properties relevant to storage;
- -- current storage practice;
- -- advanced storage concepts;
- -- hazard analysis; and
- -- recommendations.

Existing storage and handling requirements for plutonium metal and oxides are currently covered in DOE Order 460.1D, <u>Hazardous Materials Packaging and Transportation</u> <u>Safety</u> (DOE, 2016d). DOE M 441.1-1, <u>Nuclear Material Packaging Manual</u> (DOE, 2008e) and DOE-STD- 3013-2018, <u>Stabilization, Packaging, and Storage of Plutonium-Bearing Materials (DOE, 2018b</u>), also provide information on packaging plutonium material.

The following property summaries adapted from Haschke and Martz (1993), are useful for determining potentially unsuitable storage and containment conditions for plutonium metal and oxide. Given that plutonium metal is chemically reactive in air and other environments, it also:
- -- Exhibits spontaneous self-sustained ignition (becomes pyrophoric) only if the metal dimension
 - is <0.1 mm and T >150°C
 - is >0.2 mm and T >500°C
- -- reacts slowly in air at room temperature (maximum of about $1 \mu m/day$)
- -- has limiting (T-independent) oxidation rate in air above 500°C
- -- is not a dispersible form (<10 μm geometric size) until oxidation occurs:
 - oxide from Pu+Air at ambient T: 100 mass % (ssa = $10-20 \text{ m}^2/\text{g}$)
 - oxide from PuH_2+O_2 : ~25 mass % (ssa ~ 1 m₂/g)
 - oxide from Pu+O₂and Pu+Air at T >500°C: $< 0.1 \text{ mass } \% \text{ (ssa } <0.1 \text{ m}^2/\text{g)}$
- -- radiolytically decomposes organic and covalently bound specific species in the environment
- reacts with most radiolytically produced gases and with nonequilibrium surface:
 limits pressurization by gases
 - forms low-density (pressure-generating) and pyrophoric products
- -- retains helium from alpha decay
- -- is stabilized by certain storage atmospheres (reactivity decreased by 10^{12})
- -- is stable if isolated from reactive species
- -- has good storage history when stored properly.

A similar property summary for plutonium dioxide, the most commonly used form of plutonium, shows it to be stable and unreactive in air. Storage and containment recommendations, based on the properties of plutonium metal and dioxide, are shown in Table 2.10.

Table 2.10. Storage Recommendations for Plutonium Metal Dioxide (adapted from Haschke and Martz, 1993)

- Metal and oxide are both suitable storage forms for plutonium (100 years).
- Organics (plastics, elastomers) shall be excluded from the primary container for both forms.
- Converting between metal and oxide is not recommended (negative impact of waste, cost, environmental safety and health risk).
- Both forms shall be properly prepared and certified:
 - Procedures for metal already exist (technology transfer needed).
 - Procedures for oxide need development (stabilization, desorption, loss on ignition.
- Both forms shall be in sealed primary containers for extended storage:
 - Positive seals (e.g., welds and metal seals) are necessary.
 - Seal certification or double sealing is necessary.
- Requirements diverge for short-term/retrievable storage:
 - Containers with metal gaskets are advantageous for metal storage.
 - After stabilization, oxide is best stored in a container fitted with a rupture disk in series with a vented stainless-steel frit container.
- Surveillance of stored materials is required.

Note: DOE M 441.1-1, <u>Nuclear Material Packaging Manual</u> (DOE, 2008e) and DOE-STD-3013-2018, <u>Stabilization, Packaging, and Storage of Plutonium-Bearing Materials</u> (DOE, 2018b), also provide information on packaging plutonium material.

3.0 RADIATION PROTECTION

The radiation protection field is concerned with the protection of individuals, their progeny, and humanity as a whole, while still allowing for necessary activities which might involve radiation exposure. The aim of radiation protection is to prevent deterministic effects and to limit the probability of stochastic effects. Most decisions about human activities are based on an implicit form of balancing risks and benefits leading to the conclusion of whether or not the application of a particular practice produces a positive net benefit. Because the probability of health effects is not zero, the ICRP in Publication 26 (ICRP, 1977) recommended the following criteria for a system of dose limitation:

- -- No practice shall be adopted unless its introduction produces a positive net benefit.
- -- All exposures shall be kept as low as reasonably achievable, with economic and social factors being taken into account.
- -- The dose equivalent to individuals shall not exceed the limits recommended for the appropriate circumstances.

These criteria and related information have been incorporated into DOE regulations, instructions, and manuals for radiation protection.

The successful operation of a plutonium facility requires scrupulous attention to providing adequate radiation protection and maintaining contamination control through the implementation of a quality health physics program. (In this section, "health physics" and "radiation protection" can be used interchangeably when referring to programs or personnel.) Prompt dose assessment is important for demonstrating compliance with standards, providing information to workers, establishing an accurate historical record, and for responding to accident and incident situations. This section defines the basis for the establishment of a sound health physics program at a plutonium facility.

3.1 REGULATION AND STANDARDS

Regulations on radiation protection in DOE and DOE contractor facilities are found in 10 CFR 835, <u>Occupational Radiation Protection: Final Rule (DOE, 2011)</u>. Guidance is found in the supporting document <u>Radiological Control</u> (DOE, 2017) and the 10 CFR 835 Implementation Guide G 441.1-1C, Ch 1 (DOE, 2011a).

10 CFR 851 specifies health and safety regulations, which also apply to workers in DOE facilities. Other related source documents include publications of the U. S. Environmental Protection Agency (EPA), American National Standards Institute (ANSI), ICRP, National Council on Radiation Protection and Measurements (NCRP), and United Nations Scientific Committee on the Effects of Atomic Radiation. 10 CFR 851, <u>Worker Safety and Health</u> <u>Program</u> (DOE, 2019) provides requirements for worker safety and health. The worker safety and health program integrates the Rule's requirements with other site worker protection activities and the integrated safety management system (ISMS) [851.11(a)(3)(ii)].

In addition, each site that handles radioactive materials and/or radiation generating machines is required to establish and maintain its own documented radiation protection program, following the Federal regulations.

3.2 RADIATION PROTECTION PROGRAMS

Radiation protection programs include provisions for quality assurance, administrative controls, protection of visitors, visits by regulatory personnel, and onsite packaging and transportation of hazardous materials.

3.2.1 Quality Assurance

It is highly desirable for laboratories and industrial facilities handling plutonium to have a well-integrated quality assurance program. Such a program should have high visibility and strong management support. Quality assurance should be effectively applied throughout facility activities, including the radiation protection program. The basis for quality assurance programs in DOE facilities is established in 10 CFR 830, Nuclear Safety Management (DOE, 2011j). In addition, 10 CFR 830.120, Quality Assurance Requirement, requires the development of a Quality Assurance Program, specifies an implementation schedule, and provides the elements that the program shall address.

An effective quality assurance program for radiation protection will include establishment of appropriate standards of performance for essential activities and equipment, with an effective system of documentation and traceability of those activities and of the use of the equipment. Proper maintenance of those records will be necessary for reference purposes.

3.2.2 Administrative Controls

In any facility that handles radioactive materials, the major controls protecting workers, the public, and the environment are structures and installed equipment, which shield, contain, and confine the radioactive materials. However, to allow useful work to be performed in the facility and to assure that its protective features remain effective, a number of administrative controls are ordinarily required. These administrative controls are usually contained in a series of procedures related to the operations and maintenance activities to be carried out in the facility. All personnel who work in controlled areas should be familiar with the administrative controls are made, these changes or additions should be effectively communicated to all persons who may be affected.

3.2.2.1 Radiation Protection Procedures

A plutonium facility should have a written policy on radiation protection, including a policy on keeping exposures as low as reasonably achievable (ALARA). All radiation protection procedures and controls should have formal, recognizable technical bases for limits, methods, and personnel protection standards. Procedures should be adequately documented, updated periodically, and maintained in a centralized historical file. A control system should be established to account for all copies and ensure that all new procedures are included in the historical files. A designated period of time for maintaining historical files should be established. DOE Order 200.1A, Information Technology Management (DOE, 2008b) and ANSI/HPS N13.6 (ANSI, 2010) provide guidance on how to maintain historical files. In addition, radiation protection procedures should have a documented approval system and established intervals for review and/or revision. A tracking system should be developed to ensure that the required reviews and revisions occur.

Radiation protection procedures should be provided for but not limited to the following topics:

- -- Posting and labeling of facilities
- -- development and maintenance of all radiation protection records
- -- reporting of unusual radiation occurrences
- -- use of radiation monitoring instruments
- -- use of radiation sources (e.g., reference calibration)
- -- reporting of radiation exposures
- -- use of protective clothing
- -- responding to radiological emergency events
- -- surveying and monitoring
- -- counting room equipment and use
- -- instrument maintenance and control
- -- development and use of Radiological Work Permits (RWPs)
- responsibilities of operations staff for contamination control and personnel surveys.

Two topics, RWPs and facility posting and labeling, are discussed below in more detail.

3.2.2.2 Radiological Work Permits

10 CFR 835.501(d) requires written authorizations to control entry into and perform work within radiological areas. These authorizations shall specify radiation protection measures commensurate with the existing and potential hazards. Radiological Work Permits are a type of written authorization used for entry into high and very high radiation areas, high contamination areas, and airborne radioactivity areas. The RWPs also are used to control entry into radiation and contamination areas and for handling materials with removable contamination. The RWPs should be initiated by the work group responsible for the activity. All RWPs should be reviewed and approved by the radiation protection staff. Radiological

Work Permits are recommended for other radiological work in accordance with the standard, <u>Radiological Control</u>, (DOE, 2017). Guidance for posting of RWPs and for their contents is contained in the standard, <u>Radiological Control</u>.

Radiological workers should read and understand the applicable RWP before performing work in a radiological area. The RWPs should be located at the access point to the applicable radiological work area. Workers should acknowledge by signature or through electronic means that they have read, understood, and will comply with the RWP before they initially enter the area and after changes. Out-of-date RWPs should be removed.

3.2.2.3 Radiological Surveys and Data Trending

Area monitoring in the workplace shall be routinely performed, as necessary, to identify and control potential sources of personnel exposure (10 CFR 835.401(a) (6)). This monitoring should include surveys in areas that are not ordinarily expected to be contaminated. The program should define minimum requirements, survey types, and frequencies.

Surveys should be performed at frequencies adequate to identify changes in posting required or an activity buildup, and to ensure that current radiological controls are appropriate. The surveys suggested by this section are minimum recommendations; additional surveys should be conducted, recorded, and reviewed as necessary to ensure full protection of personnel.

Contamination surveys should be performed to determine contamination area (CA) boundaries, the appropriate posting of sources or areas, and the location and extent of localized contamination.

Contamination surveys should be performed and documented prior to the start of radiological work, during general work activities at times when changes in contamination level may occur, and following work to assure that final radiological conditions are acceptable and documented. See Munson et al. (1988).

A sufficient number of points should be surveyed to adequately assess the radiological status of the area being surveyed.

Routine radiological surveys should be regularly conducted, recorded, and reviewed for all areas where personnel could be exposed to alpha, beta, gamma, X-ray, or neutron radiation throughout the site. Surveys should be performed at frequencies adequate to ensure protection of personnel. The following surveys should be considered the minimum. Additional surveys should be conducted, recorded, and reviewed as necessary to ensure that personnel exposures are maintained ALARA.

General radiation and contamination surveys should be performed:

- -- To identify and verify the boundaries of areas which shall be radiologically controlled.
- -- to verify that radiation and contamination Levels outside of radiological areas remain less than specified limits.
- -- to determine the appropriate posting of localized higher radiation levels, beams, or hot spots.
- -- to ensure that radiological conditions are acceptable and documented prior to, during, and at the completion of work that may cause changes in radiation levels to occur (see Munson et al., 1988, p. 6.1.2).
- -- to satisfy required predetermined procedure hold-points in work areas and adjacent areas, whenever operations are performed that may cause significant increases in radiation levels. The survey may be required as part of a radiological inspection step required by the work procedure. This includes areas above and below the work area as appropriate during special processing operations or cell decontamination, movement of permanent or temporary shielding, radioactive waste processing, and relocation of highly radioactive materials.

Routine radiation and contamination level surveys should be performed in the workplace at a frequency commensurate with the radiation hazard, to detect trends related to equipment, systems, environment, and work habits.

Non-routine surveys of radiation and contamination levels in the workplace should be performed:

- -- Before initial use of a new installation, system, or equipment, or as soon as possible after a radiation source is brought into the area.
- -- whenever changes in procedures, equipment, or sources have occurred that may cause changes in the external radiation levels.
- -- after modification to a shield or changes in shield materials.
- -- as the basis for trend evaluation of external radiation level conditions.
- -- when a radiological accident has occurred or is suspected.
- -- when requested by the personnel performing the activity (see Munson et al., 1988, p. 6.1.2).

Radiation surveys should be performed upon initial entry into process cells and tanks that contain radioactive piping or components.

Surveys should be conducted when performing operations that might result in personnel being exposed to small intense beams of radiation (e.g., removing shielding for shielded X-ray devices).

Every reasonable effort should be made to maintain the radiation dose of the surveyor at levels that conform to ALARA guidance.

Surveys should be performed and documented according to established procedures.

Only fully trained and qualified personnel should conduct surveys that are to be the official records of radiation levels or for the protection of personnel; these surveys should be reviewed and approved by the Radiological Protection Manager or his/her designee.

Surveys should be performed with calibrated instrumentation appropriate for the intensity and energy of the radiation anticipated in the area to be surveyed.

Survey instruments should meet the performance check requirements established by the facility in accordance with ANSI N323a (ANSI, 1997b).

Combinations of survey instruments should be used as necessary to provide the capability to measure all types of radiation and dose rates characteristic of that which could be encountered at the facility being surveyed.

Records that establish the conditions under which individuals were exposed to external radiation (such as facility radiological conditions records generated by the monitoring programs) should be retained to provide a chronological and historical record. See ANSI/HPS-N13.6 (ANSI, 2010).

A sufficient number of points should be surveyed in order to adequately assess the radiological status of the area. Regular predetermined points may be used, but additional spot monitoring should be done to ensure that all changes in dose rates are identified, recorded, and reviewed.

All records of surveys should clearly identify, as a minimum:

- -- The name, signature, and employee number of the surveyor.
- -- survey instrument(s) model number, serial number, and calibration date.
- -- the type(s) of radiation being monitored (e.g., neutron, gamma, etc.).
- -- the dose rates.
- -- the date and time the survey was performed.

-- locations where radioactive material is located temporarily (or is being temporarily stored) or where equipment that generates ionizing radiation is being operated.

Records of the results of radiation surveys should be retained in accordance with facility policy.

Survey data should be reviewed by supervisory personnel. Significant findings should be presented to the facility manager in a timely manner.

Health physics personnel should summarize survey data in each building or area at least once a quarter. Significant changes or trends in area dose rates and/or radiological contamination should be noted and corrective actions assigned. The survey summary should be presented to the facility management quarterly.

Survey results and data summaries should be made available to the ALARA team chair periodically and should be used:

- -- To provide a basis for evaluating potential worker exposure on a job and in ALARA preplanning.
- -- to provide a baseline for trend analysis, investigation, and correction of unusual conditions.
- -- to track the status of jobs (including identification of good practices) and to detect departures from good operating procedures and/or the failure of radiation controls.
- -- to identify the origin of radiation exposures in the plant by location, system, or component.

Health physics personnel should post the results of radiation surveys or survey maps at the entrance to all permanent radiation areas, high radiation areas, and very high radiation areas. The results should be posted in the form of a survey map so that personnel can be aware of the locations of higher and lower levels of radiation within the area.

A survey data trending program should be conducted; to indicate the continuing effectiveness of existing control; to warn of deterioration of control equipment or effectiveness of operating procedures; to show long-term variations in radiation levels; and to identify and correct improper radiation work practices.

Health physics should perform trend analyses on all permanent radiation, high radiation, and very high radiation areas. At a minimum, one complete survey record should be evaluated and included in the trend analysis program for each survey required to be performed by the facility routine control program.

Health physics should use the facility reporting system to identify discrepancies and abnormal trends and should summarize the data review results in their monthly reports to the Radiological Protection Manager.

Survey data trends should be investigated when either:

- -- an upward trend in general area radiation level occurs, causing a significant increase.
- -- an abrupt change in radiation level occurs that cannot be directly correlated to normal activities.

3.2.2.4 Facility Posting and Labeling

Areas in plutonium facilities shall be posted in accordance with the requirements in 10 CFR 835 (DOE, 2011). Chapter 12 of Implementation Guide G 441.1-1C, Ch. 1 (DOE, 2011a) provides guidance to ensure compliance. The technical criteria and dose rate and/or levels for defining radiation, high radiation, very high radiation, contamination, high contamination, and airborne radioactivity areas are established in 10 CFR 835. The health physics staff should identify:

- -- Areas to be barricaded and marked to prevent personnel from inadvertently entering them.
- -- Areas to be physically controlled per 10 CFR 835, Subpart F.

Entrance to radiological areas shall be controlled (10 CFR 835.501(a and b)) commensurate with the existing and potential radiological hazard within the area.

The health physics staff should post current radiation surveys of radiation areas at the health physics access control point for use in prejob planning. Airborne Radioactivity Areas shall be posted with the words, "Caution, Airborne Radioactivity Area" or "Danger, Airborne Radioactivity Area" when the airborne radioactivity levels in the occupied area exceed, or are likely to exceed, the derived air concentration (DAC) value listed in Appendix A or Appendix C of 10 CFR 835 or where an individual could receive 12 DAC hours in a week (10 CFR 835.603(d)). These areas are posted to alert personnel of possible respiratory protection requirements.

3.2.2.5 Unposted Areas

Certain areas of facilities that handle radioactive materials should be maintained free of detectable radioactive contamination. These areas should also be maintained at ambient radiation levels equivalent to the environmental background of the facility. Parts of the facility that should meet these requirements include lunchrooms, offices, restrooms, janitor rooms, corridors outside operational areas, foyers, and outside areas surrounding the facility, including the building roofs.

To assure these areas meet the requirements of radiological cleanliness, they should be surveyed with count-rate instruments sensitive to the radioactive isotopes of interest. In a plutonium facility, the instruments should meet the requirements listed in ANSI Standard N317-1991, <u>Performance Criteria for Instrumentation Used for In-Plant Plutonium Monitoring</u> (ANSI, 1980a). These clean areas should be maintained below the surface contamination levels cited in 10 CFR 835 (DOE, 2011).

3.2.3 Visitors

Regardless of the general radiation safety knowledge of visitors to a plutonium facility, they should be escorted at all times when they go into the posted areas of the plant. In addition, before going into such an area, they should be given a general orientation to the facility radiation protection program and informed about the potential radiation conditions in the areas where they will be going. They also should be provided with the same protective devices worn by facility personnel engaged in similar activities.

Visitors with a demonstrated need to enter the following areas may be allowed access if such access is controlled with a combination of training and the use of escorts trained for the specific area:

- -- Radiological Buffer Areas
- -- Radiation and High Radiation Areas
- -- Contamination Areas
- -- Radioactive Material Areas

Guidance for training for visitors is provided in the standard, <u>Radiological Control</u>, (DOE, 2017), Article 622:

- -- Persons under 18 years of age should not be permitted to enter Radiation Areas or Contamination Areas without the approval of the Radiological Protection Manager.
- -- Area entry requirements and access restrictions for visitors should be in accordance with established facility procedures.
- -- Individuals, visitors included, shall (10 CFR 835.502(b)) be prevented from entering Very High Radiation Areas when dose rates are in excess of the posting requirements of 10 CFR 835.603(c), and visitors should be prohibited from accessing High Contamination and Airborne Radioactivity Areas.

In addition the following is recommended:

All facility personnel serving as a qualified escort should ensure that each visitor under his/her cognizance completes a facility radiological visitor form. The qualified escort should also sign the visitor form and complete it as appropriate.

Facility-sponsored visitors should provide the following before entering radiological areas, unless these records have already been entered into the facility entry control system:

- -- Evidence of completing required training, as applicable.
- -- visitor radiation exposure disclosure.

The host facility manager should forward the visitor radiation exposure and medical disclosure forms to Dosimetry.

The use of offsite respirator fit test certification may be authorized (if in accordance with the applicable Radiation Protection Program) under the following conditions:

- -- A respirator fit test has been completed within the previous year.
- -- The individual presenting the respirator fit test certification card has not changed physical appearance in a way that would affect the seal of the respirator facepiece to the face.
- -- The facility has the respirator facepieces available that the individual is certified to wear.

3.2.4 Visits by Regulatory Personnel

Periodically, personnel from the DOE and other Federal and state agencies visit radiation facilities for audit purposes or to discuss regulatory changes. In most cases, they will want to look at records of the radiation protection program and, in some cases, will also want to enter posted areas of the facility. They should have ready access to the facility provided that dosimetry and other requirements are met. They should have complete access to facility personnel knowledgeable in the subjects they wish to discuss.

3.2.5 Onsite Packaging and Transportation

DOE Order 460.1D (DOE, 2016d) establishes safety requirements for the proper packaging and transportation of DOE, including NNSA, offsite shipments and onsite transfers of radioactive and other hazardous materials and for modal transportation. DOE M 441.1-1, <u>Nuclear Material Packaging Manual</u> (DOE, 2008e) and DOE-STD-3013-2018, <u>Stabilization, Packaging, and Storage of Plutonium-Bearing Materials (DOE, 2018b)</u>, also provide information on packaging plutonium material

3.3 RADIOLOGICAL CONTROL ORGANIZATIONS

The radiological control organization shall be structured so that all of the activities required to provide support to line management and workers can be accomplished.

3.3.1 Management Commitment

Management commitment to safety is the most important characteristic of an effective safety program, including a radiological control program. If the management commitment to safety is strong, the radiological control program will be valued and respected. The radiological control program should be provided adequate authority to permit performance of necessary assignments and program implementation. Management commitment to the ALARA concept is particularly important [see Article 111, <u>Radiological Control (DOE, 2017)</u>]. Adequate personnel, equipment, and funding should be available as a part of this commitment.

3.3.2 Radiological Control Organization Independence and Reporting Level

The radiological control organization should be independent of the line organization responsible for production, operation, or research activities and should have an equivalent reporting level. Because health physics personnel should have the authority to balance operations with safety, they should not report directly to the administrators of operations. When shift work is involved, the operations shift supervisor may make minor health physics decisions in support of the shift's Radiological Control Technicians (RCTs); however, decisions involving basic policies and procedures should be directed to a separate health physics organization.

If a safety organization includes the health physics program, it should be high enough in the company to assure direct access to the company president or equivalent. If the health physics program is administered by a separate radiological control organization, that organization should also be in a position to assure direct access to the company president. This is to safeguard the program from the pressures of production that exist in the operational environment, by keeping it independent of operating organizations.

A system of guides, policies, and procedures should be established to clearly identify the interrelationships, responsibilities, and authorities of those involved with the development, operation, and maintenance of the facility and the health and safety of the employees. These guides, policies, and procedures should be documented and should be reviewed at least once every year.

3.3.3 Adequacy of Personnel and Equipment

A sufficient number of qualified and, where required, certified radiological control personnel should be available to perform necessary tasks for support of plutonium facility startup and operation (See Section 3.4 for guidance concerning staffing and staff qualifications). Sufficient equipment, including protective clothing, respiratory protective equipment, and radiation detection instrumentation should be

available to support RCTs and operating personnel in the performance of work in controlled areas.

3.3.4 Assignment of ALARA Responsibility and Authority

Limiting radiation exposures to the lowest levels commensurate with the benefit of the work to be accomplished has long been a part of health physics and radiological protection programs of DOE and its contractors. 10 CFR 835 (DOE, 2011) establishes the policy of maintaining ALARA exposures of workers and the public to radiation from DOE operations. Procedures are required to be prepared (10 CFR 835.104) and implemented and records shall be maintained as required by 10 CFR 835.701 to demonstrate the implementation of ALARA. The DOE standard, <u>Radiological Control (DOE, 2017)</u>, provides additional guidance. Munson et al. (1988) and Chapter 4 of Implementation Guide G 441.1-1C, Ch. 1 (DOE, 2011a), may be used in developing an ALARA program.

An ALARA committee should be established at the plutonium facility. The membership should include managers and workers from the line, the technical support organization, and the radiological control organization. A line manager, such as Director of Operations, Research, Training, or Maintenance should serve as the committee chair. The ALARA committee should make recommendations to management to improve progress toward minimizing radiation exposure and radiological releases (DOE, 2017).

3.4 STAFFING AND STAFF QUALIFICATIONS

A cadre of operating and maintenance personnel that has experience in the operation of a plutonium facility should be established during the construction of a new facility. The remainder of the operating and maintenance staff should be hired as soon as possible and should receive formal and informal training from the experienced personnel. This step is extremely important to enable all personnel to grow with the facility and learn the details of the operations. Once operations start, potential problems already should have been identified and engineering or administrative changes should have been made to resolve them.

Staffing in the radiological control organization requires technicians and professionals in many support areas. A successful health physics program is highly dependent on the availability of adequate staff support in areas such as environmental monitoring, instrument maintenance and calibration, internal and external dosimetry, meteorology, safety analysis, and risk management.

3.4.1 Professional Staffing and Qualifications

The senior staff of the radiological control organization should include health physicists and other professionals with four-year degrees in science or engineering. A continuing training program should be established for facility personnel. Pursuit of certification by the American Board of Health Physics for senior and professional staff members is encouraged (DOE, 2017).

At least one professional staff member at the plutonium facility should have a minimum of three years of health physics experience in the operation of plutonium facilities.

3.4.2 Technician Staffing and Qualifications

Recommendations for minimum entry-level requirements for RCTs are given in the DOE standard, <u>Radiological Control</u> (DOE, 2017), and DOE STD-1122-99, <u>Radiological Control Technician Training</u>, (DOE, 2009a). They include a high school education or equivalency and knowledge of certain scientific fundamentals. If a two-year degree in nuclear technology or an equivalent discipline is locally available, completion of such a program should be encouraged.

Where possible, RCTs and other members of the health physics staff should have a minimum of one year's experience working at a plutonium facility. Such experience is an important prerequisite to allowing them to work unsupervised. Personnel hired without such experience should work an internship of six months under the leadership of a qualified RCT or supervisor with experience in that facility.

The RCTs should be encouraged to pursue registration by the National Registry of Radiation Protection Technologists.

3.4.3 Staffing Levels

At least one professional health physicist is recommended to be on the staff of each major plutonium facility as a full-time employee.

There is no rule of thumb for determining the number of RCTs needed for a given plutonium facility. The number of RCTs should be based on an analysis that provides for sufficient coverage on each shift, given the number of samples, surveys, and other work to be performed; the time of training, donning and doffing protective clothing; shift turnover procedures; and other similar considerations. The site collective dose and individual dose limits in the facility may also lead to the need for additional personnel. Consideration should be given to having sufficient personnel to respond to off-normal conditions and emergencies as well as routine work. Major maintenance, modifications, or decommissioning activities may require additional personnel.

3.5 INSTRUMENTATION CONSIDERATIONS

The radiation from the radioactive decay of plutonium includes alpha, beta, gamma, X-ray (photons), and neutron radiation. An effective monitoring program for plutonium requires radiation detection instruments that are responsive to all of these forms of radiation. It is essential that instruments meet the performance criteria outlined in the applicable U.S. and international standards and be properly calibrated for their intended use.

3.5.1 Types of Instruments and Measurements

Alpha-sensitive instruments are necessary for most contamination control surveys. Exposure rate surveys are normally conducted with photon-sensitive instruments with known energy responses. Neutron surveys become important when processing tens of grams of 238 Pu or hundreds of grams of mixed isotopes of plutonium, particularly compounds (i.e., PuO₂, PuF₄, etc.). The neutron survey is important in instances where photon shields, such as leaded glass, are used; such shields normally stop all of the charged particles, most of the low-energy photons, and essentially none of the neutrons. Under these circumstances, neutron radiation is likely to be the major contributor to whole body dose. See the 10 CFR 835 Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a) for a discussion of acceptable approaches for evaluating neutron radiation levels using the radiation weighting factors from the 2007 amendment to 10 CFR 835 (DOE, 2011).

Continuous air monitors (CAMs) are used extensively in plutonium facilities. Continuous air monitors and sample extraction lines that go to CAMs and continuous radiation dose monitors should be placed outside the glove boxes and hoods. In-line processing instrumentation is critical to accurately monitor the work stations and a review should be performed to determine instrument locations. Continuous air monitors may not have adequate detection capabilities for real-time monitoring at the DAC level. For ²³⁹Pu, the annual limit on intake (ALI) is 12 nCi for absorption type M compounds based on the DAC of 5 x 10⁻¹² µCi/mL, as given in Appendix A to 10 CFR Part 835 (DOE, 2011). DOE G 441.1-1C, Ch. 1 recommends that real-time air monitors be capable of measuring 1 DAC when averaged over 8 hours (8 DAC-hours) under laboratory conditions. Alarm set points for real-time air monitors used for routine monitoring should be set at the lowest practical level so as to accurately indicate loss of containment or the need for corrective action without causing a significant number of false alarms. When monitoring for alpha emitters in areas with high radon concentrations an alarm set point greater than 8 DAC-hours may be necessary.

Continuous air monitors (CAMs) typically have had poor large-particle response due to particle loss during transport to the filter inside the system. Newer alpha air monitors are able to handle large particles more efficiently. Background levels of radon-thoron decay products may be present in concentrations up to 50 to 100 times greater than the level of plutonium of interest. If calibrated properly, alpha CAMs will subtract background levels of radon-thoron decay products; however, in practice the detection limit for plutonium may be as high as 40 DAC-h in the presence of high radon levels. A new generation of alpha CAMs is able to compensate for radon more effectively and meet the desired 8 DAC-h alarm level.

Transuranic aerosol measurement units have been developed and adapted to be used in the workplace. These units avoid preferential plate-out of larger particles by using an in-line filter. Higher flow rates than those normally used with CAMs may be used. Increased detection is obtained on a quasi-real-time basis by high-volume air sampling and counting in a separate vacuum chamber. Detection levels of less than 0.5 DAC-h have been quoted for these units. It has been demonstrated that high-volume impact samplers used at some facilities have demonstrated detection capabilities of 0.1 DAC-h in the laboratory and 1 DAC-h in the field. Other monitoring systems that use diffusion, impaction, or electronic discrimination to

reduce the effect of background resulting in an increased detection capability have also been used and are being improved upon. However, it is suggested that sitespecific testing be performed on any new equipment to ensure compatibility and verify expected performance. See the <u>Health Physics Manual of Good Practices for</u> <u>the Prompt Detection of Airborne Plutonium in the Workplace</u> (Mishima et al., 1988) for additional information on the selection, placement, and operation of plutonium air monitors.

3.5.2 General Performance Criteria for Instruments

Programs for in-plant monitoring of plutonium consist mainly of airborne and surface contamination surveys and dose rate surveys. The general and specific performance criteria for the instrumentation needed to conduct these programs are described in ANSI N317-1991 (ANSI, 1980a). Performance specifications are also given in ANSI N323 (ANSI, 1997b), ANSI N42.17A (ANSI, 2003), and ANSI N42.17C-1989 (ANSI, 1987b) for portable health physics instrumentation and IEC Publication 60325:2002 (IEC, 2002) for alpha and beta contamination meters and monitors. Criteria for air monitoring instrumentation are contained in ANSI N13.1 (ANSI, 2011a), IEC Publication 761-2 and draft IEC Publication 761-6 (IEC, 1983), and ANSI N42.17B-1989 (ANSI, 1987a). Criticality alarm systems are discussed in ANSI/ANS 8.3-1986 (ANSI, 1997c). The criteria discussed in the following subsections are specified in these standards as referenced.

3.5.2.1 Portable Survey Instruments

ANSI N317 (ANSI, 1980a) discusses several criteria related to the performance of portable survey instruments; these include the following requirements:

- -- The overall accuracy shall be within $\pm 20\%$, and the precision shall be within $\pm 10\%$ at the 95% confidence level.
- -- The response time (i.e., the time for the instrument reading to go from zero to 90% of full scale) shall be < 10 seconds on the most sensitive scale and < 2 seconds at readings of 100 mrem/h, 100 mR/h, and 500 dpm or greater. (This criterion is unrealistic with current neutron instrument capabilities. Response time is typically 30 to 60 seconds.)
- -- The instrument shall be able to maintain accuracy and precision for a minimum of 24 hours of continuous operation.
- -- The instrument shall have a minimum battery lifetime of 200 hours of continuous operation.

ANSI N42.17A (ANSI, 1988a) specifications differ slightly.

-- The response of the instrument shall not change by more than ±15% from a reference value taken at 20°C over the anticipated temperature range for operation.

-- The instrument system shall function within specifications over all anticipated combinations of temperature and humidity (e.g., 15° to 65°C, 40% to 95% relative humidity).

Photon survey instruments should meet the accuracy requirements stated in ANSI N317 (ANSI, 1980a) over the energy range of 0.01 to 1.25 MeV. The angular response of this type of instrument should be within $\pm 15\%$ over a 2 π steradian frontal direction using at least two photon sources with energies ranging from 0.06 to 1.25 MeV. Experience has shown that this response specification is not met by most instruments at lower energies due to attenuation of the photon. The energy dependence should be within $\pm 15\%$ over the range of very low energy to 1.25 MeV and the operating range should be from 0.5 mR/h to at least 5000 mR/h.

Experience has shown that $\pm 20\%$ over very low energy to 1.25 MeV is more realistic. This specification applies to a specific window selection (e.g., below 0.05 MeV, the electron equilibrium cap or beta shield shall be removed).

ANSI N42.17A (ANSI, 2003) has a broader scope than ANSI N317 (ANSI, 1980a) but the criteria in it apply to portable survey instruments. Additional criteria include geotropism (maximum change of 6% from reference reading for all orientations), temperature shock (a critical parameter for instruments stored inside, but used outdoors during very cold or hot periods), mechanical shock, vibration, and ambient pressure (maximum change of 15% from reference reading for the latter four criteria). Some differences exist between ANSI N42.17A and ANSI N317. In most cases, the criteria for ANSI N42.17A are more applicable because these criteria are based on substantial testing, which was sponsored by DOE. In ANSI N42.17A, precision is tied into a measurement level; for example, it quotes a precision of 15% at <500 cpm and 10% at >500 cpm. Also, with the advent of liquid crystal displays and other digital readouts, "response time" is defined as the time it takes for the reading to move from 10% to 90% of the equilibrium or steady-state reading. Another significant difference in the standard is that the battery lifetime specification is 100 hours instead of the 200 hours mentioned in ANSI N317.

For direct alpha contamination surveys, the use of audible signals (headphones or speaker) greatly facilitates the detection of "hot spots."

IEC Publication 325 (IEC, 1981) provides additional guidance on the uniformity of probe response for alpha and beta contamination meters. Surface sensitivity measurements are also discussed in this standard.

3.5.2.2 Performance Criteria for Fixed Monitoring Instruments

Airborne contamination monitors, surface contamination monitors, photon and neutron area monitors, and emergency instrumentation are

fixed monitoring instruments subject to the following standard performance criteria.

Airborne Contamination Monitors. Airborne contamination monitors, normally CAMS (see Section 3.5.1), should meet the following criteria according to ANSI N317 (ANSI, 1980a). The primary purpose of any CAM is to detect the presence of airborne radioactivity and activate an alarm to warn personnel in the area so that actions can be taken to minimize personnel exposures. The goal for any CAM should be to perform this function as quickly as possible and at the lowest detectable level of radioactive airborne concentration. The quantity of airborne radioactivity that will result in an alarm within a given time interval is defined in units of DAC-h for a particular radionuclide and is a function of the nuclide's airborne concentration in DACs, the sampling rate, the lower limit of detection of the instrument, and the time needed for the alarm to occur. Mishima et al. (1988) provides guidance on each of these functions.

The minimum detection level of ²³⁹Pu, in terms of derived air concentration (DAC), should be 8 DAC-h at the point of sampling in the presence of nominal amounts of naturally occurring alpha-emitters such as radon and thoron and their decay products. (No guidance is provided on what a "nominal" amount is, however.) The operating range should be at least 100 minimum detection levels (i.e., up to 800 DAC-h for ²³⁹Pu). Instrument error should not exceed $\pm 20\%$ of the reading over the upper 80% of the operating range. The reproducibility of the system for any given measurement should be within $\pm 10\%$ at the 95% confidence level for a mid-scale or mid-decade reading. The instrument should be capable of operating with less than a 5% change in calibration over the ambient temperature range expected. The instrument should be equipped with an adjustable alarm set point (audible and visible alarms) that can be set at any point over the stated range. The air flow rate should be indicated and adjustable. Voltage and frequency variations of $\pm 15\%$ within design values should result in reading variations of no greater than 5% at the minimum detection level.

ANSI N42.17B (ANSI, 1987a) provides additional performance criteria for air monitors used to detect plutonium. This standard provides specifications for general criteria (sampler design, units of readout, alarm threshold, etc.), electronic criteria (alarms, stability, response time, coefficient of variation, and line noise susceptibility), radiation response, interfering responses (radiofrequency, microwave, electrostatic, and magnetic fields), environmental criteria (temperature, humidity, and pressure), and air-circuit criteria. More detailed specifications are provided in ANSI N42.17B than in ANSI N317 (ANSI, 1980a); however, the environmental criteria and the limits of variation are not as restrictive as those in ANSI N317. With respect to accuracy, ANSI N317 requires less than $\pm 20\%$, and ANSI N42.17B requires 40% at the 95% confidence level. For the environmental criteria, ANSI N317 requires that the readings change less than 5% under ambient conditions, while ANSI N42.17B gives a 15% limit of variation. As discussed previously,

criteria from ANSI N42.17B are more applicable because they are supported by instrument testing.

ANSI N13.1 (ANSI, 2011a) provides detailed guidance on sampling methods. One criterion that relates to CAMs is that air sample lines between air inlet and filter media are to be eliminated where possible; where not possible, they are to be designed to meet the sampling criteria contained in the standard (e.g., short lines, proper sampling rate, smooth bends). The use of Tygon tubing as sample lines before the collection filter should be minimized or eliminated. Air in-leakage from surrounding areas can be a problem when using sampling lines. Testing for air in-leakage shall be performed at least annually or when seals or "O" rings are replaced.

Surface Contamination Monitors. Surface contamination monitors include hand and/or shoe counters and instruments (or probes) with sufficient flexibility to survey pieces of equipment, including exterior clothing. ANSI N317 (ANSI, 1980a) states that these instruments shall have an audible alarm, a frequency that is proportional to the count rate, or a preselectable trip setting, and that upon reaching that level shall activate an audible or visible alarm or both. These instruments should be calibrated according to the requirements in ANSI N323 (ANSI, 1997b) and be equipped with a traceable check source. Fixed instruments should be powered by alternating current (AC) and provided with an emergency power source.

Photon and Neutron Area Monitors. Photon and neutron area monitors measure the intensity of photon and neutron radiation in areas where significant quantities of plutonium are stored and/or handled. ANSI N317 (ANSI, 1980a) states that these monitors shall have a preselectable trip setting with audible annunciators, shall provide electronic signals for remote alarms if they are used as alarming devices, and shall be equipped with a visual meter or digital readout. All neutron and photon area monitors should be AC-powered and all critical monitors should be provided with an emergency power source. Many of the requirements that apply to portable survey instruments, as stated in ANSI N317 may also apply to this type of instrumentation. Calibrations should be performed according to the requirements in ANSI N323 (ANSI, 1997b).

3.5.2.3 Performance Criteria for Emergency Instrumentation

Meeting the criteria for criticality accident alarm systems, fixed nuclear accident dosimeters, and other emergency instrumentation is essential.

Criticality Alarm Systems. ANSI/ANS 8.3 (ANSI, 1997c) discusses the performance and design criteria for criticality accident alarm systems. The criteria include the following:

-- Criticality alarm systems shall be designed to detect immediately the minimum accident of concern; the minimum accident may be assumed to deliver the equivalent of an absorbed dose in free air of

20 rad at a distance of 2 meters from the reacting material within 60 seconds.

-- Systems shall be designed so that instrument response and alarm latching shall occur as a result of radiation transients of 1millisecond duration. The alarm signal shall be for evacuation purposes only and of sufficient volume and coverage to be heard in all areas that are to be evacuated. Very high audio background noise in some areas may require that the alarm be supplemented with visual signals; however, high background noise is a dangerous situation that should be prevented by design. Instrument response to radiation shall be calibrated periodically to confirm the continuing performance of the instrument. The calibration interval may be determined on the basis of experience but shall be no less frequent than annually. Tests should be performed at least monthly and the results of testing should be documented.

The standard does not quantify criteria for reliability or the rejection of false alarms. Consideration should be given to the avoidance of false alarms as accomplished by providing reliable single detector channels or by requiring concurrent response of two or more detectors to initiate the alarm. (ANSI 1986a).

Fixed Nuclear Accident Dosimeters. All DOE facilities that have sufficient quantities and kinds of fissile material to potentially constitute a critical mass, such that the excessive exposure of personnel to radiation from a nuclear accident is possible, shall provide nuclear accident dosimetry for those personnel (10 CFR 835.1304). Requirements for fixed nuclear accident dosimeters are found in DOE Order 420.1C (DOE, 2012b).

Effluent Monitors. Facilities that deal with unencapsulated plutonium should have continuously operating effluent monitors to determine whether or not plutonium is being released to the environment. Effluent monitor criteria is found in IEC Publications 761-1 and 761-6 (IEC, 1983) and ANSI N42.18 (ANSI, 2004) and should be performed. Similar to airborne contamination monitors, effluent monitors should be tested for air in-leakage at least annually or when seals or "O" rings are replaced.

Other Emergency Instrumentation. Other emergency instrumentation should provide ranges for all radiation dose rates and contamination levels potentially encountered at the time of an accident. Normally, dose rate capabilities from a few millirem per hour to a few hundred rem per hour should be required. Performance specifications for emergency radiological monitoring instrumentation can be found in ANSI N320-1979 (ANSI, 1975) and BNWL-1742 (Andersen et al., 1974).

3.5.3 Instrument Calibrations and Testing

Radiation doses and energies in the work areas should be well characterized. Calibration of instruments should be conducted where possible under conditions and with radiation energies similar to those encountered at the work stations. Knowledge of the work area radiation spectra and instrument energy response should permit the application of correction factors when it is not possible to calibrate with a source that has the same energy spectrum. All calibration sources should be traceable to recognized national standards, such as NIST. Neutron energy spectral information is considered particularly important because neutron instruments and dosimetry are highly energy-dependent.

When the work areas have been well characterized, the calibration facility used by the plutonium plant should be set up to represent as closely as possible the work area's radiation fields. Californium-252 or PuBe calibration sources should be used for work areas that process plutonium metal and plutonium oxide because their neutron energy distribution is similar to those compounds. Facilities that process PuF_4 should use a PuF_4 source. Most work areas at processing plants are high-scatter areas and thus have significant quantities of low-energy neutrons. Because it may not be feasible to have sources and scatter geometries representative of all work locations at the facility, it should be important to determine specific spectra and correction factors for work locations to correct for the calibration. Scatter conditions should be taken into account when setting up a calibration facility. The effect of room scatter in a neutron calibration facility can be significant and may account for as much as 20% of the measured dose rate. Modeling, such as a Monte Carlo N-Particle (MCNP) code, should be used to correct for room scatter.

ANSI N323 (ANSI, 1997b) provides requirements on the calibration of portable instruments and periodic performance testing (e.g. source response checking) of instruments. Section 9.4 of G 441.1-1C, Ch. 1(DOE, 2011a) has additional guidance on this topic.

The reproducibility of the instrument readings should be known prior to making calibration adjustments. This is particularly important if the instrument has failed to pass a periodic performance test (i.e., the instrument response varies by more than $\pm 20\%$ from a set of reference readings using a check source) or if the instrument has been repaired. The effect of energy dependence, temperature, humidity, ambient pressure, and source-to-detector geometry should be known when performing the primary calibration. Primary calibration should be performed at least annually.

Standards referenced in Section 3.5.2 discuss specific performance testing of radiation detection instruments. Testing procedures in these standards should be used for periodic requalification of instruments or detailed testing of instruments.

The calibration of photon monitoring instruments over the energy range from a few keV to 300 keV is best accomplished with an x-ray machine and appropriate filters that provide known x-ray spectra from a few kiloelectron volts to approximately 300 keV. Radionuclide sources should be used for higher energies. Most ion chambers used to measure photon radiations have a relatively flat energy response

above 80 to 100 keV; ¹³⁷Cs or ⁶⁰Co are typically used to calibrate these instruments. These sources also may be used to calibrate Geiger-Mueller (GM) type detectors used for dose rate measurements. It should be noted that some GM detectors (e.g., those with no energy compensation) can show a large energy dependence, especially below approximately 200 keV. GM detectors should not be used if not energy compensated.

The calibration of alpha-detection instruments normally should be performed with ²³⁹Pu, ²⁴¹Am, or ²³⁰Th sources. Several sources of different activities should be used to calibrate different ranges.

Whenever possible, beta detectors should be calibrated to the beta energies of interest in the workplace. A natural or depleted uranium slab source can be used for calibration of beta detectors when beta radiations in the workplace have energies similar to the uranium. The energy dependence of beta detectors can be tested using the calibration sources listed in the International Organization for Standardization (ISO) Publication 1980 (1984); such as ⁹⁰Sr.

The calibration and testing of crucial monitoring systems are extremely important to the overall radiation protection program but have often been neglected. Effluent monitoring and sampling systems and remote area monitoring systems should be given several tests. The radiological, environmental, and mechanical characteristics of the instrumentation portion of the system should be fully evaluated prior to its first use to ensure its compatibility with performance requirements and facility operating conditions. The effluent sampling losses from the sample probe to the collector/detector should be determined. This test should be repeated at least annually and when a significant change in the sampling equipment is made. The sample probe should be examined at least once a year to verify that its design or performance has not been changed by corrosion. The recorder of the sample flow rate should be calibrated when it is installed and annually thereafter. The operability of the overall system should be completely tested once, with repeat tests only after modification, repair, or maintenance. Operability checks should be scheduled at least monthly and calibration performed at least annually.

The operation of criticality or other radiation alarm signal systems should be checked periodically to ensure that the alarms are audible at all potentially occupied locations. To prevent any desensitizing of staff, the staff should be aware that the tests will be performed, and where possible, tests should be scheduled during off-shift hours. Building systems should be tested semiannually and the area-wide system should be tested at least annually. Any portion of the detector/alarm system that is affected by the test should be reconfirmed for operability after the test is completed (e.g., if a detector is disconnected and a signal is injected at that point, the detector should be tested immediately after it has been reconnected).

3.6 RADIATION SAFETY TRAINING

A thorough radiation protection training program should be established at plutonium facilities. Separate training programs should be established for general employees, radiation workers, and RCTs. The training of all staff members should be carefully documented. The DOE standard, <u>Radiological Control</u> (DOE, 2017), and DOE standardized training programs (DOE, 2007b and DOE, 2008) provide guidance on information to be presented during the training programs.

The frequency requirements for Radiation Safety Training are specified in 10 CFR 835.901. Refresher training in the alternate year when retraining is not performed is recommended. Individuals who work with plutonium should have special plutonium facilities training, such as DOE HDBK-1145-2008 <u>Radiological Safety Training for Plutonium Facilities</u> (DOE, 2008c) in addition to <u>Radiological Worker Training</u>, (DOE, 2008).

Training requirements shall ensure that personnel have the training to work safely in and around radiological areas and to maintain their individual radiation exposure and the radiation exposures of others ALARA.

3.6.1 Radiological Worker Training

Before working in plutonium operations, all radiological workers shall be trained and qualified according to 10 CFR 835.901. A thorough radiation protection training program should be established at plutonium facilities. Before beginning plutonium training, each plutonium worker should receive Radiological Worker Training and other radiation safety topics as required by 10 CFR 835.901(a).

The level of radiation worker training should be determined in accordance with the standard, <u>Radiological Control</u>, Table 6.1 (DOE, 2017). All training should be in accordance with <u>Radiological Worker Training</u>, (DOE, 2008) and implemented by the guidance of Chapter 14 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a). All training dispositions and records shall be documented in accordance with 10 CFR 835.704 (DOE, 2011).

3.6.2 Radiological Control Technician Training

A thorough RCT training program should be established at plutonium facilities. Before plutonium operations begin, a trained and qualified staff of RCTs should be present. All RCT training should be accomplished in accordance with DOE HDBK-1122-99 (DOE, 2009a)

3.6.3 Training for Other Facility Personnel

Nonradiological workers in a plutonium facility should be given a general orientation on the radiation safety concerns for working with plutonium, the general protective measures used for work with plutonium, and the engineered safety features of the facility.

3.6.4 General Public Education

If there are members of the public who live or work near a plutonium facility, a plan for orientation of members of the public should be developed to inform them of facility activities. Such a plan should include information on the concerns that require protection of people from potential injuries by plutonium, the general protective measures used at the facility to confine it and keep it out of the public domain, and solicitation of information on the concerns of members of the local public about plutonium. To the extent possible, efforts should be made to allay those concerns. The information in the public education plan should also be provided to local news media.

3.6.5 Training Qualifications

All training instructors and materials should meet the requirements in DOE Order 426.2, (DOE, 2010a) and should meet the guidance in the standard, <u>Radiological</u> <u>Control</u> (DOE, 2017).

Each plutonium facility should develop performance-based training that reflects radiological conditions present at the facility. This training should be monitored to ensure that site specific, worker-performance-based measures, and practical factors are included in the plutonium training.

3.6.6 Health Physicist Training Involvement

Facility health physicists should have comprehensive knowledge of all of the material on plutonium radiation safety that is included in the training programs for radiation workers and RCTs.

3.7 RADIOLOGICAL RECORDS

The systematic generation and retention of records relating to the occupational radiation protection program are essential to describe the occupational radiation exposure received by workers and the conditions under which the exposures occurred. Such records have potential value for medical, epidemiological, and legal purposes.

Regulation 10 C FR 835 (DOE, 2011) establishes radiation protection program records requirements. The standard, <u>Radiological Control</u> (DOE, 2017), provides guidance for radiation protection program records.

10 CFR 835 Subpart H requires that records be maintained that document compliance with 10 CFR 835. Subpart H requires specific information on the following types of records:

- -- Individual monitoring records
- -- Monitoring and workplace records
- -- Administrative Records

Most of the required radiological records have established retention periods. The retention periods are discussed in DOE Order 200.1A (DOE, 2008b). Individual records may be covered by the Privacy Act; the DOE has codified the Privacy Act in 10 CFR 1008, <u>Records Maintained on Individuals</u> (Privacy Act) (DOE, 2011i).

Detailed guidance on development and maintenance of a radiological exposure recordkeeping and reporting system can be found in Chapter 13 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a).

3.8 ALARA AND OPTIMIZATION

The policy of maintaining radiation exposures ALARA has existed in principle since the early 1940s. The evolution of ALARA into a formal program began in the early 1960s. It is well to remember that the ALARA approach was applied to radiation protection far earlier and is much more institutionalized than any comparable approach to other hazards.

Although there is, and has been since the 1940s, a series of official established dose limits, they do not represent ALARA. ALARA is a continuous process of controlling and managing radiation exposure to workers, the general public, and the environment. Although ALARA is based upon protection of people and the environment, the philosophy is also grounded on sound economic and operating principles. The responsibility for maintaining radiation exposures ALARA is not a unique responsibility of management or health physics personnel. It is a responsibility of everyone involved in managing, supervising, or performing radiation work. It is imperative to teach administrative personnel to support the principles and practice of ALARA, and to train all radiation workers to consider ALARA as they prepare for and perform their work.

10 CFR 835 Subpart K "Design and Control" contains specific requirements relating to ALARA considerations for facility design and modification. Also, DOE Order 458.1, Ch 3. "Radiation Protection of the Public and Environment" (DOE, 2011c) contains environmental ALARA requirements.

3.8.1 Current Status of ALARA Programs

Currently, it is common practice in a DOE facility to have a well-structured ALARA plan for the entire facility, with more detailed plans in the various buildings or functional subunits of the facility. There is ordinarily a facility coordinator who administers the overall ALARA plan and reports to top-level management of the facility. Coordinators for the various buildings or subunits of the facility receive guidance from the overall facility coordinator and report the results of their ALARA programs to that individual.

3.8.2 Achievement of Goals

The standard, <u>Radiological Control</u> (DOE, 2017), provides guidance to contractors (facility) to provide documentation of the ALARA process. To ensure improving radiological performance, at the beginning of each fiscal year, each facility prepares and submits Radiological Performance Goals. At least quarterly, the contractor (facility) provides the contractor senior site executive with an interim status report of the goals. At the end of the calendar year, an Annual Goal Status Report is issued.

Identifying specific ALARA goals in plutonium facilities requires close coordination between the facility ALARA team members (operations, maintenance, and health physics personnel) made up from a cross-section of personnel

representing the various work elements of the facility. ALARA goals may be formulated as qualitative or quantitative types of goals, but shall be measurable and achievable, with clearly defined endpoints.

3.8.3 Quality Assurance

Important aspects of any ALARA program are the measurement of beneficial effects and the determination that important factors, such as economic impacts, the time involved in accomplishing tasks, and the utilization of personnel, are being optimized. To accomplish these objectives, it is necessary to have a written plan for the ALARA program and high quality records of activities involving exposures to workers, the public, and the environment. These permit comparisons with past experiences and analysis of the recorded activities. In many cases, such studies of the recorded activities not only confirm satisfactory execution of the work, but reveal opportunities for future improvements.

One approach which works very well is the inclusion of an ALARA worksheet along with the RWP. Such a worksheet should be prepared by an individual with responsibilities for the work to be performed, a relatively detailed knowledge of the radiological conditions, and knowledge of what is required to accomplish the task. The worksheet should contain estimates of the time to complete the task and the expected radiation doses that will be received. If any special engineered devices are used to control or reduce personnel exposure, they should be noted on the ALARA worksheet, along with any special instructions that they require. These worksheets provide valuable information for analysis of the effectiveness of the ALARA program for each job.

3.8.4 Technical Aspects

The technical aspects of ALARA programs include not only the standard equipment regularly used in controlling dose to workers, the public, and the environment, such as facility shielding, ventilation filters, installed and portable radiation measuring instruments, but also many special devices that may be used temporarily. Special devices can be used to provide exposure control and/or containment when it may not be practical without them. These include temporary shields, tents or greenhouses, portable fans, ductwork and filters, and special fixtures to hold highly radioactive materials requiring detailed inspections, repairs, modification, or fabrication. Such devices can permit doing difficult work at low radiation doses, which might not be possible otherwise.

Some of these special devices may have general application and can be kept on hand for use as needed. In some cases, devices would have to be especially fabricated for a specific task. Since this would ordinarily have a significant effect on the cost of doing that job, the economic aspects of doing or not doing the job would have to be carefully evaluated.

3.8.5 Attributes of Effective Review and Audit

Evaluation of the effectiveness of an ALARA program requires both reviews and auditing. The reviews will include detailed examination of the written ALARA program plan and the records of ALARA activities. The objectives in such reviews

are to find if the written plan is being followed, and what is working or not working well. Such reviews can be performed adequately by either a knowledgeable member of the facility staff or an equally knowledgeable outsider. The written report of a review should be directed to a member of management who is responsible for implementation of the ALARA program.

Audits are best performed by an outside health physicist who is sufficiently knowledgeable about work with plutonium and its radiological characteristics that he/she knows where to look for problems and can make appropriate evaluations and recommendations. He should not only examine the ALARA program plan and records, but should also visit the working areas and laboratories in the facility, with a knowledgeable escort who can answer questions about activities and conditions in the facility.

There is nothing really unique in ALARA programs at plutonium facilities, compared with facilities handling other kinds of radioactive materials. However, the radioactivity of plutonium, its potential for criticality, and its relatively high radiotoxicity require somewhat more meticulous surveillance and control than many other radionuclides. Therefore, the detail in ALARA programs for plutonium facilities is likely to be somewhat greater than would be found in ALARA programs for many other facilities.

In any plutonium facility, it is highly desirable to have well-structured ALARA teams in each building or subunit of the facility. Facility goals should be developed by the facility ALARA teams. All facility-specific goals should be categorized using the facility-specified format and should include the following:

- -- Exposure Reduction. Goals listed under exposure reduction may reflect occupational or nonoccupational exposure reduction. Exposure to radiological hazards or nonradiological hazards are relevant. Specific jobs for which exposure reduction plans have been developed should be covered in this section. Exposure may be reduced by reducing other hazards that contribute to the difficulty of performing work in radiological areas. For example, reducing noise, reducing heat stress conditions, or improving lighting may facilitate the completion and accuracy of work performed in radiological areas and, thus, reduce exposure. Such opportunities for exposure reduction should be carefully evaluated and appropriate ALARA goals established to make the most of these opportunities.
- -- Source Reduction. Source reduction should concentrate on minimizing or eliminating the sources of radiation exposure. Reducing the number of areas with radiological contamination and reducing dose rate are examples of sourcereduction goals. Where the presence of nonradiological hazardous materials results in mixed waste, the removal of the hazardous material may have ALARA benefits by reducing the waste classification. Such changes may also reduce exposure at a later time by eliminating the need to store or further treat the waste. In these cases, eliminating the hazardous material may be an appropriate source-reduction ALARA goal.

-- Administrative. Administrative goals typically encompass training, program improvements, procedure revision, or other administrative-type activities. Administrative goals are generally qualitative, so it is difficult to develop endpoints for them. Specific efforts shall be made to ensure that adequate closure mechanisms exist for administrative goals.

During all phases of ALARA goal-setting, the facility health physics personnel should be intimately involved in providing advice and expertise on ALARA actions.

When addressing exposure reduction, a cost/benefit analysis should be made to determine the real cost of implementing a dose reduction plan. The Health Physics Manual of Good Practices for Reducing Exposures to Levels that are as Low as Reasonably Achievable (Munson et al., 1988), provides an excellent methodology for conducting a cost/benefit analysis by health physics personnel.

The application of ALARA principles to the performance of work in the field is the main objective of any ALARA program. ALARA design, engineering, planning, and administration come to fruition in maintaining exposures ALARA to workers and the public. The operational application of ALARA requires cooperation and coordination of many functional groups, including radiation protection, operations, maintenance, planning and scheduling, training, engineering, and administration.

The primary responsibility for controlling radiation exposure during operations rests with the individual and his/her immediate supervisor. The support functions provide the training, resources, guidance, and measurements, but it is in the application that the effectiveness of an ALARA program is realized. Operational measures for controlling exposure shall be applied to assure that any work with radioactive materials is carried out in the safest manner reasonable. Both engineered and administrative control measures should be used for limiting exposure.

Engineered controls should be utilized whenever possible. In addition, periodic verification of the continued effectiveness of these controls should be performed by facility health physics personnel. Ventilation and filtration systems should be routinely checked and inspected to assure that operation is maintained within the design criteria. The integrity of shielding, the reliability of equipment, and the calibration of instruments should likewise be routinely verified.

Although administrative controls are not an adequate substitute for engineered features, they are necessary. They are a part of the management systems developed and implemented to provide guidance, direction control, and limitations for activities. Administrative controls include the documents that describe organizational interfaces and prescribe controls for radiation protection. Administrative controls, especially procedures, should be reviewed by those responsible for ALARA to ensure that radiation exposure activities include dose limitation considerations.

Factors that shall always be considered in an ALARA program are the costs and benefits. This is especially important when the identified benefit represents a very

small increment of radiation dose reduction. Funds for dose reduction should always be applied to actions which will achieve the greatest dose reduction for the cost.

The final decontamination and decommissioning (D&D) of a plutonium facility should be given consideration in both the original design of the facility and any modifications done to the facility during its operating lifetime. Likewise, D&D should be given consideration in choosing operating processes and practices for the facility, including any changes in processes and practices during its operating lifetime. Both design and operating activities can affect the radiation levels and personnel doses encountered by workers who perform the D&D activities. To the extent practicable, design and operations should provide for radiation levels that are ALARA during D&D activities.

The successful implementation of an ALARA program requires the commitment, support, attention, and efforts of all members of an organization. In facilities in which the radiation exposures are already relatively low, implementation of the ALARA concept is particularly challenging. The reduction of radiation doses to ALARA levels demonstrates to workers and the public a continued emphasis, commitment, and concern for health and safety.

3.9 CONDUCT OF OPERATIONS

The organization and administration of operations should ensure that a high level of performance in DOE facility operations is achieved through effective implementation and control activities. Administration of operations activities should recognize that protection of the environment, maintaining high-quality safety, and productivity are compatible goals. The DOE policies should describe the standards of excellence under which the facility is expected to operate. Clear lines of responsibility for normal and emergency conditions shall be established. Effective implementation and control of operating activities are achieved primarily by having readily accessible written standards for operations, periodical monitoring and assessment of performance, and personnel accountability for performance. For a more detailed discussion, see DOE Order 422.1, <u>Conduct of Operations (DOE, 2010b)</u>.

A high level of performance in DOE operations is accomplished by management establishing high operating standards and then by communicating the operating standards to workers:

- -- By providing sufficient resources to the operations department
- -- by ensuring that personnel are well trained by closely monitoring performance in operations
- -- by holding workers and their supervisors accountable for their performance in conducting activities.

Senior management establishes operating standards, considering input from workers when appropriate. Working-level personnel will more strongly support the standards when they have had appropriate input into their development. The standards should define operating objectives, establish expected performance levels, and clearly define responsibilities in

plant operations. Standards for operating activities should be integrated into operations department procedures and programs. Operating standards should also be communicated to workers by training them in operating practices and by having supervisors monitor and guide work involving facility operations. Sufficient staff, facilities, equipment, and funding should be allocated to permit the operations department to effectively perform its functions. Performance in operations should be closely monitored by facility management, preferably using operating reports and goals, so that the performance of the operations department can be effectively measured. Operations personnel should be held accountable for their performance through supervisor counseling, performance appraisals, and, when necessary, disciplinary measures. Remedial training should be provided when appropriate.

The health physics organization, as a support element, shall ensure that all aspects of radiation safety are considered in the establishment of operations standards and policy. A well instituted cooperative relationship between operations and health physics is paramount to the health and safety of workers and the public and to protection of the environment.

A plutonium facility should have a written policy on radiation protection, including an ALARA policy. All radiation protection procedures and controls should have recognizable or formal technical bases for limits, methods, and personnel protection standards. Procedures should be adequately documented, updated periodically, and maintained in a centralized historical file. A control system should be established to assure that all copies are accounted for and that all new procedures are included in the historical files. A designated period of time for holding the historical files should be established. DOE Order 200.1A (DOE, 2008b) and ANSI/HPS N13.6 (ANSI, 2010) provide guidance on how long to keep historical files. In addition, radiation protection procedures should have a documented approval system and established intervals for review and/or revision. A tracking system should be developed to ensure that the required reviews and revisions occur.

The radiation protection procedure system should provide for but not be limited to, the following topics: radiation work procedures, posting and labeling, instrument calibration, and provision for audits.

3.9.1 Radiation Work Procedures

Radiation work procedures, including RWPs, survey procedures, ALARA reviews, sample counting, and other task procedures, fall within the requirements for conduct of operations. All sections of DOE Order 422.1 (DOE, 2010b) apply. The guidance and requirements of Section XVI, "Operations Procedures," is especially pertinent to radiation work procedures. Procedures are a key factor affecting radiation protection performance. Appropriate attention should be given to writing, reviewing, approving, and monitoring implementation of radiation protection procedures. There should be documented qualification and training requirements for those who prepare and approve procedures. A formal approval process should be established. Procedure changes and revisions should be subject to the same review and approval process as the initial procedure.

Personnel should be trained in the use of the procedures they will be expected to perform. For RWPs, workers should read the RWP and verify by signature that they have read it, understand its contents, and will comply with its requirements in the conduct of the work. Procedures should be available for personnel use. The

RWPs should be posted at the entrance to the work location. There should be a system in place to assure that posted copies of all work procedures, including RWPs, are current.

3.9.2 Posting and Labeling

The requirements for posting and labeling of working areas because of the presence, or potential presence, of radiation and/or radioactive material are specified in 10 CFR 835, Subpart G (DOE, 2011). Guidance in implementing the regulatory requirements can be found in Chapter 12 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a), and the standard, <u>Radiological Control</u> (DOE, 2017). Conformance of conduct of operations requirements should assure a reasonable degree of uniformity in the posting and the signs used, as well as verifying that operator aids and other posted information do not interfere with necessary radiological posting. It is necessary to formally review posting of radiological areas in the same manner that the posting of operating aids is reviewed, in conformance with DOE Order 422.1 (DOE, 2010b).

3.9.3 Calibration of Instruments

The status of installed and portable radiological instruments should be well known and appropriate to the use. (Calibration of radiological instruments is discussed in Section 3.5.2.)

"Ownership" of installed radiological dose rate and airborne contamination monitoring instrumentation should be well known and the responsibility and authority for calibration, repair, and notification clearly established. Because such information is often used by more than one group, formal notification procedures should be established to cover those times when the instrument is out of service or beyond the required calibration schedule. Configuration control and quality assurance requirements for installed systems should be established commensurate with their safety significance.

For portable instrumentation, conduct of operations requirements are normally built into the routine calibration and survey program. Functional checks are routinely made to verify calibration, instruments are checked to assure that they are within the calibration period, and survey procedures require identification of the instruments used so that if a problem is later found, measurements can be repeated.

3.9.4 Audits

Conduct of operations does not, in itself, contain requirements on auditing. Inspections, audits, reviews, investigations, and self-assessments are part of the checks and balances needed in an operating program. Auditing is one of the many tools that line management has at its disposal to identify problems. Regulation 10 CFR 835.102 requires internal audits of all functional elements of the radiation protection program no less frequently than every 3 years. These audits are to include program content and implementation. Each one of the 18 topics addressed in DOE Order 422.1 (DOE, 2010b) should be subject to both internal selfassessment and external auditing to assure effective implementation of their requirements. Any deficiencies identified should be documented and corrective

actions aggressively pursued and tracked to completion. The self-assessment and audit process should include conducting trend analyses and root cause evaluations of deficiencies and communication of results throughout the organization.

3.9.5 Decommissioning of Weapons and Weapon Facilities

Decommissioning of nuclear weapons and nuclear facilities is subject to the same conduct of operations requirements as operating facilities. In general, some components, once they are separated, can be downgraded in safety significance. Also, facilities undergoing decommissioning will have fewer safety systems. During decommissioning, status control and shift turnover are extremely important considerations and shall be done in accordance with DOE Order 422.1 (DOE, 2010b). Posting and labeling of radiological areas are also an increasing challenge because of the rapidly changing radiological status. In extreme cases, it may be desirable to have workers review or sign the RWP each day to ensure they are aware of the status.

4.0 CONTAMINATION CONTROL

The primary control for contamination in a plutonium plant is the facility design. Contamination is confined primarily by enclosing the process areas and using controlled ventilation systems. The design objective for the confinement system is to essentially prevent or minimize exposure of plant personnel and the public to airborne contamination. To ensure that this objective is met, additional attention should be given to airborne contamination control, surface contamination control, and personnel contamination control. Radiological controls for the workplace should ensure that radionuclides are contained and handled properly and that intakes, if they occur at all, are negligible to the extent achievable with state-of-the-art technology. However, much of the current effort involves decommissioning of no-longer-needed production facilities. The lack of engineered controls or the systematic removal of existing controls during the decommissioning process introduces a completely different set of circumstances that requires special attention for adequate contamination control and worker and public protection.

4.1 AIRBORNE CONTAMINATION CONTROL

To achieve the design objective of preventing (or at least minimizing) internal exposure of plant personnel, airborne contamination shall be confined to process enclosures which have adequate air cleaning systems. Because both equipment and personnel errors can compromise designed protection and because older facilities may already have unconfined plutonium, air monitoring and other contamination control measures are needed. Experience has shown that the most common route for inadvertent plutonium deposition in man is by inhalation even though intakes may also occur by accidental ingestion or by wound contamination. In facilities being decommissioned, the use of temporary containment structures, interim ventilation systems, and administrative controls such as protective clothing and respirators may be required to replace engineered systems.

10 CFR 835.1002 requires that for the control of airborne radioactive material, the design objective shall be, under normal conditions, to avoid releases to the workplace atmosphere and in any situation, to control the inhalation of such material by workers to levels that are ALARA; confinement and ventilation shall normally be used.

Note: The use of ventilation systems may require the approval of Facility Criticality Safety personnel because these systems may concentrate fissionable material.

4.1.1 Internal Versus External Dose Philosophy

The overall goal of radiological protection is to minimize the total dose to the individual. However, because of the difficulties and cost of evaluating internal exposures to plutonium, it is best to avoid all internal exposures during routine operations and anticipated abnormal events by engineered controls and personnel protective equipment. As stated above, this is an extremely challenging goal for those facilities undergoing decontamination/decommissioning activities or facilities/sites in environmental remediation. The conditions encountered in decommissioning and environmental restoration will typically place a heavy reliance on administrative controls.

4.1.2 Purpose of Air Monitoring

Airborne contamination surveys are performed for the following reasons:

- -- Prompt detection of airborne contaminants for worker protection.
- -- Personnel exposure assessment.
- -- Monitoring of trends within the workplace.
- -- Special studies.

Of primary importance is the prompt detection of airborne contaminants. The rapid, early detection of airborne releases requires knowledge of the potential sources and characteristics of the airborne material, the locations of the personnel who are at risk, and the capabilities of the detection devices. Optimally, the samples should be taken between the source and the person to measure the potential airborne radioactivity exposure to the individual. With the numerous sources and mobility of the workers, accurate measurement of potential exposure to the individual using area air monitoring devices under all conditions is difficult, if not impossible, to achieve. To aid in early detection of unanticipated airborne radioactivity (e.g., as a result of an undetected pinhole leak in a containment glove), samples of airborne materials should be taken as close to their points of potential origin as practicable to maximize the probability of detection (airborne concentrations are at a maximum at their points of origin). To aid in monitoring of an individual's exposure to airborne radioactivity where area sampling is not representative of the individual's exposure, the use of lapel air samplers on the individual is recommended. Detailed guidance for the placement of air samplers and monitors, selection of system characteristics and requirements, and maintenance and calibration of the equipment is available in the Health Physics Manual of Good Practices for the Prompt Detection of Airborne Plutonium in the Workplace (Mishima et al., 1988) and Air Sampling in the Workplace (NRC, 1993).

4.1.3 Regulations and Limits

The regulations for control of radiation work are covered in 10 CFR 835 (DOE, 2011). Additional requirements and guidance for implementation is provided in the DOE standard, <u>Radiological Control</u> (DOE, 2017), and the Implementation Guide. While many of the topics included in the Implementation Guide relate to plutonium contamination control, specific guidance on contamination control has not been provided. The limits established for plutonium and other transuranic elements for contamination areas, high contamination areas, and airborne radioactivity areas are given in 10 CFR 835.603 and Appendix D of 10 CFR 835. The Appendix D values are summarized in Table 4.1.

Radionuclide	Removable ^{2,4}	Total (Fixed + Removable) ^{2,3}
U-nat, U-235, U-238, and associated decay products	⁷ 1,000	⁷ 5,000
Transuranics, Ra-226, Ra-228, Th-230, Th-228, Pa-231, Ac-227, I-125, I-129	20	500
Th-nat, Th-232, Sr-90, Ra-223, Ra-224, U-232, I-126, I- 131, I-133	200	1,000
Beta-gamma emitters (nuclides with decay modes other than alpha emission or spontaneous fission) except Sr- 90 and others noted above ⁵	1,000	5,000
Tritium and special tritium compounds ⁶	10,000	See Footnote 6

Table 4.1 Surface Contamination Values

¹The values in this appendix, with the exception noted in footnote 5 below, apply to radioactive contamination deposited on, but not incorporated into the interior or matrix of, the contaminated item. Where surface contamination by both alpha- and beta gamma-emitting nuclides exist, the limits established for alpha- and beta-gamma-emitting nuclides apply independently.

² As used in this table, dpm (disintegrations per minute) means the rate of emission by radioactive material as determined by correcting the counts per minute observed by an appropriate detector for background, efficiency, and geometric factors associated with the instrumentation.

³ The levels may be averaged over one square meter provided the maximum surface activity in any area of 100 cm² is less than three times the value specified. For purposes of averaging, any square meter of surface shall be considered to be above the surface contamination value if: (1) from measurements of a representative number of sections it is determined that the average contamination level exceeds the applicable value; or (2) it is determined that the sum of the activity of all isolated spots or particles in any 100 cm² area exceeds three times the applicable value.

⁴ The amount of removable radioactive material per 100 cm² of surface area should be determined by swiping the area with dry filter or soft absorbent paper, applying moderate pressure, and then assessing the amount of radioactive material on the swipe with an appropriate instrument of known efficiency. (Note - The use of dry material may not be appropriate for tritium.) When removable contamination on objects of surface area less than 100 cm² is determined, the activity per unit area shall be based on the actual area and the entire surface shall be wiped. It is not necessary to use swiping techniques to measure removable contamination levels if direct scan surveys indicate that the total residual surface contamination levels are within the limits for removable contamination.

⁵ This category of radionuclides includes mixed fission products, including the Sr-90 which is present in them. It does not apply to Sr-90 which has been separated from the other fission products or mixtures where the Sr-90 has been enriched.

⁶ Tritium contamination may diffuse into the volume or matrix of materials. Evaluation of surface contamination shall consider the extent to which such contamination may migrate to the surface in order to ensure the surface contamination value provided in this appendix is not exceeded. Once this contamination migrates to the surface, it may be removable, not fixed; therefore, a "Total" value does not apply. In certain cases, a "Total" value of 10,000 dpm/100 cm² may be applicable either to metals of the types from which insoluble special tritium compounds are formed, that have been exposed to tritium, or to bulk materials to which insoluble special tritium compound particles are fixed to a surface.

7 These limits apply only to the alpha emitters within the respective decay series.

Note: This document concerns release to controlled areas only. Requirements for unrestricted release of materials and equipment are found in DOE Order 458.1, Ch 3. (DOE, 2011c). Refer to that document for guidance regarding unrestricted releases.
4.1.4 Uncertainties and Limitations

Because of the large exposure per unit intake associated with plutonium and the difficulties in evaluating certain exposures, it is important to consider the uncertainty in the measurements when designing a plutonium monitoring program. Although the design objective of the facility will likely be no airborne plutonium contamination, the reality will be a measurement that ensures airborne plutonium is below an acceptable lower limit of detection. The sampling and monitoring program will need to be designed not only for prompt detection of airborne contamination, but to assure that samples are representative of the air that the workers are breathing and have a low enough limit of detection that only negligible doses could go undetected. This is especially important because of the technology shortfall for routine bioassay and in vivo analysis in detecting small intakes of plutonium. Air samples are typically considered representative if they are taken using a personally worn lapel air sampler or if the sample head is within 1 foot of the workers head. The need for an effective sampling and monitoring program is even more critical in the rapidly changing environment of decommissioning activities.

Numerous factors enter into any determination of plutonium contamination levels and the risk to workers. Some of these factors are detection efficiency of the measuring instrument, collection efficiency of the smear media or air sample filter, the location of the smear or air sample in relation to the source of contamination, the physical and chemical properties of the contamination, the representativeness of the air sample to the air being breathed by the worker, the engineered controls available, and the protective equipment used. All of these factors shall be considered in the development of a plutonium contamination control program and in evaluating the actions required for personnel protection.

4.1.5 Samples and Instrumentation

For plutonium facilities, both air sampling and air monitoring are essential elements of the radiological control program. Real-time air monitoring using alpha-sensitive CAMs should be used to alert workers to rapid degradation of radiological conditions. The air sampling system with a lower limit of detection shall be adequate to provide continuing assurance that personnel exposures are within limits and ALARA.

The characteristics of a good plutonium CAM include:

- -- A lower limit of detection equal to or better than 8-DAC-h
- -- high reliability with a minimum of spurious alarms
- -- a stable and constant flow air mover
- -- stable and documented detector efficiency with geometry, filter collection efficiency, self-attenuation, etc., considered
- -- methodology for radiation discrimination and natural radioactivity discrimination
- -- system for activating an alarm

- -- shielding for extraneous sources of interference such as radiation, radiofrequency, temperature, and vibration
- -- mechanical and electrical ruggedness
- -- ease of maintenance and calibration.

A plutonium air sampling program typically includes a system of fixed head air samplers to quantify air concentrations in the workplace. The basic characteristics of the sampling equipment remain the same except that there is normally less flexibility in locating the sampling heads but more flexibility in selecting and operating the counting instrumentation. In many instances, installed sampling systems may no longer be operational or may be in the wrong locations. In those instances, portable air sampling systems, either impactor-head type or filter type may be used to provide required worker protection.

4.1.6 Sample Analysis

Plutonium air samples are typically analyzed by alpha counting, alpha spectral analysis, or chemical analysis. The technique used will depend upon the filter media used, the physical and chemical state of the contaminate, the urgency for the data, interfering radionuclides, and other factors. Authoritative guidance in establishing plutonium air sampling counting and analysis methods can be found in NCRP Report No. 58, <u>A Handbook of Radioactivity Measurements Procedures (NCRP, 1985)</u> and in <u>Air Sampling in the Workplace (NRC, 1993)</u>.

4.1.7 Monitoring Strategies and Protocols

The rapid, early detection of airborne releases requires knowledge of the potential sources and characteristics of the airborne material, the locations of the personnel who are at risk, and the capabilities of the detection devices. Optimally, the samples should be taken between the source and the potentially exposed worker (or member of the public) to intercept the airborne materials before they reach the individual. With the numerous sources and mobility of the workers, interception under all conditions is difficult, if not impossible to achieve. Samples of airborne materials should be taken as close to their points of origin as practicable to maximize the probability of their detection (airborne concentrations are at a maximum at their points of origin).

Fixed probes that are positioned to intercept releases from recognized major potential sources should be used along with portable air samplers for planned activities with known potentials for airborne release of contaminants and for temporary storage of contaminated materials in areas of low air flow. If the workplace exhaust system can be shown to provide rapid, essentially quantitative clearance of airborne contamination, fixed probes that sample the exhaust system may be adequate for routine coverage of unplanned activities. If justified by documented studies, other sampling arrangements may be used that provide improved "total" coverage of the workplace environment for the early detection of airborne contamination.

Those responsible for the rapid and reliable detection of airborne plutonium should consider the following workplace characteristics in evaluating monitoring systems and working environments (Mishima et al., 1988):

- -- The airflow patterns and airborne transport of plutonium in the workplace
- -- The location of personnel within the workplace during various processing procedures
- -- The location at which the airborne plutonium sample should be taken to accurately monitor the activity inhaled by workers
- -- The ability of the system to transport an undistorted sample to the collection media or measurement device
- -- The collection and retention efficiency of the collection medium
- -- The efficiency of the measurement device in measuring the plutonium collected and differentiating the plutonium from other materials present
- -- The accuracy and reliability of the system.

Guidance for each area listed above is provided in Mishima et al. (1988).

4.2 SURFACE CONTAMINATION CONTROL

Controlling plutonium surface contamination is essential because it may easily be resuspended in air and/or transferred to other surfaces. The following elements are important for controlling surface contamination: keeping plant surfaces clean; monitoring, reporting, and tracking contamination levels; and establishing appropriate control zones with limits and action levels for those zones.

4.2.1 Plant Surfaces

Good housekeeping practices are essential in keeping plant surfaces clean. Periodic housekeeping should be performed within contaminated areas to minimize the buildup of contamination and contaminated waste. Periodic decontamination both within contaminated glove boxes and in the general work area should be conducted to minimize removable contamination.

In some instances, it may be appropriate to apply fixatives to minimize the movement of plutonium contamination. However, it is generally desirable to attempt decontamination first. If decontaminating is not successful or perhaps, not appropriate for the job scope, a fixative may be appropriate. If a fixative is used, typically a paint, two layers of fixative should be used, with the bottom coat yellow and the top coat a different color. When the yellow begins to show through the top coat, additional fixative should be applied. Also, for areas which have had a fixative applied over plutonium contamination, a routine contamination survey should be conducted to assure that no contamination has become movable over time.

In some cases a strippable coating may be used to allow easy decontamination at the completion of a job. These strippable coatings are sometimes used to decontaminate areas. An aerosol fixative is also available that can be pumped into a room, glovebox, or other work space, that coats all exposed surfaces, including the underside of components. This allows work to proceed without disturbing contamination.

Note: The use of fixatives may require the approval of Facility Criticality Safety personnel because fixatives may concentrate or moderate fissionable material.

Outside areas may also require a fixative to minimize the spread of contamination. Historically, some outside contaminated areas have been covered with asphalt to fix contamination. This is not a desirable material to use because it creates a mixed hazardous waste as well as significantly increasing the volume of contaminated material for disposal. Two substances that currently are used as an interim fixative for outside soil/surface contamination areas are (1) a derivative of pine tar (toll oil), which forms a non-toxic surface fixative that is hard and appears to have a relatively durable surface and (2) a mixture of white glue and water (enduro seal), which is easily sprayed on and sets rapidly to a firm surface. A water to glue ratio of about 25 to 1 appears to perform well in preliminary tests. Both of these fixatives are only interim measures because of eventual degradation from the elements. For more localized areas where a permanent fixative/cover is needed, a sprayable concrete (Shotcrete) is available. A disadvantage of this material is cracking, which defeats the sealing surface. Another material that can be used as a carpeting for outside contamination is a spray-on two-part polymer that provides a flexible, semidurable cover.

The characteristics of the cover can be adjusted to vary water transmission and the color can be changed to inhibit growth under the covering. The major problem for outside use of all of these fixatives is the invasion and actions of biota. Mice, rabbits, other wildlife as well as plant growth tend to burrow under any covering and spread the contamination. While these measures do not permanently solve the problem, they may provide a method of preventing the spread of contamination until a permanent, acceptable solution is determined.

4.2.1.1 Housekeeping

The three housekeeping practices listed below should be followed in a plutonium facility as part of the Conduct of Operations [see DOE Order 422.1 (DOE, 2010b)]:

- -- The inventory of contaminated and potentially contaminated scrap and equipment should be kept to a minimum because all such materials are subject to special monitoring and accountability.
- -- Radioactive contamination should be controlled and the spread of contaminants and the potential for accidents involving contaminants shall be minimized. (In at least one instance, poor housekeeping contributed to a serious criticality accident.) Management at all levels should continuously emphasize the importance of good housekeeping, and operating procedures should be written to ensure good housekeeping practices.

-- Measures shall be taken to maintain radiation exposure in controlled areas as low as is reasonably achievable through engineered and administrative controls (10 CFR 835.1001).

Where possible, materials that are not absolutely necessary to an operation should be kept out of the contaminated or potentially contaminated area. It is very important to minimize the creation of TRU waste. All packaging and unnecessary protective coverings should be removed before materials are introduced into the process area. Likewise, items that are not necessary to the process should be promptly removed, particularly from glove boxes, and not left to accumulate and become safety hazards, potential fire hazards, sources of radioactive (dust) accumulation, or sources of exposure.

Good housekeeping practices inside glove boxes should emphasize fire and explosion control. Only metal or nonflammable plastic containers should be used for the accumulation of scrap and wastes of any kind in the glove boxes and throughout plutonium facilities. Accumulation of combustible materials in glove boxes should be minimized. When explosive, flammable, or volatile liquids are allowed, they should be rigidly controlled and used only in inert gas atmospheres unless a safety analysis review shows it is safe to do otherwise. All residues should be removed immediately at the conclusion of each job or cleaning operation.

Considerable effort has been expended on the development of coated and corrosion-resistant tools. Some efforts have been marginally successful, but in most cases throw-away tools are favored. Electropolishing of contaminated metal tools and equipment has been shown to be a good method of decontamination and allows for their reuse in some cases or disposal as non-contaminated waste. Where possible, all tools with sharp edges or points (e.g., screwdrivers, ice picks, scissors) should be kept out of glove boxes.

Management should constantly demand good housekeeping. Mandatory, routine clean-up periods are becoming more common due to the increasing cost of storing and disposing of contaminated materials. Better housekeeping is required due to real-time, computerized accountability for nuclear materials. It has been demonstrated that kilogram quantities of plutonium oxide dust can accumulate in glove boxes unless they are routinely cleaned. Much of the exposure to workers originates from layers of plutonium oxide dust on the surface of gloves and the internal surfaces of glove boxes. In processes where plutonium oxide powder is handled, the glove boxes should be cleaned weekly to reduce the accumulation of dust layers and to reduce worker exposure. Although difficult to achieve and maintain, good housekeeping is equally essential during decommissioning of plutonium facilities.

4.2.1.2 Vacuuming

The subject of vacuuming within a glove box is somewhat complex. Experience has shown vacuuming to be the most effective and quickest way

to clean a controlled-atmosphere (dry) glove box. It is not particularly effective for high-humidity or wet-process glove boxes, particularly those that involve acids. After acids have been used in a glove box, washing and wiping is the preferred method of cleaning the etched surfaces.

Vacuuming is particularly effective in dry-atmosphere and inerted enclosures where the levels of radioactive dust can quickly increase personnel exposure. In many cases, vacuuming reduces the exposure level more than a wipe down with a damp cloth, and it can be done more quickly and with less waste material generated. Two factors weigh against vacuuming: possible safety hazards from electrical sparks, and the occasional difficulty of operating in inert atmospheres (although the last item need not be of importance). However, in dry glove boxes with dusty operations using high-exposure plutonium, personnel exposure control is a problem and vacuuming is a quick and effective method of keeping the dust and exposure rates under control and should be considered.

Note: The use of vacuum cleaners may require review by Facility Criticality Safety personnel because vacuum cleaners are likely to concentrate fissionable material.

The use of vacuum cleaners for contamination control requires careful consideration and strict controls to assure that the process does not spread contamination. As a minimum, all vacuums used for radioactive material should have high efficiency particulate air (HEPA) filtration on the exhaust. In some instances, the additional precaution of having the exhaust vented into a process ventilation system should be considered.

4.2.2 Reporting and Documenting Contamination Levels

Radiological control programs require the performance of contamination surveys to determine existing conditions in a given location. Maps with sufficient detail to permit identification of original survey locations should be maintained. Records should contain sufficient detail to be meaningful even after the originator is no longer available. Contamination surveys should be recorded on appropriate standard forms and include the following common elements:

- -- Date, time, and purpose of the survey
- -- General and specific location of the survey
- -- Name and signature of the surveyor and analyst
- -- Pertinent information needed to interpret the survey results
- Reference to a specific Radiological Work Permit if the survey is performed to support the permit [see DOE standard, <u>Radiological Control</u> part 751.1 (DOE, 2017)].

Records should be maintained to document changes in monitoring equipment, techniques and procedures [see DOE standard <u>Radiological Control</u>, part 751.2 (DOE, 2017)].

In addition, records of contamination surveys should include, at a minimum, the following information:

- -- Model and serial number of counting equipment
- -- Contamination levels (using appropriate units) and appropriate supporting parameters, including counting efficiency, counting time, correction factors, type of radiation, and whether the contamination was fixed or removable
- -- Location of areas found to contain hot particles or high concentrations of localized contamination
- Follow-up survey results for decontamination processes cross-referenced to the original survey (see DOE standard, <u>Radiological Control</u> part 754 (DOE, 2017)).

Records for the release of material and equipment from radiological areas to controlled areas should describe the property, the date on which the release survey was performed, the identity of the individual who performed the survey, the type and identification number of the survey instrument used, and the results of the survey shall be documented (10 CFR 835.703(c)). Additional details on radiation records can be obtained from Chapter 13 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a).

All skin and personal property contaminations should be documented and evaluated to help improve the contamination control program. Documentation should include the following:

- -- The person's name and work group
- -- The location, amount, and type of skin or personal property contamination
- -- The results of decontamination
- -- A description of circumstances involved in the occurrence, such as radiation work permit number, protective clothing required, and protective clothing actually used.

4.2.3 Characteristics of Plutonium Contamination

There are few characteristics of plutonium contamination that are unique. Plutonium contamination may be in many physical and chemical forms. (See Section 2.0 for the many potential sources of plutonium contamination from combustion products of a plutonium fire to radiolytic products from long-term storage.) One characteristic of plutonium is its ability to migrate with no apparent motive force. Whether from alpha recoil or some other mechanism, plutonium contamination, if not contained or removed, will spread relatively rapidly throughout an area. This is especially true for

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plutonium newly released to the environment. For plutonium contamination in soil, after contact with soil, the plutonium tends to have its migration hindered and thus moves slower.

4.2.4 Monitoring

Radiation workers are often assigned tasks that conceivably could expose them to radioactive material. It is not sufficient to rely exclusively on equipment design to minimize contamination and exposure in the workplace. A radiation protection program shall include both monitoring of the workers (discussed in Section 4.3) and monitoring of the conditions in the workplace (10 CFR 835 Subpart E). Both functions are essential to a good radiation monitoring program.

Continuous radiation monitoring should be provided during the periods of high or unusual risk associated with the work in the area. Periods of high or unusual risk include the potential or actual breaching of the integrity of the glove-box or associated systems, including such maintenance as replacement of panels, glove changes, bag-out operations, replacement of filters, or repair of vacuum systems. Work that involves the use of temporary enclosures (greenhouses or glovebags) may also be provided with continuous coverage by an RCT, if the hazard is sufficient to warrant such measures. For decommissioning, most activities will be new, unique, and have no historical precedent. Consequently, high and unusual risks may become the norm and the use of temporary controls and continuous coverage the routine.

Monitoring of the workplace is an essential element of every routine surveillance program. It can be effectively accomplished using any or all of the techniques that are discussed in this section. The rigor with which all of the various elements of a radiation monitoring program are applied should be tailored to meet the needs of the individual work areas and should depend on the kind and quantity of radioactive material present and its potential for dispersion. Each program should be designed to meet existing needs, but also should be flexible to allow for incorporation of the possible advantages to be provided by the various available monitoring practices. Monitoring practices include, but are not limited, to the following:

- -- Contamination surveys of the workplace
- -- Release surveys
- -- External exposure surveys
- -- Airborne contamination surveys
- -- Routine surveillance by an RCT.

4.2.4.1 Contamination Surveys of the Workplace

The radiation monitoring program should include documented survey procedures, a system for maintaining survey results, and contamination control limits for "fixed" and "removable" contamination. The results of contamination surveys should be reported in activity per area (e.g., dpm/100 cm²) except for large-area swipes and swipes of very small items. This

permits interpretation of the recorded data without requiring knowledge of instrument efficiency or geometry.

All workplaces should be monitored for contamination levels on a regularly scheduled basis. The frequency of such surveys will depend on the potential for dispersion of the radioactive material. As a minimum, all gloves, work surfaces, floors, equipment, etc., within the workplace should be surveyed according to the frequencies listed in the DOE standard, <u>Radiological Control</u> (DOE, 2017).

The change room and other support facilities within the controlled area should be surveyed for contamination daily. Continuous air monitors, survey instruments at step-off pads, and hand and shoe counters should be functionally tested daily or once per shift in support of the weekly and monthly surveys. These frequent surveys are also part of the routine surveillance program and permit immediate follow-up if low-level contamination is detected to minimize the potential for major incidents. Some fixtures and support areas outside the controlled area, such as door knobs and telephones of adjacent offices and the lunchroom, should also be surveyed daily. Other support areas should be surveyed monthly. If routine survey results detect any contamination in a given area, more detailed surveys shall be performed to determine the extent of the contamination. An investigation should be initiated to determine the source of the contamination and the cause.

To preclude the possibility that contaminated waste would be disposed of as ordinary waste, (1) all process and controlled area waste should be considered contaminated, and (2) mechanisms should be established that prevent the mixing of contaminated and noncontaminated waste.

4.2.4.2 Release Surveys

For transuranic radionuclides, the contamination level (fixed and removable) at which surfaces are considered contaminated are listed in Appendix D of 10 CFR 835 (DOE, 2011). That document also specifies the criteria for the release of materials and equipment to Controlled Areas.

This document concerns release to controlled areas only. The detailed requirements for unrestricted release of materials and equipment are found in DOE Order 458.1, Ch 3 (DOE, 2011c). Refer to that document for guidance regarding unrestricted releases.

4.2.4.3 External Exposure Surveys

To delineate the levels involved, measurements of external exposure should be made at the time a program is established at all locations where personnel exposure occurs. Additional photon and neutron measurements should be made at the same frequency as the contamination surveys. The buildup of plutonium contamination in glove boxes and on gloves and equipment may contribute substantially to the external dose rates.

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4.2.4.4 Measurement and Survey Techniques

This section discusses four types of contamination surveys that are typically used in DOE facilities. Surveys for removable contamination include a largearea wipe survey and a technical swipe or smear survey. Surveys for total/fixed contamination include a scan survey and a statistically based survey. These surveys, or a combination of them, are used to survey material for release from radiological control. The appropriate use of each type of survey is discussed.

Surveys for Removable Contamination

Two types of surveys are used for removable contamination: a large-area wipe survey and a technical swipe or smear survey.

A large-area wipe survey is used to detect gross removable contamination. A large-area wipe survey is typically performed using a large floor cloth and a dust mop type handle to wipe large areas. This technique tends to concentrate any low levels of removable contamination that may be present. The surface to be wiped and the wiping material should be industrially clean (i.e., free of debris, grease, etc.) to reduce self-absorption of alpha contamination. The buildup of material (radioactive or otherwise) that would attenuate alpha radiation needs to be considered in establishing the size of large-area wipe surveys. The survey is performed by wiping the surface of the area being surveyed and conducting frequent checks of the cloth using a portable instrument. For detection of alpha-emitting isotopes, a nonabsorbent material should be used. Removable contamination will be accumulated and concentrated on the wipe, increasing the probability of its detection. Checking for contamination is conducted by placing an alpha measurement instrument approximately 0.25 in. (0.6 cm) from the surface of the wipe for 5 seconds, and the count rate observed. If there is no increase above background, then the wipe may be placed in contact with the detector. If radioactivity above background is measured, the material is contaminated. Depending upon the specific circumstances, a series of technical smears may be required to locate and quantify the contamination within the area covered by the large-area wipe. In most instances, if contamination is detected on the large-area wipe, decontamination should be considered.

For transuranic radionuclides, the guideline values for removable contamination may be lower than the detection limit or MDA (minimum detectable activity or amount) of the portable instrument. See G 441.1-1C, Ch. 1(DOE, 2011a) for a discussion on MDA. If this is the case, the surface area of wipe surveys needs to be large enough that the quantity of radioactivity collected on the swipe will be greater than the detection limit of the instrument. Wipe surveys of areas smaller than this minimum surface area require more sophisticated measuring instruments, such as a scaler measurement, and the entire surface of the material should be wiped. The minimum area (A) for using a large-area wipe survey is given by:

$$A = \frac{MDA}{L} \times 100 \text{ cm}^2$$
(4.1)

where L is the removable surface radioactivity value in $dpm/100 \text{ cm}^2$ of the potential contaminant, given in Table 4.1. and MDA is in dpm.

The purpose of a technical smear survey is to locate and quantify removable contamination that is known or suspected to exist. For small items, a technical smear may be used at any time to verify the item's contamination status. A technical smear or swipe survey is performed by wiping a cloth, paper, plastic foam, or fiberglass disk over a 100-cm² area of the surface. The wipe should be taken with a dry medium using moderate pressure. A common field practice is to use two fingers to press the wipe medium against the surface to be wiped. The wipe is then moved along an "S" shaped path that has a nominal length of 8 in. (20 cm) to 10 in. (25 cm).

When the potential contaminant emits alpha radiation, paper or fiberglass filter papers should be used to assure that alpha activity is not attenuated by becoming imbedded in the wipe. To improve the MDA, smears may be taken over areas larger than 100 cm². However, for purposes of demonstrating compliance with contamination limits, 100-cm² smears need to also be taken and evaluated. The size of the area smeared should be limited to prevent buildup of material (radioactive or otherwise) that would attenuate alpha radiation. Appropriate corrections should be made for objects smaller than 100 cm².

If contamination is detected during a scan survey for fixed contamination, a survey for removable contamination should be performed to determine if the contamination is fixed and to quantify any removable contamination. The survey should be performed using a small piece of absorbent material, such as a standard paper smear. This type of survey for removable contamination is often called a technical smear survey. If no contamination above the values for removable contamination in Table 4.1 is detected during the smear survey, the contamination is fixed, and the area should be appropriately marked.

In addition, a technical smear survey may be used routinely to detect removable contamination, especially for contamination surveys of radiological areas.

Scan Survey for Fixed Contamination

A scan survey for fixed contamination requires passing a portable instrument over the surface of the area being surveyed at a fixed, known scan speed and at a specified distance from the surface. Typically, the scan speed is 1 - 2 in./s (2.5 - 5 cm/s) and the maximum distance is 0.25 in. (0.6 cm) for alpha contamination instruments, but this can vary depending on the instrument, probe configuration and background. A scan survey should be used to survey

material that resides in an area controlled for contamination purposes, an area where unsealed radioactive sources are used, or a radiological buffer area surrounding an area controlled for contamination purposes. A scan survey in conjunction with a wipe survey should be used to release from radiological control material with a total surface area less than 5 ft² (0.46 m²). A statistically based survey, which will be discussed later, should be used to release from radiological control material with a surface are greater than 5 ft² (0.46 m²).

During the performance of scan surveys, the audible response of the instrument is faster than the needle deflection (some instruments do not have a needle, e.g., digital read out instruments). Therefore, audible response should be used in conjunction with meter readings. For alpha surveys, the surveyor should pause for 3 to 5 seconds each time an individual pulse is detected in order to allow a longer count time at the location of the detected pulse, until it is determined whether the response indicates random background noise or detected contamination.

Several important factors affecting scan survey detection sensitivity are: instrument detection efficiency, background, size of the effective probe area, and the speed at which scan surveys are performed. For a given instrument, scan speed can be a critical factor as counting time is inversely proportional to scan speed. For instruments with larger detector faces, the scan speed is faster for a given rate of meter movement because a point on the surveyed surface remains beneath the window longer. To ensure that low levels of contamination can be detected, it is necessary that a maximum scan speed be mandated and that this speed be implemented during field measurements. As noted above, a typical scan speed for instruments in current use is 1 - 2 in./s (2.5 - 5 cm/s). However, the scan speed for a specific application should consider the instrument, probe, guideline value, and confidence level desired. The MARSSIM (DOE, 2000) contains guidance for determination of scan rates. It also suggests that an empirical method be used to verify scan rates. The equipment and method used in this determination may be incorporated into training for survey personnel to enhance their survey skills.

Similarly, the Multi-Agency Radiation Survey and Assessment of Materials and Equipment (MARSAME) (DOE, 2009b) document for releasing equipment such as earth movers, trucks and other equipment has been published.

4.2.5 Release Criteria

The release of material from radiological areas shall be performed according to 10 CFR 835.1101. In these areas, material and equipment should be treated as radioactive material and should not be released from radiological areas to controlled areas if either of the following conditions exist:

- -- Measurements of accessible surfaces show that either the total or removable contamination levels exceed the values specified in Table 4.1
- -- Prior use suggests that the contamination levels on the inaccessible surfaces are likely to exceed the values specified in Table 4.1.

Wire rope and electronic gear with cooling fans are examples of equipment that are difficult to survey and require special procedures to be released from contaminated and airborne radioactivity areas. Additional release criteria can be found in Section 4 of the DOE standard, <u>Radiological Control</u> (DOE, 2017).

It may be noted that Appendix D of 10 CFR 835 allows that surface radioactivity values be averaged over 1 m^2 provided that the activity in any 100 cm² is not more than three times the specified value.

The material release methodology has four main components: material evaluation, scan survey for fixed contamination, large-area wipe survey for removable contamination (described above), and statistical survey for fixed contamination. The material evaluation process involves consideration of the previous known uses of the material, as well as typical uses and the environment in which the material was used. Material evaluation places the material into one of two categories: not potentially contaminated.

Non-radioactive material can be released without an instrument survey if its documented history ensures

- -- That it has never been used or stored in an area controlled for contamination purposes (i.e., a Contamination Area, High Contamination Area, or Airborne Radioactivity Area)
- -- That it has never come into contact with unsealed radioactive material
- -- That it has not been stored or used in a Radiological Buffer Area (RBA) surrounding a Contamination Area, High Contamination Area, or Airborne Radioactivity Area.

This material may be considered to be not contaminated and an instrument survey is not necessary according to the DOE standard, <u>Radiological Control</u> (DOE, 2017). A material history release form should be used to document the release of material that is known to be free of contamination by its history of use. If the material history release form cannot be completed, or if the history of the material is unknown, an instrument survey shall be made of the material. Material released from RBAs around Contamination Areas, High Contamination Areas, or Airborne Radioactivity Areas should also be evaluated using an instrument survey. Material cleared for unrestricted release from a radiological area shall be done in accordance with DOE O 458.1.

The material evaluation process should also consider the nuclides to which the material was potentially exposed. If the material was exposed to significant quantities of nuclides that are difficult to detect, including tritium, ¹⁴C, ¹²⁵I, or ¹²⁹I, an appropriate survey methodology should be applied.

4.2.6 Plutonium Contamination Detection

The detection and measurement of plutonium contamination is necessary to ensure control of contamination and compliance with DOE requirements. Typically,

detection of plutonium contamination has been performed using survey instruments that detect the alpha activity. Routinely used health physics instruments (i.e., alpha survey instruments) may not be adequate for some D&D operations. Self-absorption of plutonium alpha particles within the source or in an irregular surface area may require the use of special X-ray and low energy photon instruments (e.g., a NaI detector). The NaI detector should also be used to detect plutonium contamination that has been painted over.

Discussions of methods used to detect plutonium contamination for past D&D operations can be found in publications by Umbarger (1982) and West et al. (1991). Umbarger reported on nondestructive assay techniques (including portable field instrumentation and laboratory-based methods) for sorting waste in low-level (class A) and TRU waste. Portable field instruments included the field FIDLER (i.e., thin NaI detector), phoswich detector (i.e., thin NaI detector coupled with a thicker CsI detector), ZnS alpha scintillation detector, a portable multichannel analyzer, and a hand-held gamma-ray spectrometer gun. The advantage of a phoswich detector over a NaI detector is its lower operating background. Laboratory-based systems include active and passive gamma-ray spectroscopy, passive neutron detection, and pulsed portable neutron generator interrogation.

During the decommissioning of a mixed-oxide fuel fabrication facility, West et al. (1991) used a nondestructive assay system to provide criticality safety monitoring, track the plutonium inventory, provide measurement of decontamination effectiveness, and provide quantitative characterization/assay of the waste. The system consisted of an integrated set of two passive neutron networks, two pulsed active neutron units, a high-resolution gamma spectrometer [high-purity germanium (HPGe)], and a neutron-coincidence counting unit. Waste determined to be less than 10 nCi/g was certified as class A low-level waste (LLW).

4.2.7 ALARA Guidelines

Contamination levels should be maintained ALARA to minimize the potential for the spread of contamination and to reduce the protective measures and equipment required. Control of radioactive material at the source and prevention of the generation of contamination are more effective and less costly than remediation.

4.3 PERSONNEL CONTAMINATION CONTROL

As described earlier, the purpose of contamination control is to prevent the ingestion or inhalation of plutonium by workers. This is primarily achieved by the engineered barriers discussed previously, containment, confinement, and ventilation control. Only if the primary controls fail or if there is a potential for personnel contamination during an activity are administrative controls such as protective clothing and respirators advisable.

4.3.1 Monitoring Philosophy

Monitoring the worker is necessary, not only to ensure that a potential intake is detected promptly and that the resulting internal dose is assessed, but to confirm the integrity of the engineered containment system and ensure the effectiveness of the overall radiation protection program.

There are several types of worker monitoring, some during and immediately following work with radioactive material and some scheduled for a later time at a preset frequency. This section addresses only methods of monitoring the worker at the workstation. Other methods are discussed in the section that deals with internal and external exposure controls.

Techniques to monitor the individual worker at the work site include:

- -- Frequent/routine surveys of gloves
- -- Exit surveys
- -- Nasal swipes
- -- Personal air sampling.

4.3.2 Monitoring Program

Instrumentation shall be provided and persons entering a plutonium work station shall be required to survey themselves at established frequencies. The requirements for radioactive contamination control and monitoring are found in 10 CFR 835.1102. As a minimum, workers should survey their gloves and coverall sleeves each time they are withdrawn from a glove box (or similar containment system) and after each glove replacement or bag-out operation.

Personnel monitoring for contamination should be mandatory at the egress from controlled areas and should be conducted in a verifiable manner. Assurance should be provided that personnel are monitored prior to breaks, meals, or exits from the plant site. Portal monitors, hand-and-shoe counters, and/or portable survey instruments may be used for this purpose. If employees are instructed to perform self-monitoring, the equipment should be set up in a "go/no-go" mode and employees should be clearly instructed in the required actions to take if predetermined action levels are exceeded. Frequent audits should be performed to verify that controls are adequate. Limiting the number of egress points and controlling personnel movement can minimize the numbers of locations where positive control of personnel monitoring shall be maintained.

4.3.3 Protective Clothing

Various types of protective clothing, including laboratory coats, shoe covers, gloves, coveralls, plastic or rubber suits, and air-purifying or atmosphere-supplying respiratory protective equipment, may be required for operations with transuranic radionuclides. The use of company-issue shoes and clothing for employees with work assignments in process areas can be a major aid in contamination control. Recently, some facilities are using disposable anti-contamination clothing. This may be a cost savings from a handling standpoint. However, disposal costs shall be considered. Additionally, consideration should be given to the potential for heat stress.

4.3.3.1 Requirements for Routine Operations

As a minimum, personnel who handle or work with unsealed sources of plutonium should wear coveralls, gloves, and shoe covers. For inspections or visits, lab coats and shoe covers may be permissible in those same areas. When contaminated wet areas are to be entered, water-repellent (plastic or rubber) clothing should be worn. No personal outer clothing should be permitted under coveralls.

When working with unsealed plutonium sources or in glove boxes, hands should be protected by a minimum of two barriers, for example, at least one pair of surgeon's gloves and one pair of glove-box gloves. Where manual dexterity is not required and the work involves a potential for piercing one or both layers of rubber gloves, leather gloves should be worn over the surgeon's gloves. Automated methods should be considered for replacing routine manual methods that have a high risk of piercing the gloves.

Protective clothing should be removed at the exit to radiologically controlled areas and personnel monitoring for contamination performed. If for some reason this is not practical, the movement of personnel should be strictly controlled from the exit area to a location where protective clothing can be removed.

4.3.3.2 Requirements for Special Maintenance

For special maintenance work that involves significant quantities of plutonium, a double barrier concept should be implemented. An example of minimum requirements for protective clothing is provided below:

- -- Two pairs of coveralls (and sometimes a plastic suit)
- -- Canvas boots taped to the inner pair of coveralls, with rubber boots over the canvas boots
- -- One pair of surgical gloves taped to the inner coveralls, with a leather, cotton, or rubber outer pair of gloves
- -- Respiratory protective device with hood taped to respirator.

To create a double barrier between the source and all extremities, surgeon gloves should be worn in addition to the glove-box gloves. In general, black Neoprene gloves are the standard glove-box glove and the most economical to use where process conditions do not produce rapid glove deterioration. However, alpha particles from surface dust layers can induce surface cracking in black Neoprene. Hypalon 0 is more resistant to surface cracking, acid deterioration, and ozone effects, and this characteristic will, in many cases, make Hypalon gloves the most economical, despite their higher unit cost.

In recent years many new types of glove-box gloves have been developed. Glove usage should be tailored to the particular needs of the job. For

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processes that require maximum dexterity, the 0.014-in. (0.038-cm) Neoprene gloves are still superior. Coated Hypalon gloves are superior to Neoprene for glove-box process operations that involve nitric acid or ozone levels that may cause deterioration. Ethylenepropylenediamine monomer (EPDM) gloves are used in some facilities and have good flexibility and are resistant to degradation caused by radiation and ozone. Greenhalgh et al. (1979) reported that Hypalon and EPDM gloves have greater than 30 times the longevity of Neoprene in low-level ozone concentration atmospheres. Viton gloves have proven to have a longer life than Neoprene gloves under many operating conditions, but suffer somewhat from stiffness. Where high gamma radiation levels are encountered, lead-loaded gloves may be necessary. However, their stiffness and workers' loss of manual dexterity should be considered in determining their influence on work efficiency and the total dose received.

Persons who perform operations that involve microspheres of 238 Pu, coated or uncoated, should be aware that the heat generation of a single 100-µm- to 200-µm-diameter sphere can melt through glove material. In addition, containment of a quantity of microspheres, especially coated microspheres, is difficult because of electrostatic repulsion. Microspheres have been observed climbing the walls of a glass beaker and spreading throughout a glove box.

Glove storage problems occur occasionally. Experiments and static tests have not provided an adequate explanation of the sporadic problems that have been encountered. Test results in which gloves were stored under different lighting conditions (ultraviolet and fluorescent) and under stressed conditions (creased or bent) have not been consistent. Tests of gloves seem to indicate that glove degradation is caused by the combined effect of ionizing radiation, ozone, and lighting. The glove inventory should be rotated to prevent the inventory from becoming outdated while on the shelf.

All gloves in normal use at plutonium processing installations should be inspected prior to each use. All operating personnel should perform contamination self-surveys after every glove usage. The glove inspections should be made each time by the same team of trained individuals, and the condition of each glove should be recorded so that glove failures can be anticipated and preventative measures can be taken. The development of a statistical basis for establishing the frequency of glove changes should be considered because such a basis may be cost-effective. For example, the change-out frequency could be planned so that gloves are changed at some fraction of the mean time between failures or more preferably some fraction of the minimum time between failures. This type of change-out program could also minimize personnel doses and potential contamination spread incidents associated with too-frequent glove replacement. This procedure may require that each glove use be categorized. A routine replacement program will not replace an inspection program, but it is a supplement to the inspections. The inspector's surgeon gloves should, of course, be surveyed after the inspection of each glove-box glove. Gloves that are in questionable condition should be changed without delay. Gloves that are not in use for the remainder of that shift should be capped off with a glove cover or plastic bag.

Gloves not in use should be stored inside the glove box in such a manner that they do not interfere with operations.

4.3.4 Respiratory Protection

Respiratory protection should be readily available. Respiratory protective equipment should be used for all bag-out operations, bag and glove changes, and any situation involving a potential or actual breach of confinement. Alternatively, the operation could be performed in a glovebag to maintain confinement. In any case, protection, in the form of air-purifying or atmosphere-supplying respirators, should be considered whenever concentrations of radionuclides in the air are likely to exceed 30% of the Derived Air Concentration (DAC) (i.e., where an individual without respiratory protection could receive 12 DAC-hrs in a week). For good performance, the respirator shall fit closely on the facial contours and make an impenetrable seal so that all air enters through the filter or is supplied by the breathing-air supply. ANSI Z88.2-1992 (ANSI, 1980b) describes qualitative and quantitative tests that should be used for verification of respirator fit at plutonium facilities. Respirator fit tests should be performed annually.

The respiratory protective device selected should provide a protection factor appropriate for the air concentration anticipated. ANSI Z88.2 provides protection factors guidance.

Air-supplied hoods are becoming more popular because a fitting is not required and facial hair does not prohibit their use. Protection factors up to 1000 are allowed for air-supplied hoods. All respirators, including air-supplied hoods, require approval. While NIOSH approves most respirators, some respirator types in use at DOE facilities are not part of the NIOSH testing program.

4.3.5 ALARA Guidelines

The total dose to an individual and the collective dose to the work force should be ALARA. When applied to personnel contamination or internal intakes, this generally means less than detectable dose with the best available commercial technology.

4.3.6 Personnel Release Criteria

The decision to release personnel with detectable plutonium contamination is made on a case-by-case basis. If the individual is injured and needs prompt medical attention, medical treatment will always take precedence, with compensatory measures made for the protection of medical personnel and facilities. If injuries are absent or do not require immediate attention, decontamination is preferable to ensure that the dose to the contaminated individual and the potential for inhalation by the victim and medical staff are minimized and the spread of contamination is prevented.

In a case where decontamination is incomplete due to injury to the skin or other reasons, the individual may be provisionally released with measures to prevent the spread of contamination. Module 2.13, Radiological Considerations for First Aid, of DOE-HDBK-1122-99 (DOE, 2009a) provides guidance on control of contamination for medical events. Guidance should be provided to the individual on control of potentially contaminated wound dressings if they are removed after the individual is released.

4.4 PERSONNEL DECONTAMINATION

Skin decontamination should be performed by RCTs or other members of the health physics staff. The treatment and decontamination of wounds should be performed by medical staff.

Nonabrasive methods should be used for skin decontamination to protect the tissues from deeper contamination. Masking tape should be used to remove dry contamination. Wet decontamination should be used to remove residual contamination. The skin should be gently scrubbed with soap and water. The following procedure is recommended:

- 1. Survey the worker to determine the contaminated areas of the skin. Have the medical staff treat and decontaminate breaks in the skin.
- 2. Wipe loose contamination with a gauze sponge or cotton applicators dipped in mild antiseptic detergent. Do not spread contamination to uncontaminated areas.
- 3. Rub the skin with the applicators to produce good sudsing.
- 4. Use soft bristle scrub brushes for fingernails and other difficult-to-clean areas as long as the skin barrier is maintained intact. It may be difficult to decontaminate the cuticles and under the nails.
- 5. Dry the skin area with cleansing tissue.
- 6. After the skin is thoroughly dry, survey it for any remaining contamination.
- 7. If no contamination is detected, apply a good-quality hand cream to prevent chapping.

Another effective nonabrasive decontamination method involves placing the contaminated hand in a cotton glove and then a Latex glove (causing the hand to perspire).

The decontamination factor is the ratio of the initial contamination level to the contamination level after decontamination methods are applied, as determined by survey instrument readings. Nonabrasive methods should be repeated until the decontamination factor between washes drops below 2 or 3 with significant contamination still remaining.

If contamination persists on the skin, a more abrasive decontamination method may be necessary. The decision to proceed with a more abrasive method should be based on the effectiveness of the decontamination. An abrasive soap should be applied with a moist gauze sponge or soft handbrush while rubbing the skin to develop a soapy lather. Care should be exercised to prevent damage to the skin surface.

Liberal irrigation with lukewarm water or saline solution is recommended for eye, nose, and mouth contamination. These procedures are performed by the medical staff to remove contamination.

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APPENDIX A

GLOSSARY

Terms used consistent with their regulatory definitions.

abnormal situation: Unplanned event or condition that adversely affects, potentially affects or indicates degradation in the safety, security, environmental or health protection performance or operation of a facility. **(RCS)**

activity median aerodynamic diameter: The diameter of a sphere having a density of 1 g cm₋₃ with the same terminal settling velocity in air as that of the aerosol particle whose activity is the median for the entire aerosol. (Internal Dosimetry Chapter of the IG)

air sampling: A form of air monitoring in which an air sample is collected and analyzed at a later time, sometimes referred to as retrospective air monitoring.

air monitoring: Actions to detect and quantify airborne radiological conditions by the collection of an air sample and the subsequent analysis either in real-time or off line laboratory analysis of the amount and type of radioactive material present in the workplace atmosphere. (Internal Dosimetry Chapter of the IG)

airborne radioactive material: Radioactive material in any chemical or physical form that is dissolved, mixed, suspended, or otherwise entrained in air.

alarm set point: The count rate at which a continuous air monitor will alarm, usually set to correspond to a specific airborne radioactive material concentration by calculating the sample medium buildup rate.

ambient air: The general air in the area of interest (e.g., the general room atmosphere) as distinct from a specific stream or volume of air that may have different properties.

breathing zone air monitoring: Actions conducted to detect and quantify the radiological conditions of air from the general volume of air breathed by the worker, usually at a height of 1 to 2 meters. See *personal air monitoring*. (Workplace Air Monitoring Chapter of the IG)

continuous air monitor (CAM): An instrument that continuously samples and measures the levels of airborne radioactive materials on a "real-time" basis and has alarm capabilities at preset levels.

decision level (*DL*, *L*c): The amount of a count or a count rate or the final instrument measurement of a quantity of analyte at or above which a decision is made that the analyte is definitely present. (ANSI, 2011b)

decontamination: The process of removing radioactive contamination and materials from personnel, equipment or areas. **(RCS)**

detector: A device or component that produces a measurable response to ionizing radiation. (Portable Instrument Calibration Chapter of the IG)

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DOELAP: The Department of Energy Laboratory Accreditation Program for personnel dosimetry. **(RCS)**

dose: The amount of energy deposited in body tissue due to radiation exposure. (RCS)

exposure: The general condition of being subjected to ionizing radiation, such as by exposure to ionizing radiation from external sources or to ionizing radiation sources inside the body. In this document, exposure does not refer to the radiological physics concept of charge liberated per unit mass of air. (Internal Dosimetry Chapter of the IG)

fissionable materials: A nuclide capable of sustaining a neutron - induced fission chain reaction (e.g., uranium-233, uranium-235, plutonium-238, plutonium 239, plutonium -241, neptunium-237, americium- 241 and curium-244) (**10 CFR 830**).

fixed contamination: Any area with detectable removable contamination less than the removable contamination values of Appendix D of 10 CFR 835 and fixed contamination at levels that exceed the total contamination values of Appendix D of 10 CFR 835. (Posting and Labeling Chapter of the IG)

fixed-location sampler: An air sampler located at a fixed location in the workplace.

grab sampling: A single sample removed from the workplace air over a short time interval, typically less than one hour.

hazardous waste: Because of its quantity, concentration, and physical, chemical, or infectious characteristics, hazardous waste may cause or significantly contribute to an increase in mortality, or an increase in serious irreversible or incapacitating reversible illness; it may pose a potential hazard to human health or the environment when improperly treated, stored, transported, disposed of, or otherwise managed. (DOE/S-0101)

high-efficiency particulate air (HEPA) filter: Throwaway extended pleated medium dry-type filter with 1) a rigid casing enclosing the full depth of the pleats, 2) a minimum particle removal efficiency of 99.97% for thermally generated monodisperse di-octyl phlalate smoke particles with a diameter of 0.3 µm, and 3) a maximum pressure drop of 1.0 in. w.g. when clean and operated at its rated airflow capacity. (**RCS**)

HLW: High-level waste (HLW) is the material that remains following the reprocessing of spent nuclear fuel and irradiated targets from reactors. The HLW is highly radioactive and generates heat on its own. Some of its elements will remain radioactive for thousands of years. Because of this, HLW shall be managed very carefully and all handling shall be performed from behind heavy protective shielding. (DOE/S-0101)

intake: The amount of radionuclide taken into the body by inhalation, absorption through intact skin, injection, ingestion or through wounds. Depending on the radionuclide involved, intakes may be reported in mass (e.g., µg, mg) or activity (e.g., µCi, Bq) units. (Internal Dosimetry Chapter of the IG)

LLW: Low-level waste (LLW) is any radioactive waste that is not HLW, spent nuclear fuel, TRU waste, or uranium mill tailings. The LLW is typically contaminated with small amounts of radioactivity dispensed in large amounts of material. The LLW is generated in every process

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involving radioactive materials in the DOE including decontamination and decommissioning projects. (DOE/S-0101)

minimum detectable amount/activity (MDA): The smallest amount (activity or mass) of an analyte in a sample that will be detected with a probability β of non-detection (Type II error) while accepting a probability α of erroneously deciding that a positive (non-zero) quantity of analyte is present in an appropriate blank sample (Type I error). (ANSI N13.30-2011)

MW: Mixed waste (MW) is waste that contains both radioactive and hazardous wastes. Any of the types of radioactive waste described can be a mixed waste if it contains any hazardous wastes. In fact, all of DOE's HLW is mixed waste because of the chemicals used to reprocess the fuel that resulted in the generation of the material or because it is suspected to contain hazardous materials. **(DOE/S-0101)**

personal air monitoring: The monitoring of air for radioactive particles in the immediate vicinity of an individual radiation worker's nose and mouth, usually by a portable sampling pump and collection tube (such as a lapel sampler) worn on the body. Personal air monitoring is a special case of breathing zone air monitoring. (Workplace Air Monitoring Chapter of the IG)

portable air sampler: An air sampler designed to be moved from area to area.

radiation-generating device (RDG): The collective term for devices which produce ionizing radiation, sealed sources which emit ionizing radiation, small particle accelerators used for single purpose applications which produce ionizing radiation (e.g., radiography), and electron-generating devices that produce x-rays incidentally. (Radiation-Generating Devices Chapter of the IG)

radioactive material: For the purposes of the standard, radioactive material includes any material, equipment or system component determined to be contaminated or suspected of being contaminated. Radioactive material also includes activated material, sealed and unsealed sources, and material that emits radiation. **(RCS)**

radiological work permit (RWP): The permit that identifies radiological conditions, establishes worker protection and monitoring requirements, and contains specific approvals for radiological work activities. The Radiological Work Permit serves as an administrative process for planning and controlling radiological work and informing the worker of the radiological conditions. **(RCS)**

radiological protection organization: A contractor organization responsible for radiation protection activities within contractor facilities. This organization is independent of the line organizational element responsible for production, operation, or research activities and should report to the contractor senior site executive. (Sealed Source Chapter of the IG)

real-time air monitoring: Collection and real-time analysis of the workplace atmosphere using continuous air monitors (CAMs).

refresher training: The training scheduled on the alternate year when full retraining is not completed for Radiological Worker I and Radiological Worker II personnel. **(RCS)**

removable contamination: Radioactive material that can be removed from surfaces by nondestructive means, such as casual contact, wiping, brushing or washing. **(RCS)**

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representative air sampling: The sampling of airborne radioactive material in a manner such that the sample collected closely approximates both the amount of activity and the physical and chemical properties (e.g., particle size and solubility) of the aerosol to which the workers may be exposed.

sanitary waste: Sanitary waste is waste that is neither hazardous nor radioactive. (DOE/S-0101)

source-specific air sampling: Collection of an air sample near an actual or likely release point in a work area using fixed-location samplers or portable air samplers.

survey: An evaluation of the radiological conditions and potential hazards incident to the production, use, transfer, release, disposal, or presence of radioactive material or other sources of radiation. When appropriate, such an evaluation includes a physical survey of the location of radioactive material and measurements or calculations of levels of radiation, or concentrations or quantities of radioactive material present.

TRU: Transuranic (TRU) waste refers to waste materials containing elements with atomic numbers greater than 92. These elements are generally alpha-emitting radionuclides that decay slowly. The TRU waste contains a concentration of these elements greater than 100 nCi/g. The TRU waste is not as intensely radioactive as HLW. The TRU waste also decays slowly, requiring long-term isolation. (DOE/S-0101)

workplace monitoring: The measurement of radioactive material and/or direct radiation levels in areas that could be routinely occupied by workers.