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Exam Preview:

1. The equivalent dose limit for the embryo/fetus of a declared pregnant worker is ___ rem for the entire gestation period, defined as the summation of external dose received and internal dose received during the gestation period.
    a. 0.25
    b. 0.5
    c. 0.75
    d. 0.85
2. According to the reference material, internal doses are not directly measured but are estimated or calculated based on knowledge of the material to which a worker may be exposed, and it's known or assumed biokinetic behavior.
    a. True
    b. False
3. Using Table 5.3. Lung Measurement Bioassay Goals for ^{241}Am as an Indicator of Aged Weapons-Grade Plutonium, what is the IRF value associated with weapons-grade plutonium that has been aged for 200 days?
    a. 4.21E-02
    b. 1.89E-01
    c. 3.14E-02
    d. 2.59E-02
4. The physical-chemical form of plutonium also affects the internal hazard posed. Oxides of plutonium tend to exhibit inhalation absorption type M behavior, whereas other compounds such as nitrates are assigned absorption type S by the ICRP.
    a. True
    b. False

5. The americium-tracer method has the advantage of better detection capability for some mixtures of plutonium. The detection level for this method with a plutonium/americium ratio of __ is typically 2-nCi plutonium in the lung.
    a. 10
    b. 15
    c. 20
    d. 25
6. The volume normalization technique typically normalizes whatever volume is collected to the ICRP Reference Man daily urine excretion volume of 1400 mL. Reference Woman excretion (1000 mL/d) may be used for gender-specific programs.
    a. True
    b. False
7. Using Table 6.2. Tissue Weighing Factors, what is the tissue weighting factor associated with the stomach?
    a. 0.01
    b. 0.05
    c. 0.12
    d. 0.20
8. According to the reference material, it is very difficult to accurately calculate dose rates from plutonium because of the wide range of photon energies and the relatively high abundance of photons.
    a. True
    b. False
9. The age and isotopic composition are very important in determining the dose rate from plutonium because of the ingrowth of ^{241}Am from the decay of ^{241}Pu, which has a half-life of only __ years.
    a. 10
    b. 15
    c. 20
    d. 22.5
10. Using Isotopic Composition of the Plutonium Used in the Extremity Dosimetry Measurements, which of the following isotopes has the highest weight percent?
    a. ^{236}Pu
    b. ^{241}Pu
    c. ^{242}Pu
    d. ^{239}Pu

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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, Occupational Radiation Protection. The Standard also supplements the DOE 10 CFR 835 Implementation Guide, DOE Orders, and DOE standard, DOE-STD-1098-2008, Radiological Control, (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).

5.0 INTERNAL DOSIMETRY

Internal dosimetry is an essential part of a quality health physics program at every facility where plutonium is handled or processed. The purpose of an internal dosimetry program is to monitor workplace activities, to assess accidental or inadvertent intakes of radioactive material, and to conduct internal dose assessments from bioassay measurement data.

It is DOE policy that facilities are designed, operated, and remediated to prevent intakes of radioactive materials. 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. In spite of excellent design and operation policies, inadvertent intakes of radioactive material can occur as a result of equipment malfunction, failure to follow procedures, or the unanticipated presence of radioactive material.

Experience has shown that the most common route for inadvertent plutonium intake is inhalation. Intakes can also occur by accidental ingestion or by wound contamination. Surveillance programs should be designed to rapidly detect a release in the event of a loss of radioactive material containment. Internal dosimetry programs should be tailored to the needs of each plutonium handling facility so that inadvertent intakes are discovered and quantified and workers' doses are determined by appropriate methods.

When workers are inadvertently exposed to radioactive material, appropriate corrective action should be taken to ensure that control and containment have been re-established. Prompt detection by routine workplace monitoring practices is essential to regaining control after any contamination spread or loss of containment. Prompt workplace indications of potential intake are also crucial to start special bioassay monitoring for intake and dose assessment. An early assessment of the probable severity of an intake and its corresponding dose, preferably within the first two hours of the intake, is needed for decisions on dose reduction therapy and event reporting. For plutonium and americium intakes, the bioassay data necessary for final dose assessment may require long periods of time (many months) to obtain. Until such data become available, ongoing preliminary assessments of intake and dose may be necessary to provide guidance for the administrative and medical management of the workers.

5.1 INTERNAL DOSE EVALUATION PROGRAM

Internal doses are not directly measured but are estimated or calculated based on knowledge of the material to which a worker may be exposed and it’s known or assumed biokinetic behavior. The common approach to internal dosimetry is to calculate an occupational intake based on worker bioassay measurements or workplace air-sample data and assumed breathing rates. Once an intake is calculated, appropriate internal doses to organs and tissues of concern can be estimated by using fundamental dosimetry principles, by various intake-to-dose conversion factors, which incorporate assumed biokinetic models, or by an appropriate computer code. Intake-to-dose conversion factors can be found in ICRP Publication 68 (ICRP, 1994b). Further discussion on intake and dose assessment is provided in Section 5.8.

Participation in internal dose evaluation programs is required by DOE for conditions identified in 10 CFR 835.402(c) (DOE, 2011). The internal dose evaluation program shall address both general workplace conditions and individual intakes.

Workplace conditions are monitored through air sampling programs as well as contamination surveys. For work that can have variable or changing conditions, more intensive surveillance may be required, using supplemental portable air samplers, continuous air monitors, or personal air samplers.

Individual worker monitoring for intakes is commonly performed using bioassay procedures. Bioassay monitoring includes both direct (in vivo) measurements of radioactivity in the body and indirect (in vitro) measurements of material excreted or removed from the body. Refer to Section 5.7.4 for information on assessing internal exposures from air monitoring data.

10 CFR 835.402(c) (DOE, 2011) specifies the requirements for participation in a radiological bioassay program. Because most plutonium facilities have a high degree of radiological control and containment for plutonium, chronic exposure to levels of occupational concern is unlikely and it is not considered likely that a worker would incur more than one unplanned intake in a year. Thus, participation in a bioassay program is generally based on the possibility that a single intake causing a dose in excess of 100-mrem committed effective dose CED might occur. Bioassay is also required if an intake is suspected for any reason.

Indications of intake include (but are not limited to) detection of facial or nasal contamination, air monitoring or sampling that indicates internal exposure, or any wound in which contamination is detected or suspected (See Section 5-9 for internal dosimetry recommended indicator and action levels.) The most common internal exposure monitoring program for workers is the bioassay monitoring program, which shall be designed for the specific nuclides and forms of material at a particular facility. Likely candidates for internal exposure monitoring include personnel who may be routinely exposed to surface or airborne contamination, or those identified by the foregoing workplace indicators.

Workplace monitoring for potential internal exposures is performed to verify the adequacy of containment and work practices. This monitoring includes air sampling, continuous air monitoring, personal contamination surveys, and workplace contamination surveys. Facilities are to be designed and operated to minimize internal exposure. Details regarding workplace monitoring and control practices are discussed in Section 4.0.

5.1.1 Performance Capabilities for Internal Exposure Monitoring

Bioassay monitoring programs shall be capable of showing compliance with the 5-rem/year stochastic and 50-rem/year deterministic dose limits of 10 CFR 835.202 (DOE, 2011). 10 CFR 835.402(c) (1) (DOE, 2011) identifies 100-mrem CED for all likely intakes as a level above which workers shall participate in a bioassay program. Therefore, ideally, such bioassay monitoring programs should be capable of detecting this level. In fact, this is not technically achievable for most routine plutonium bioassay programs. In order to meet this requirement, reliance shall be placed on workplace monitoring to identify potential intakes at the time they occur so that special bioassay monitoring can be initiated. Routine, periodic bioassay measurements have little chance of detecting a CED of 0.1 rem and can even have difficulty showing compliance with dose limits.

Performance capabilities for bioassay and internal dosimetry programs can be expressed as the minimum detectable dose, based on some combination of minimum detectable activity and frequency of measurement or time post-intake at which the measurement is made. The term “minimum detectable dose” is preferred over any variants of the occasionally encountered terms “dose-missed” or “potentially undetected dose,” which were usually defined as the same thing. The connotation of the latter terms is that of an actual intake which was not detected, whereas the intent was to define a measure of program sensitivity to doses that might have gone undetected had an intake occurred. The preferred term” minimum detectable dose” (MDD) ties the concept to the recognized terminology of minimum detectable activity (MDA). See G 441.1-1C, Ch. 1(DOE, 2011a), definition of MDA, for information on evaluating measurement results below these quantities.

The MDD for a bioassay monitoring program shall meet the aforementioned dose limit requirements of 10 CFR 835.202. A design goal of 100-mrem CED from all intakes of similar nuclides in a year is desirable but unrealistic for a routine program. To meet these requirements, bioassay programs should have measurement sensitivities (i.e., MDAs for bioassay measurements) established based on the material to which workers might be exposed. Examples of such sensitivities are given in Tables 5.1 and 5.2 (O'Connell, 2009) for pure ^{239}Pu monitored by urinalysis and fecal analysis, respectively. Table 5.3 (O'Connell, 2009) provides an example of the ^{241}Am sensitivity required for monitoring a mixture of weapons-grade plutonium, aged 30 years for ingrowth at time of intake. These tables illustrate the difficulty in relying on routine bioassay to demonstrate compliance with the limits and design goal.

Table 5.1. Urine Bioassay Goals[(a)] for ^{239}Pu

	Type M	Inhalation		Type S	Inhalation	
Days Post-Intake	Urine Intake Retention Fraction[(b)]	Dose Limit Goal[(c)] dpm	100-mrem Committed Effective Dose Goal[(d)] dpm	Urine Intake Retention Fraction[(b)]	Dose Limit Goal[(c)] dpm	100-mrem Committed Effective Dose Goal[(d)] dpm
1	2.46E-04	7.36E+00	4.63E-01	2.50E-06	8.23E-01	1.79E-02
7	2.40E-05	7.18E-01	4.52E-02	3.08E-07	1.01E-01	2.21E-03
30	9.51E-06	2.85E-01	1.79E-02	1.72E-07	5.67E-02	1.23E-03
60	8.11E-06	2.43E-01	1.53E-02	1.65E-07	5.43E-02	1.18E-03
90	7.12E-06	2.13E-01	1.34E-02	1.61E-07	5.30E-02	1.15E-03
200	5.12E-06	1.53E-01	9.63E-03	1.61E-07	5.30E-02	1.15E-03
400	3.71E-06	1.11E-01	6.98E-03	1.70E-07	5.60E-02	1.22E-03
1000	2.44E-06	7.30E-02	4.59E-03	1.77E-07	5.83E-02	1.27E-03
10000	6.87E-07	2.06E-02	1.29E-03	8.25E-08	2.72E-02	5.91E-04
20000	4.83E-07	1.45E-02	9.09E-04	5.83E-08	1.92E-02	4.18E-04

(a) The goals reflect the activity in a 24 hour urine void corresponding to either a 50 rem committed equivalent dose or a 0.1 rem committed effective dose.

(b) Incremental (i.e., sample collected in a 24-hour period ending at the time indicated) values for excreta obtained from "Intake Retention Functions Developed from Models Used in the Determination of Dose Coefficients Developed for ICRP Publication 68 – Particulate Inhalation" (Potter, 2002). See Section 5.8.1.

(c) Calculated as Goal (dpm) = Intake x IRF x 2220 dpm/nCi, where Intake (nCi) is the 50 rem committed equivalent dose limit/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from the ICRP Publication 68 Database (ICRP, 1994b) is shown below:

	Type M, rem/nCi	Type S, rem/nCi
Bone Surface	3.70	0.337

(d) Calculated as Goal (dpm) = Intake x IRF x 2220 dpm/nCi, where Intake (nCi) is the 0.1 rem committed effective dose threshold/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from the ICRP Publication 68 Database (ICRP, 1994b) is shown below:

	Type M, rem/nCi	Type S, rem/nCi
Effective Dose	0.118	0.031

Table 5.2. Fecal Bioassay Goals[a] for ^{239}Pu

	Type M	Inhalation		Type S	Inhalation	
Days Post-Intake	Fecal Intake Retention Fraction[b]	Dose Limit Goal[c] dpm	100-mrem Committed Effective Dose Goal[d] dpm	Fecal Intake Retention Fraction[b]	Dose Limit Goal[c] dpm	100-mrem Committed Effective Dose Goal[d] dpm
1	1.10E-01	3.30E+03	2.07E+02	1.16E-01	3.82E+04	8.31E+02
7	2.29E-03	6.87E+01	4.31E+00	2.42E-03	7.97E+02	1.73E+01
30	2.81E-04	8.43E+00	5.29E-01	3.51E-04	1.16E+02	2.51E+00
60	1.31E-04	3.93E+00	2.46E-01	1.86E-04	6.13E+01	1.33E+00
90	6.65E-05	2.00E+00	1.25E-01	1.07E-04	3.52E+01	7.66E-01
200	1.42E-05	4.26E-01	2.67E-02	3.32E-05	1.09E+01	2.38E-01
400	4.67E-06	1.40E-01	8.79E-03	2.13E-05	7.02E+00	1.53E-01
1000	1.04E-06	3.12E-02	1.96E-03	1.12E-05	3.69E+00	8.02E-02
10000	2.96E-07	8.88E-03	5.57E-04	9.53E-08	3.14E-02	6.82E-04
20000	2.11E-07	6.33E-03	3.97E-04	3.20E-08	1.05E-02	2.29E-04

(a) The goals reflect the activity in a 24 hour fecal sample corresponding to either a 50 rem committed equivalent dose or a 0.1 rem committed effective dose.

(b) Incremental (i.e., sample collected in a 24-hour period ending at the time indicated) values for excreta obtained from "Intake Retention Functions Developed from Models Used in the Determination of Dose Coefficients Developed for ICRP Publication 68 – Particulate Inhalation" (Potter, 2002). See Section 5.8.1.

(c) Calculated as Goal (dpm) = Intake x IRF x 2220 dpm/nCi, where Intake (nCi) is the 50 rem committed equivalent dose limit/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from the ICRP Publication 68 Database (ICRP, 1994b) is shown below:

	Type M, rem/nCi	Type S, rem/nCi
Bone Surface	3.70	0.337

(d) Calculated as Goal (dpm) = Intake x IRF x 2220 dpm/nCi, where Intake (nCi) is the 0.1 rem committed effective dose threshold/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from the ICRP Publication 68 Database (ICRP, 1994b) is shown below:

	Type M, rem/nCi	Type S, rem/nCi
Effective Dose	0.118	0.031

Table 5.3. Lung Measurement Bioassay Goals for ^{241}Am as an Indicator of Aged Weapons-Grade Plutonium[a]

		Type M Plutonium	Inhalation
Time - days	IRF[d]	Dose limit Goal[b] nCi ^{241}Am	100-mrem Committed Effective Dose Goal[c] nCi ^{241}Am
1	1.25E-02	3.39E-02	2.21E-03
3	1.69E-02	4.58E-02	2.99E-03
7	3.10E-04	8.40E-04	5.49E-05
10	1.54E-05	4.17E-05	2.73E-06
30	3.79E-14	0	0
		Type S Plutonium	Inhalation
Time - days	IRF[d]	Dose limit Goal[b] nCi ^{241}Am	100-mrem Committed Effective Dose Goal[c] nCi ^{241}Am
1	1.89E-01	1.73E+00	8.43E-02
7	6.01E-02	5.49E-01	2.68E-02
30	4.96E-02	4.53E-01	2.21E-02
60	4.21E-02	3.84E-01	1.88E-02
90	3.79E-02	3.46E-01	1.69E-02
200	3.14E-02	2.87E-01	1.40E-02
400	2.59E-02	2.36E-01	1.16E-02

(a) Defined as a Pu mixture consisting of, by weight %, 93% ^{239}Pu, 6.1% ^{240}Pu, 0.8% ^{241}Pu, 0.05% ^{238}Pu, and 0.05% ^{242}Pu, with 30 years allowed for ^{241}Am ingrowth (Table 5.4). Additional ingrowth of ^{241}Am post intake is negligible.

(b) Intake of a 30 year aged 6% mix of Pu, with the specific activities listed in Table 5.5, giving a committed equivalent dose of 50 rem to the bone surfaces. Calculated as:

Dose Limit Goal (nCi ^{241}Am) =

$$\frac{\text{50 Rem x specific activity (Ci/g) of the } ^{241}\text{Am in the 30 year aged 6\% Pu Mix x IRF}}{\text{Mixture dose coefficient (Rem/g) x 1.0E-9 Ci/nCi}}$$

Where: Mixture Dose Coefficient (Rem/g) =

∑ (Specific Activity (Ci/g) of a 30 year aged 6% Pu Mix (from Table 5.5) x applicable radionuclide dose conversion factor (Rem/Ci) derived from the ICRP Publication 68 Database (ICRP, 1994b). See table below.

(c) Intake of a 30 year aged 6% mix of Pu, with the specific activities listed in Table 5.5, giving a committed effective dose of 0.1 rem.

100-mrem committed effective dose goal (nCi ^{241}Am) =

$$\frac{\text{0.1 Rem x specific activity (Ci/g) of the } ^{241}\text{Am in the 30 year aged 6\% Pu Mix x IRF}}{\text{Mixture dose coefficient (Rem/g) x 1.0E-9 Ci/nCi}}$$

Where: Mixture Dose Coefficient (Rem/g) =

∑ (Specific Activity (Ci/g) of a 5year aged 6% Pu Mix (from Table 5.5) x applicable radionuclide dose conversion factor (Rem/Ci) derived from the ICRP Publication 68 Database (ICRP, 1994b). See following table.

Dose Coefficients (Rem/Ci)

Absorption Type/		Pu-238	Pu-239	Pu-241	Pu-242	Am-241
M	Bone Surface	3.40E+09	3.70E+09	7.40E+07	3.60E+09	4.10E+09
S	Bone Surface	3.00E+08	3.40E+08	7.40E+06	3.20E+08	4.10E+09*
M	Whole Body	1.10E+08	1.20E+08	2.15E+06	1.10E+08	1.00E+08
S	Whole Body	4.10E+07	3.10E+07	3.10E+05	2.80E+07	1.00E+08*

* Type M dose coefficient for Americium-241 is used.

(d) Incremental (i.e., sample collected in a 24-hour period ending at the time indicated) values for excreta obtained from "Intake Retention Functions Developed from Models Used in the Determination of Dose Coefficients Developed for ICRP Publication 68 – Particulate Inhalation" (Potter, 2002). See Section 5.8.1.

The problem is simply that the measurement technology is not available to provide the sensitivities required for the 100-mrem goal using routine, periodic measurements at reasonable frequencies. For example, routine Hanford analyses of plutonium in urine has a detection limit of 0.02 dpm/sample, plutonium in feces has a detection limit of 0.2 dpm/sample and americium lung counting has a detection limit of 0.16 nCi (Carbaugh, 2003). As shown in Table 5.1, for material type M, monthly routine bioassay measurements would not achieve a sensitivity for the 100-mrem goal. For material type S, even weekly routine bioassay measurements would not have the sensitivity for the 100-mrem goal. Monthly, or even bi-monthly, fecal bioassay, as shown in Table 5.2, could achieve the requisite sensitivity for the100-mrem goal for material type M (worst case). However, as discussed in Section 5.3.3.2, there are associated difficulties in including fecal analysis as part of a routine bioassay program. As shown in Table 5.3, lung counting for material types M or S would also not achieve a sensitivity for the 100-mrem goal.

Therefore, because the goal of 100-mrem CED cannot, typically, be met through routine bioassay, the radiation protection organization should take the following administrative actions:

-- ensure that adequate control measures are applied to prevent intakes

-- document the adequate control measures for auditing purposes

-- upgrade bioassay measurement systems and workplace monitoring practices to provide state-of-the-art measurements

-- ensure that internal dose assessments use commercially viable technology.

-- ensure workplace monitoring programs are designed to identify potential intakes.

All confirmed occupational intakes of plutonium, regardless of magnitude, should be assessed. The results of all bioassay and other measurements needed to support the quality of measurements and dose assessment should be recorded and maintained. The recording and reporting requirements for internal dosimetry data are set forth in Section 3.7 of this report; however, the following is a summary list of internal dosimetry information for which recording is required:

-- Total CED from all intakes during a year

-- committed equivalent dose to organs or tissues of concern from all intakes during a year

-- magnitude of intake for each radionuclide during a year

-- data necessary to allow subsequent verification, correction, or recalculation of doses

-- gestation period equivalent dose to the embryo/fetus from intake by the declared pregnant worker during the entire gestation period.

Radiation exposure records programs shall also provide for the summation of internal and external doses, as required by 10 CFR 835.702 (DOE, 2011). While the summation process is not necessarily performed under a site internal dosimetry program, it behooves the program to recognize what is required. The following summations are identified by 10 CFR 835.702(c) (5) and (6):

-- Total effective dose (TED) defined as the summation of effective dose (for external exposures) and the CED

-- summation of the equivalent dose to the whole body from external exposure and the committed equivalent dose to organs or tissues of concern

-- cumulative TED

-- for the embryo/fetus of a declared pregnant worker, the summation of the equivalent dose to the whole body to the mother from external exposure during the entire gestation period and the gestation period equivalent dose to the embryo/fetus from intakes by the mother during the entire gestation period.

Doses should be calculated and recorded for any confirmed plutonium intake. What constitutes a confirmed intake is discussed in Section 5.7. Along with the doses, supporting records shall be maintained, including the bioassay data, assumptions, biokinetic models, and calculational methods used to estimate the doses. These may be included in letter-report dose assessments, databases, technical basis documents, and similar records, either singly or in combination.

5.1.2 Protection of the Embryo/Fetus, Minors, and Members of the Public

The equivalent dose limit for the embryo/fetus of a declared pregnant worker is 0.5 rem for the entire gestation period, defined as the summation of external dose received and internal dose received during the gestation period (not the 50-year committed internal dose). Internal exposure monitoring is required if an intake is likely to result in more than 10% of that limit (i.e., 50 mrem for the gestation period). As discussed in more detail in Section 5.6., providing adequate protection to keep the mother's intakes below the occupational limits will also provide adequate protection for the embryo/fetus. Thus, special bioassay for plutonium or americium related to pregnancy is not required. As a matter of caution, some sites try to obtain baseline bioassays as soon as a pregnancy is declared, with another baseline bioassay

following the end of pregnancy. Some sites also offer to restrict pregnant workers from jobs with relatively high potential for occupational intakes.

Minors and members of the public are limited, in part, by 10 CFR 835.207 and 10 CFR 835.208 (DOE, 2011) to a TED of 0.1 rem/year. Internal exposure monitoring is required if an intake is likely to result in 50% of that limit (0.05 rem). As noted in Section 5.1.1, because bioassay monitoring is not likely to be sufficiently sensitive to identify such intakes on a routine basis, enhanced workplace surveillance or restriction of access may be required.

5.2 CHARACTERIZATION OF INTERNAL HAZARDS

Plutonium can be encountered in a wide range of mixtures, e.g., a pure isotope in a standard solution, a highly variable combination of isotopes in so-called “weapons grade” or “fuels grade” Pu, or commercial spent fuel. In addition, the age of a mixture significantly affects its isotopic composition. As a typical weapons or fuels grade mixture ages, the ^{241}Pu decays to ^{241}Am. Although the mass changes may be quite small, the overall result can be a significant build-up of ^{241}Am radioactivity with time. This buildup can make the mixture somewhat easier to detect by in vivo methods. Table 5.4 shows some example plutonium mixtures which might be encountered in DOE facilities. Isotopically pure forms of radionuclides can also be encountered. Table 5.5 demonstrates the impact of aging on the activity composition of two mixtures. The composition of plutonium in the facility can significantly affect the design and capabilities of an internal dosimetry program. As part of the program technical basis, the plutonium mixtures need to be determined. In addition, determinations should be made at the time of identified incidents of potential intake. Methods for such determination may include radiochemical analysis or chemistry followed by mass spectrometry.

The physical-chemical form of plutonium also affects the internal hazard posed. Oxides of plutonium tend to exhibit inhalation absorption type S behavior, whereas other compounds such as nitrates are assigned absorption type M by the ICRP. However, as noted in Section 2.4.1, extremes have been observed with regard to both highly soluble and highly insoluble forms, leading to the good practice of performing dissolution rate (i.e., solubility) tests on standard materials in a facility.

As plutonium ages in a residual, loose contamination form, such as might be found in old duct work, glove boxes, or other such components, it can be expected to undergo slow oxidation to a more insoluble form. Thus, absorption type S forms of plutonium may be reasonable assumptions of what to expect during many decommissioning operations.

Particle size is an important consideration for inhalation exposures. The normal practice for an aerosol is to identify the activity median aerodynamic diameter and its associated particle-size distribution. Particle sizes of 10 μm or less are considered respirable. It is acceptable to assume a 5-μm particle size for dosimetry purposes because actual particle size information is usually lacking. Particle size data are most readily obtainable for chronic exposure situations.

Unless representative air sampling is performed in the immediate proximity of a worker during abnormal working conditions, the practical likelihood of obtaining good particle-size information is slim.

5.3 SCOPE OF BIOASSAY PROGRAM

The relatively low annual limit on intake of plutonium renders its radiation hazard substantially more restrictive than its industrial hygiene or chemical toxicity hazard. Thus, internal radiation dose or intake monitoring is the appropriate focus of bioassay monitoring.

Table 5.4. Example Plutonium Isotope Mixtures Immediately Post-Separation wt%

Isotope	Weapons-Grade Plutonium (6% ^{240}Pu Mixture)	Fuels-Grade Plutonium (12% ^{240}Pu Mixture)	Spent Commercial Fuel (25% ^{240}Pu Mixture)
^{238}Pu	0.05	0.10	1.49
^{239}Pu	93.0	84.4	59.50
^{240}Pu	6.1	12.4	23.98
^{241}Pu	0.8	3.0	10.33
^{242}Pu	0.05	0.1	4.0
^{240}Am	0.0	0.0	0.0

Table 5.5. Activity Composition with Age for Reference 6% and 12% ^{240}Pu Mixtures

(Carbaugh, 2003)

Isotopic Component	Reference 6 % Pu Mix [(a)] Fresh	Reference 6 % Pu Mix [(a)] Aged	Reference 12% Pu Mix [(a)] Fresh	Reference 12% Pu Mix [(a)] Aged
Specific Activity In Mixture, Ci/g				
^{238}Pu	8.6E-3	6.7E-3	1.7E-2	1.4E-2
$^{239+240}$Pu	7.2E-2	7.2E-2	8.0E-2	8.0E-2
^{241}Pu	8.2E-1	1.9E-1	3.1E+0	7.3E-1
^{242}Pu	2.0E-6	2.0E-6	3.9E-6	3.9E-6
^{241}Am	5.3E-5	2.0E-2	2.0E-4	7.6E-2
Pu-alpha	8.1E-2	7.8E-2	9.7E-2	9.3E-2
Total alpha	8.1E-2	9.8E-2	9.7E-2	1.7E-1
Activity Ratios				
$^{239+240}$Pu:^{241}Am	NA	3.5E+0	NA	1.0E+0
Pu-alpha:^{241}Am	NA	4.8E+0	NA	2.2E+0
^{241}Pu:$^{239+240}$Pu	1.2E+1	2.7E+0	3.8E+1	9.2E+0

(a) % = nominal ^{240}Pu weight percent in mixtures.
Fresh = 2 weeks of ^{241}Am ingrowth following separation.
Aged = 30 years of ^{241}Am ingrowth following separation.

5.3.1 Classification of Bioassay Measurements

Bioassay measurements can be classified according to the primary reason for their performance. This is a useful practice for historically documenting why a worker participated in a bioassay program. Numerous reasons for bioassay measurements may be defined for specific facilities; some suggested common classifications are as follows:

-- **Baseline measurements** are used to establish a pre-exposure condition, either for a new employee or as a result of a new work assignment. The standard, Radiological Control (DOE, 2017), recommends baseline measurements if workers are considered likely to receive intakes resulting in greater than 100-mrem CED. It is a good practice to perform such measurements for newly hired employees, intra-company transferees, or workers transferred from facilities where bioassay measurements may not have been required. In addition, baseline measurements can verify workers' status for special work assignments. For plutonium bioassay, baseline measurements made before any occupational exposure can be expected to yield no detectable results using current technology.

Exempting workers from baseline bioassay implies accepting any detectable results as likely attributable to current occupational exposure. However, requiring baseline measurements can potentially impact the schedule of short-term jobs; the time required to obtain a chest count and a large-volume urine sample may add a day or two delay to entry procedures. Moreover, missing a baseline for a long-term employee who will be placed on a routine bioassay program is not likely to be as troublesome as not obtaining a baseline for a short-term worker who provides a termination sample that shows detection of plutonium after the worker has left the site and is difficult to reach for follow-up.

-- **Routine, or periodic, measurements** are performed on a predetermined schedule (e.g., an annual or quarterly frequency).

-- **Special bioassay measurements** are performed as follow-up to unusual routine results or suspected intakes (See Section 5.9 for recommended internal dosimetry indicator and action levels).

-- **End of assignment or termination measurements** are performed following completion of specific work or at the time of termination of employment. The DOE Standard, Radiological Control (DOE, 2017), recommends that workers who participate in bioassay programs have appropriate termination measurements.

Bioassay classification is important because the purpose of a sample may affect the collection and analysis or monitoring method chosen. For example, single-void urine samples are not adequate for routine monitoring of potential plutonium exposure, but can provide important information for dose-reduction therapy following a suspected intake; samples representative of excretion over a 24-hour period should be collected for quantitative intake and dose assessment. The date of sample collection (and possibly the time of collection) can be very important to special monitoring performed to assess intake. However, these are

much less important with regard to periodic monitoring, for which measurements are not expected to show detectable activity and when any detection whatsoever is likely to initiate investigation and special bioassay.

5.3.2 Monitoring Requirements and Selection of Employees

Workers who are considered likely to have intakes resulting in excess of 100-mrem CED are required to participate in a bioassay program. However, because of the extensive radiological control practices for plutonium facilities, including a high degree of engineered barrier containment, no typical plutonium worker is likely to have intakes of 100-mrem CED or more. However, this should not be used as an excuse to exclude workers from routine bioassay. Although no one should be considered likely to have intakes resulting in 100-mrem CED, some workers are at significantly higher risk for incurring an intake than others and should be on routine bioassay.

The workers at highest risk of incurring an intake are the ones in closest contact with the material. Typically, these are the operators, maintenance, and health physics personnel handling plutonium or plutonium-contaminated objects in the course of routine glove-box, maintenance, or decommissioning operations. In the event of containment system failure, or failure respiratory protection devices, it is these workers who will most likely incur exposure and subsequent intake. These workers should be on a routine bioassay program designed to meet the requirements of 10 CFR 835 (DOE, 2011) as a kind of safety net to identify intakes which might have gone undetected by workplace monitoring.

Other workers (e.g., supervisors, inspectors, observers, guards, and tour groups) who work in or visit a plutonium facility but are not directly working with the material or contaminated objects are at a substantially lower risk for incurring an intake. Although these people may not need to be on a routine bioassay program, they should be subject to participation in a special bioassay program if workplace indications suggest loss of control or containment.

5.3.3 Selection of Bioassay Monitoring Techniques

Bioassay monitoring techniques fall into two broad categories, direct measurement of radioactive materials in the body (in vivo counting) and analysis of material removed from the body for laboratory in vitro analysis. In vivo counting includes measurements of the chest, lung, skeleton, liver, and wounds. In vitro measurements include urinalysis, fecal analysis, and occasionally analysis of tissue, sputum, or blood samples. Methods for in vitro analysis include liquid scintillation counting, fluorescence measurements, gamma spectrometry, chemical separation followed by electrode position, and counting with radiation detectors. Selby et al. (1994) provide a brief overview of bioassay techniques and capabilities. Further discussion of the techniques is provided below.

5.3.3.1 In Vivo Counting

Direct bioassay (in vivo counting) is the measurement of radiations emitted from radioactive material taken into and deposited in the body. Direct bioassay is appropriate for detection and measurement of photons emitted by plutonium and its

decay products. Lung, wound, liver, and skeleton counting are examples of in vivo monitoring most commonly used for plutonium and its progeny. Whole body counting, commonly used for monitoring high-energy fission and activation products in the body, is ineffective for direct measurement of plutonium due to the very low energy of photons emitted from plutonium and its decay products unless the plutonium is intimately mixed in a high-energy photon-emitting matrix, such as spent fuel.

Some low-energy x-rays emitted by plutonium decay products are energetic enough to escape the body. When direct bioassay is used, the detection system should be calibrated for the radionuclides to be measured in the appropriate organs. All calibration procedures, calibration records, and quality control data should be maintained. Energies most commonly used for plutonium monitoring are the 17-keV L X-rays and the 60-keV gamma of ^{241}Am. Mixtures of spent fuel material can lend themselves to whole body counting if the ratio of a readily detectable high-energy gamma-emitter (i.e., ^{137}Cs) to plutonium is known.

A plutonium facility should have the capability to detect and assess depositions of plutonium in the lungs of radiation workers. The major objective of lung counting is to provide measurements of suspected intakes triggered by workplace monitoring results. Lung measurements should be made to provide an early estimate of the magnitude of the intake and resulting lung deposition.

Two methods have been used to detect plutonium in the lung: the L x-ray method and the americium-tracer method. The L x-ray method is based on the measurement of L X-rays following the decay of plutonium. This method provides a direct measurement of plutonium. The detection capability of the method may be on the order of tens of nanocuries for plutonium and requires an accurate measurement of the chest wall thickness (because of the large attenuation of the low-energy X-rays by the rib cage and overlying tissues). Other problems that complicate the measurement of L X-rays are (1) the difference in attenuation in muscle and fat, (2) the possibility of nonuniform distribution of the plutonium in the lung, and (3) interferences from radionuclides in other organs or from other radionuclides in the lung.

The americium-tracer method has the advantage of better detection capability for some mixtures of plutonium. The typical MDA for ^{241}Am lung counting is 0.1 to 0.2 nCi. The americium-tracer method depends on the plutonium/americium ratio, which shall be independently determined or estimated for each intake. The detection level for this method with a plutonium/americium ratio of 15 is typically 2-nCi plutonium in the lung. The americium-tracer method also has the advantage of being less affected by attenuation in the chest wall or by variations in the muscle/fat ratio. However, it has the disadvantage of requiring an estimate of the plutonium/americium ratio, both initially and at long times post intake. This ratio may change over time because of ingrowth of ^{241}Am as the decay product of ^{241}Pu or because americium may naturally clear from the lungs and translocate among internal organs at a rate different than that for plutonium.

The most widely used systems for lung counting are high-purity germanium detectors, thin sodium-iodide detectors, phoswich detectors, and proportional counters. Multiple high-purity germanium detectors have advantages over the other detector systems because of their good resolution, allowing better identification of

the radionuclide, better detectability, and better background prediction capability. The main disadvantages of germanium detector arrays are their higher cost relative to other types of in vivo detectors and their lower reliability.

Measurement equipment to detect and measure plutonium contamination in wounds should be available at all plutonium facilities. Instrumentation used for this purpose may include thin-crystal NaI(Tl), intrinsic germanium, or Si(Li) detectors. The detection level for plutonium wound measurements is typically 0.1 nCi for ^{239}Pu. Correction for depth due to absorption of photons in the overlying tissues should be considered. Collimated detectors are useful for determining the location of the plutonium in wounds.

Estimates of the depth of plutonium contamination in a wound may be made using solid-state germanium or Si(Li) detectors to measure the relative absorption of the low-energy X-rays emitted by plutonium. Information about depth is important for determining whether tissue excision is necessary to remove the contamination.

5.3.3.2 In Vitro Analysis

The two most common forms of in vitro analysis are urinalysis and fecal analysis.

Urinalysis. Urine sampling provides useful information about the amount of plutonium excreted following an intake. After chemical isolation, the plutonium in urine samples may be determined by various methods including: alpha spectrometry (gas-flow proportional or surface-barrier detection), alpha counting (zinc sulfide or liquid scintillation counting), fission track counting, and mass spectrometry. Analytical procedures for in vitro measurement of plutonium and other radionuclides have been published (Volchok and dePlanque, 1983; Gautier, 1983).

Urine samples should be collected away from the plutonium facility to minimize cross-contamination. Samples should be collected in contamination-free containers; measures should be considered for minimizing plateout on walls of container surfaces (such as by addition of trace amounts of gold, oxalate, or nitric acid).

Fecal Analysis. Fecal analysis is a useful procedure for evaluating the excretion of plutonium and many other radioactive materials because more than half of the material deposited in the upper respiratory tract is cleared rapidly to the stomach and GI tract.

The total fecal plus urinary elimination for the first few days after exposure, combined with in vivo counts that might be obtained, may provide the earliest and most accurate assessment of intake. Fecal samples taken during the second and third day after an inhalation incident are likely to provide the most useful data because the GI hold-up time may vary from a few hours to a few days.

Fecal sampling is primarily a monitoring procedure for confirming and evaluating suspected intakes, but is used at some plutonium facilities for routine periodic monitoring as well. Workers may find fecal sampling unpleasant or objectionable, and laboratory technicians may also have aversion to fecal sample analysis. Some of these problems may be minimized if commercial fecal sample collection kits are used for convenient collection and handling of samples (Fisher et al., 1982). Collection

kits also provide a means for collecting uncontaminated samples. Fecal samples may require additional sample preparation before analysis.

5.4 ESTABLISHING BIOASSAY FREQUENCY

The bioassay measurement frequency should be based on 1) the potential risks of an intake occurring and 2) the sensitivity of a bioassay program to detecting potential intakes. The bioassay program sensitivity can be selected using specified intervals between measurements based on the MDD associated with an interval.

The rationale for the selected bioassay measurement frequency should also be documented. It is appropriate to evaluate the probability of intake and to modify the sampling frequency based on that probability.

The frequency of bioassay measurements should normally not be decreased because analytical results are below the detection level. The bioassay program should be maintained to confirm the proper functioning of the overall internal exposure control program and to document the absence of significant intakes of radionuclides.

5.4.1 Frequency Based on Program Sensitivity

The minimum detectable dose concept refers to the potential dose associated with an MDA bioassay measurement at a given time interval post-intake. The pattern of retention of activity in the body, the MDA for a bioassay measurement technique, and the frequency with which that technique is applied define a quantity of intake that could go undetected by the bioassay program. An intake of such a magnitude would not be detected if it occurred immediately after a bioassay measurement and if it were eliminated from the body at such a rate that nothing was detected during the next scheduled measurement. The dose resulting from such an intake would be the MDD for that particular measurement technique and frequency.

Estimates of MDD in terms of CED should be documented for each measurement technique, MDA, and frequency. Retention functions specific to the various chemical forms and particle size distributions found in the facility should be used. Examples of MDD tabulations can be found in La Bone et al. (1993) and Carbaugh et al. (1994). In establishing MDD tables, it is important to consider dose contributions from all appropriate radionuclides in any mixture, rather than just the dose contribution from the bioassay indicator nuclide.

5.4.2 Frequency Based on Potential Risk of Intake

As discussed in Section 5.3.2, although plutonium workers are not generally considered to be at high risk of incurring intakes that might result in CEDs of 100 mrem or more, any plutonium worker can be considered to have the potential for such an intake. However, having the potential for intake does not mean that they are likely to incur an intake.

Workers who have the highest potential risk for an intake are those most closely working with plutonium or plutonium-contaminated material. Typically, these workers are glove-box workers, maintenance workers, and operational health physics surveillance staff. These workers should be on a routine plutonium or americium

bioassay program, including urinalysis and in vivo measurements. Such programs are relatively insensitive compared to the 100-mrem CED goal and are a safety net intended to catch intakes of significance relative to regulatory limits, rather than substantially lower administrative levels. Selection of bioassay frequency depends on the facility experience with potential intakes, the perceived likelihood of intake, and the MDD of a program. Annual urinalyses and in vivo chest counts are fairly typical. More frequent (e.g., semi-annual or quarterly) measurements may permit more timely review of workplace indicators in the event that an abnormal bioassay result is obtained, but do not necessarily mean a more sensitive program.

Plutonium facility decommissioning projects may present a different set of challenges for worker protection. In particular it is likely that clean up of areas will involve more plutonium that is not contained than is the case during normal operations. In addition, the workers involved may be relatively transient as the project progress through phases requiring different craft labor mixes. This being the case, more frequent bioassay may be necessary to provide good assurance that dose limits are not exceeded. As discussed in Section 5.3.1, it is likely that program administrators will require a baseline measurement prior to the start of work and another at the termination of work. However, if the worker moves between tasks, it may be difficult to determine the source of an uptake without intermittent bioassay. In such cases, the use of breathing zone air samplers may be appropriate.

5.4.3 Special Bioassay as Supplements to Routine Bioassay Programs

Special bioassay programs for workers with known or suspected acute inhalation intakes of plutonium or other alpha-emitting radionuclides should include both urine and fecal sampling. Special bioassay measurements should be initiated for each employee in a contaminated work area when surface contamination is detected by routine surveillance if it is possible that the contamination resulted in a CED of 100 mrem or greater. Excreta samples should not be collected where they may be contaminated by external sources of plutonium. Ideally, total urine and feces should be collected for about a week following intake. This permits a sensitive assessment of potential intake and internal dose. Longer term special samples collected at various times from a month to a year following intake can help to discriminate between ingestion, absorption type M inhalation, and absorption type S inhalation. See Section 5.9 for indicator levels where special bioassay should be considered.

5.4.4 Long-term Follow-up Bioassay Programs

Following an intake a long-term follow-up bioassay program may be required for a worker to compare the actual excreta or in vivo results with those projected by the evaluation. This is important to verify the accuracy of intake and dose assessments. The frequency and duration of a special program is dependent upon the projected values; it is suggested that as long as a worker continues to have detectable bioassay results, he or she should continue to be monitored. It is particularly important to have good baseline data and projections for individuals who return to plutonium work. The ability of a bioassay program to distinguish between an established, elevated baseline and a new potential intake is important in the continued monitoring of workers once an intake has occurred. Because of statistical fluctuations in low-level plutonium and americium measurements, it can be very difficult to identify a new intake by routine bioassay if a worker has an elevated baseline.

5.5 ADMINISTRATION OF A BIOASSAY PROGRAM

Administering a bioassay program requires that the policies, procedures, materials, support facilities, and staff be in place to enable a bioassay program to commence. Among the administrative items to address are the following:

-- Management policy requiring participation in bioassay program by appropriate workers (may be part of an overall radiation protection policy)

-- implementing procedures (e.g., criteria for who should participate, scheduling, sample kit instructions, sample kit issue/receipt, follow-up to unsuccessful sample or measurement attempts, data-handling)

-- arrangements with appropriate analytical laboratories, including specifications of analysis sensitivity, processing times, reporting requirements, and quality assurance provisions

-- onsite support facilities (e.g., sample kit storage locations, sample kit issue/collection stations, measurement laboratory facilities, equipment maintenance)

-- staff selection, qualification, and training.

Recommendations for testing criteria for radiobioassay laboratories are in Performance Criteria for Radiobioassay, ANSI/HPS N13.30 (ANSI, 2011b). These recommendations include calculational methods and performance criteria for bias, precision, and testing levels.

Some sites have established brief flyers or brochures describing their bioassay measurements. These may be distributed to workers during classroom training, upon notification of scheduled measurements, or at the time of the measurement or sample.

5.5.1 In Vivo Monitoring

The scheduling and measurement process for obtaining in vivo measurements is usually straightforward. Workers are scheduled for the measurements and results are available shortly after the measurement is completed. Counting times for in vivo ^{241}Am measurements range from about 15 minutes to an hour or more, depending on the type of measurement and sensitivity required. The long counting times can impose limitations on the throughput of workers through a measurement facility, making scheduling an important issue. Procedures should be in place to assure that workers arrive for scheduled measurements and that follow-up occurs when a measurement is not completed or a worker fails to show.

Occasionally, workers are found who are claustrophobic when placed inside in vivo counter cells. Leaving the cell door partially open may help reduce some of the anxiety, but will also likely compromise the low background for which the system is designed.

Many workers want to know the results of their measurements. While a simple statement by the in vivo measurement technician may be adequate, a form letter

stating that results were normal (or showed no detection of any of the nuclides of concern) can provide permanent verification. If results are not normal, a form letter can also be used to explain what happens next.

An important aspect of any in vivo measurement program is the calibration and verification testing of the measurement equipment. In vivo measurement results are highly dependent on the determination of a background result. Likewise, calibration using known activities in appropriate phantoms is also important. Phantoms are available commercially or by loan from the USDOE Phantom Library, operated by the Radiological and Environmental Sciences Laboratory in Idaho Falls. For information on or to request loans from the USDOE Phantom Library go to the DOELAP website: http://www.hss.doe.gov/sesa/corporatesafety/doelap/index.html.

5.5.2 Urine Sampling

Urine sampling programs can be effectively administered using either workplace or home collection protocols. Workplace sampling protocols shall assure that adequate precautions are taken to prevent external contamination of the sample by levels of activity well below the detection capabilities of friskers and workplace monitors. Home collection protocols have the advantage of being sufficiently removed from the workplace to render as essentially nonexistent the potential of very low-level contamination of the sample from external sources of plutonium. Avoidance of very minor external contamination of the samples is extremely important due to the dosimetric implications of plutonium in urine.

Large-volume urine samples are necessary for bioassay monitoring due to the very small urinary excretion rates. Ideally, 24-hour total samples would be preferred; however, such samples often impose substantial inconvenience on workers, resulting in noncompliance with the instructions. As an alternative, total samples can be simulated by either time-collection protocols or volume normalization techniques.

One method of time-collection simulation (NCRP, 1987; Sula et al., 1991) is to collect all urine voided from 1 hour before going to bed at night until 1 hour after rising in the morning for two consecutive nights. This technique has been reviewed with regard to uranium by Medley et al. (1994) and found to underestimate daily urine excretion by about 14%. Such a finding is not unexpected, since the time span defined by the protocol is likely to be about 18 to 22 hours for most people.

The volume normalization technique typically normalizes whatever volume is collected to the ICRP Reference Man daily urine excretion volume of 1400 mL. Reference Woman excretion (1000 mL/d) may be used for gender-specific programs. As a matter of practicality, routine monitoring programs do not usually use gender as a basis of routine data interpretation, particularly since results are anticipated to be nondetectable under normal conditions.

A third method calls for collection of a standard volume (e.g., 1 liter) irrespective of the time over which the sample is obtained. This method uses the standard volume as a screening tool only for routine monitoring. It does not attempt to relate measured routine excretion to intake, relying on well-defined and timely supplemental special bioassay to give true or simulated daily excretion rates.

The most common sample collection containers are 1-liter polyethylene bottles. Although glass bottles are also used, they pose additional risks of breakage. Wide-mouthed bottles are preferred for convenience and sanitation. The number of bottles included in the kit should be appropriate to the protocol; for a total 24-hour protocol as much as 3 liters can be expected. Special provisions, such as a funnel or transfer cup, may improve the esthetics of sample collection and provide for added worker cooperation.

Some concerns can exist with length of sample storage before analysis. Storage may come from delays before batching samples in-house or due to transportation times to an offsite laboratory. The longer a sample stands, the more chemical and biological change it can undergo, typically manifesting itself as sedimentation and plateout on container walls. While samples can be preserved by acidification or freezing, good radiochemistry techniques should assure essentially complete recovery of any plateout or sediment. Samples sent offsite for analysis can be preserved with acid, but this method imposes hazardous material shipping requirements. Freezing samples can preserve them, but plateout and sedimentation upon thawing should still be expected.

Precautions are necessary if a lab uses an aliquot for analysis and extrapolates the aliquot result to the total sample. The aliquoting procedure should be tested using spiked samples to assure that it is representative.

A QC verification program should exist for laboratory analyses, including use of known blank samples and samples spiked with known quantities of radioactivity. Ideally, the samples should not be distinguishable by the analytical laboratory from actual worker samples. The number of QC verification samples may range from 5% to 15% of the total samples processed by a large-volume program; a small program focused on submittal of special samples following suspected intakes may have a much higher percentage of controls. An additional QC provision may be to request the analytical lab to provide results of their in-house QC results for independent review.

There are no standard or regulatory requirements for bioassay sample chain-of-custody provisions, nor has there been consensus on their need. Tampering with samples has not been a widely reported or suspected problem. Site-specific chain-of-custody requirements should be based on balancing the need with the resources required to implement them. Some sites have no chain-of-custody requirements associated with bioassay sample collection. At other sites, a simple seal placed on a sample container following collection by the subject worker is an effective means of providing a small degree of chain-of-custody. At the more complex level would be strict accountability requiring signature of issue, certification of collection, and signature of submittal.

Procedures describing details of the bioassay program should be documented. These procedures should include a description of sample collection, analysis, calibration techniques, QC, biokinetic modeling, and dose calculational methods used.

5.5.3 Fecal Sampling

A fecal sampling program shall be designed to ensure worker cooperation, whether collecting samples at home or in the workplace. Since the frequency of fecal voiding varies greatly from person to person, the sample collection program shall be adaptable. Flexibility in sample dates is important. It is suggested that when a fecal sample is required, the worker be provided with a kit and instructed to collect the sample, noting the date and time of voiding on the sample label. This practice can reduce the likelihood of unsuccessful samples. If multiple samples are required (for example, to collect the total early fecal clearance following an acute inhalation exposure), the worker may be given several kits and told to collect the next several voidings. the worker should be told to note the date and time of each sample.

Since the total fecal voiding should be collected, thought shall be given to the kit provided. Fecal sampling kits can be obtained from medical supply companies or designed by the site. A typical kit might include a large plastic zipper-closure bag to hold the sample, placed inside a 1- to 2-liter collection bucket with a tight-fitting lid. The bucket and bag can be held in place under a toilet seat by a trapezoid-shaped bracket with a hole through it sized to hold the bucket. After sample collection, the zipper bag is sealed, the lid is snapped tight on the bucket, and the bucket placed in a cardboard box.

Following collection, the sample handling, control, analytical, and quality control (QC) provisions are similar to those described above for urine samples. One particular concern for fecal analysis is the potential difficulty of dissolving class Y plutonium in the fecal matrix. While nitric acid dissolution may be adequate, enhanced digestion using hydrofluoric acid may be preferred.

5.6 MODELING THE BEHAVIOR OF PLUTONIUM IN THE BODY

A key issue to plutonium dosimetry is the modeling of how the material behaves in the body. Some of the standard models are described below, with additional discussion on the biological behavior given in Section 2.4. It is important that an internal dosimetry program establish and document the routine models and assumptions used for dosimetry. Computer codes typically incorporate standard models but may allow the flexibility to alter parameters. When altered on an individual-specific basis, the revised models need to be addressed in the pertinent case evaluations or the technical basis.

5.6.1 Respiratory Tract

The respiratory tract model of ICRP Publication 66 (ICRP, 1994a) may be used for evaluating inhalation intakes of radioactivity. The model has been widely published and internal dosimetry computer codes, hence it is not reproduced here.

Like all models, the ICRP respiratory tract model represents anticipated behavior. Once an exposure has occurred and actual data become available, deviations from the model in light of the data are appropriate.

In practice, the model has proved extremely valuable for calculating derived investigation levels and estimating intakes from bioassay data, using standard F, M,

and S absorption types of material. Model interpretation becomes more subjective when extensive data become available. Carbaugh et al. (1991) and La Bone et al. (1992) have provided excellent examples of two cases where the standard lung model assumptions did not fit the data.

Most internal dosimetry computer codes allow adjustment of particle size and selection of solubility classes. Some codes also permit detailed adjustment of the model's individual compartment parameters; with these codes, it may be possible to arrive at various subjective interpretations to explain the same data. When adjustments are made to the standard assumptions, it is important to explain what those adjustments are and why they were made.

5.6.2 Gastrointestinal Tract

The model used in ICRP Publication 68 to describe the behavior of radionuclides in the GI tract and for the calculation of doses from radionuclides in the lumen of the gut is that described in ICRP Publication 30 (1979 and 1988b). This model is also widely promulgated and used for evaluating ingestion intakes. The model is particularly subject to individual variations in fecal voiding frequency, so judgment shall be used in its application to human data.

A key parameter of the model for internal dosimetry is the f_1 factor for absorption to blood of material in the small intestine. The f_1 factor varies from 10^{-5} for plutonium oxides to 10^{-4} for plutonium nitrates and to 10^{-3} for other compounds and americium.

5.6.3 Systemic Retention and Excretion of Plutonium

Standard models for the systemic retention of plutonium are commonly used for internal dosimetry because in vivo detection of plutonium within the individual systemic compartments is not usually possible. Models proposed by the ICRP over a 10-year period are described in Section 2.4.2 of this document. Each of them has had a wide application, and ICRP has suggested that results derived using one model do not need to be rederived for compliance purposes using the newest model. Studies by the U.S. Transuranium Registry and summarized by Kathren (1994) have indicated that alternate compartments and clearance half-times may be more appropriate.

For plutonium absorbed to the blood the main sites of deposition are the liver and skeleton . ICRP Publication 78, Individual Monitoring for Internal Exposures of Workers.(ICRP, 1997) provides parameter values for biokinetic models of plutonium.

Excretion models for plutonium include the empirical models of Langham (1956) and Langham et al. (1980), Durbin (1972), Jones (1985), and Tancock and Taylor (1993), as well as study models such as Leggett (1984). This technical document does not take a position on the “best” model. Site choices of dosimetry tools such as reference tabulations (Lessard et al., 1987; ICRP, 1988a) and computer codes (such as IMBA - Integrated Modules for Bioassay Analysis), may dictate one model over another. The choice of model and explanation of its selection are among the technical bases of the site internal dosimetry program.

Note: The DOE website:
http://www.hss.energy.gov/nuclearsafety/qa/sqa/central_registry.htm
lists "toolbox" codes that are compliant with DOE's Safety Software Quality Assurance requirements. The toolbox codes are used by DOE contractors to perform calculations and to develop data used to establish the safety basis for DOE facilities and their operation, and to support the variety of safety analyses and safety evaluations developed for these facilities. IMBA is included in the DOE toolbox.

5.6.4 Natural Plutonium Balance in Man

Although plutonium can be found in members of the general public as a result of worldwide fallout from atomic weapons detonations, the levels are quite small. A summary of the literature can be found in ICRP Publication 48 (ICRP, 1986). Data from McInroy et al. (1979, 1981) suggests that median body burdens of plutonium in the U.S. population peaked at about 12 pCi during the 1960s and declined to about 2 pCi by 1977. Tissue concentration data from Nelson et al. (1993) can be used to calculate a median body burden in the early 1970s of 3 to 4 pCi.

These body burdens imply that urinary or fecal excretion associated with worldwide fallout will not be detectable by routinely available bioassay procedures. Consequently, it is reasonable to assume that any bioassay detection by a worker-monitoring problem is likely to be attributable to occupational exposure.

5.6.5 Mother-to-Fetus Transfer

Methods for evaluating embryo/fetal uptake have been described by Sikov et al. in NUREG/CR-5631 (1992) and its 1993 addendum (Sikov and Hui, 1993). For uptakes occurring during the first 2 months of pregnancy, the activity in the embryo/fetus is assumed to have the same concentration as in the mother's “other soft tissue.” For later uptakes, the embryo/fetal concentration gradually increases relative to the maternal concentration, but is assumed to remain uniformly distributed in the embryo/fetus. At 3 months, the embryo/fetal concentration is 1-1/2 times the mother's “other” soft tissues concentration. At 6 months, it is twice the mother's, and at 8 months it is thrice the maternal “other” concentration. Following transfer to the embryo/fetus, activity is assumed to remain, without clearance, until birth.

The Nuclear Regulatory Commission has developed simplified methods for assessing the gestation period dose to an embryo/fetus in Regulatory Guide 8.36 (NRC, 1992). Although the models have not been updated to reflect the 2007 amendment to 10 CFR 835 (i.e., use of dosimetric models based on ICRP Publication 60 and later publications), the Regulatory Guide is still useful in illustrating that very large maternal intakes of plutonium or americium are required to produce uptakes that would deliver 500 mrem, or even 50 mrem to the embryo/fetus. The NUREG/CR-5631 Addendum (Sikov and Hui, 1993) notes that maternal inhalation intakes of nominally 100 times the annual limit on intake (ALI) are required to give a 50-mrem embryo/fetal dose. For ingestion intakes, a 1,000 ALI maternal intake of plutonium is required to give a 50-mrem dose to the embryo/fetus. Thus, providing adequate radiation protection to limit maternal intake of plutonium and americium to the occupational limits will adequately provide for the protection of the embryo/fetus.

ANSI/HPS N13.54, Fetal Radiation Dose Calculations, (ANSI, 2008a) and ICRP Publication 88, Doses to the Embryo and Fetus from Intakes of Radionuclides by the Mother, (ICRP, 1998) provide additional guidance on assessing fetal dose. ICRP Publication 88 uses models that have been updated to reflect the 2007 amendment to 10 CFR 835.

5.7 INTERPRETATION OF BIOASSAY RESULTS

Bioassay measurements detecting plutonium or americium in workers can be initially interpreted as indicating that occupational intakes may have occurred. Standard bioassay procedures are not sufficiently sensitive to detect the worldwide environmental background levels in an in vivo or in excreta. Since most plutonium and americium bioassay measurement procedures include counting for radioactivity as the final step in the measurement process, they are subject to the statistics associated with the counting process.

Two key questions associated with bioassay data are (1) When does a sample result indicate the presence of something (i.e., when is the analyte detected)? and (2) What is the overall capability of the bioassay method for continual assurance of detection of the analyte?

The decision level, L_c (also called the critical level for detection), is the level for a given measurement that indicates the likely presence of the analyte. The L_c is dependent on the probability of obtaining false positive results (type I, or alpha, error) that is acceptable to the program. A 5% probability of false-positive results is a common design parameter of measurement programs, implying that for a large number of measurements, 5% of the time results will be indicated as positive when in fact there is no activity present. The L_c is calculated from results of analyses of blank samples. Once a measurement is performed, it is appropriate to compare it with the L_c to determine whether or not the result is "positive" (i.e., the analyte is detected).

The MDA is the level at which continued assurance of detection can be provided. The MDA is a function of the probabilities of both false positive and false negative (type II, or beta) errors and is typically based on a 5% probability for each kind of error. The MDA is also determined from analysis of blank samples, but is substantially higher than the L_c. The MDA is appropriate for use in designing bioassay programs and as the basis for estimating minimum detectable intakes and doses as indicators of program sensitivity. The MDA should not be used as a comparison with actual measurements to determine whether or not activity is present (i.e., <MDA is not an appropriate use of the concept).

Methods for calculating both L_c and MDA are given in HPS N13.30. (ANSI, 2011b).

As an alternative to the L_c and MDA of classical statistics, Miller et al. (1993) propose the use of Bayesian statistical methods for evaluating bioassay data.

General follow-up actions to abnormal bioassay measurements should include data checks, timely verification measurements, work history reviews, and performance of special in vivo measurements or excreta sample analyses for intake and dose assessments.

5.7.1 In Vivo Count Results

In vivo plutonium or americium measurements are generally relatively insensitive with regard to levels of occupational exposure concern. This applies particularly to routine chest or lung counting, skeleton counting, and liver counting. For that reason, any detection of plutonium or americium should be investigated. The investigation should address the validity of the measurement by reviewing the spectrum and its

associated background subtraction. These reviews are particularly important if the result is near the L_c. Follow-up to a positive result should include a confirming measurement. Ideally, this should be an immediate (same day) recount of equal or higher sensitivity. The farther removed in time a verification measurement is from the original measurement, the more important it becomes to factor in potential lung clearance in comparing the two measurements. A follow-up measurement taken 30 days after an initial high-routine may not be capable of providing verification if the material of concern exhibits absorption type M behavior.

Chest-wall thickness has a significant impact on chest counting. Corrections are commonly made using a height-to-weight ratio or ultrasonic methods (Kruchten and Anderson, 1990).

Corrections may be required to address apparent detection in one tissue resulting from photon crossfire from another tissue. For example, chest counting is performed primarily to estimate activity in the lung. Yet, there is substantial bone over the lungs (rib cage, sternum) and behind the lungs (vertebrae). Plutonium and americium are both bone-seeking radionuclides which will deposit on those bone surfaces and can interfere with chest counting. It is possible for a person having a systemic burden of plutonium from a wound in the finger to manifest a positive chest count from material translocated to the skeleton, axillary lymph nodes, or liver (Carbaugh et al., 1989; Graham and Kirkham, 1983; Jefferies and Gunston, 1986). Interpreting such a chest count as a lung burden can render dose estimates somewhat inaccurate.

When comparing in vivo measurements made over many years, it is important to make sure that the measurements are, in fact, comparable. One consideration is to make sure that corrections have been consistently applied to all similar measurements. It is not unusual for measurement systems to be replaced or to change the algorithms used for calculating results over time. Step changes in data can occur and should be addressed by monitoring long-term detectable trends (Carbaugh et al., 1988).

In vivo wound counting for plutonium or americium is usually one facet of special bioassay following a wound. While a portable alpha survey meter may show if surface contamination is present at the wound site or contamination of the wounding object, alpha detectors are not capable of measuring imbedded activity or activity masked by blood or serum. Thus, plutonium and americium facilities should have available a wound counter utilizing a thin sodium iodide or semi-conductor (e.g., planar germanium) detector. Such detectors are capable of measuring the low-energy photons emitted from plutonium and americium. The ability to accurately quantify wound activity is highly variable, depending on the calibration of the equipment and how deeply imbedded material is in the wound. If the object causing a wound and blood smears taken at the time of a wound show no detectable activity, then a wound count also showing no detectable activity is probably sufficient to rule out an intake. If the wounding object or the blood smears show detectable activity, special urine samples should be obtained regardless of the wound count result. In this latter circumstance, lack of detectable activity on a wound count could be attributable to deeply imbedded material at the wound site or to rapid transportation of material from the wound to the systemic compartment.

In growth of ^{241}Am from ^{241}Pu in plutonium mixtures can also significantly impact in vivo data interpretation. Rather than decreasing with time, ^{241}Am results can increase without additional intake. This circumstance is particularly likely if dealing with residual activity bound up in wound sites, but may also be observed by in vivo chest or skeleton counting. A method to evaluate ^{241}Am in growth is described is Section 5.8.4.

5.7.2 Urine Sample Results

Detection of plutonium or americium activity in a routine or special urine sample using commonly available radiochemical measurement techniques should be investigated as a potential intake. A data review should be made to assure that the sample result was correctly determined, and batch quality control sample data should be verified.

If the result is near the L_c, it is possible that statistical fluctuation of the measurement process could account for the apparent detection. Recounting the final sample preparation once or twice can be a helpful technique to verify a result or classify it as a false-positive. If the first recount also detects the analyte, it can be concluded that the sample does contain the analyte (the likelihood of two consecutive false positives at a 5% type I error per measurement is 0.0025, or 0.25 %.) If the first recount does not detect the analyte, a second recount can be performed as a tie-breaker.

An investigation should be initiated for any abnormal plutonium or americium urinalysis result. “Abnormal” for a person with no prior history of intake should be interpreted as any detectable activity.

Once an intake is confirmed, sufficient samples shall be obtained to establish a reasonably anticipated baseline against which future measurements can be compared. This is important both to provide future verification of the accuracy of the assessment and to identify potential additional intakes.

The statistical fluctuation of low-level measurements can be particularly troublesome for long-term excretion patterns. Factors of 2 can be easily expected due to day-to-day variability and imprecise adherence by the worker to urine collection protocols.

5.7.3 Fecal Sample Results

Fecal samples are much more sensitive to detection of intakes than are urine samples and, consequently, are an important part of follow-up bioassay monitoring for potential intakes initially identified by workplace indications. Pitfalls to the data interpretation include highly variable individual fecal voiding patterns, ranging from more than one per day to one every few days. This makes it extremely important to know what time interval is represented by a collected fecal sample. While normalizing a single set of fecal data to reference man daily excretion rate can be done, it is not likely to improve the quality of assessment.

The preferred fecal sampling protocol following an intake is to collect all the early fecal clearance (meaning total feces for the first five-to-seven days). This method will allow a good estimation of inhalation or ingestion intake, but does not readily permit discrimination of inhalation from ingestion, or identify whether inhaled material

exhibits absorption type F, M, or S clearance patterns. For optimum interpretation, total fecal collection should be interpreted in light of early urine and in vivo data for preliminary estimates. The urine data is likely to be particularly valuable in conjunction with fecal data to classify an intake as absorption type M or S. Longer-term follow-up fecal samples at nominally 30, 60, and 90 days post intake should substantially improve the classification of material as absorption type M or S.

Fecal sampling can also be applied to monitor excretion at long times post-intake. One caveat in such sampling is that a worker still active in a plutonium facility may be incurring very minor chronic exposure, which can significantly interfere with long-term interpretation of acute exposure data. Bihl et al. (1993) have discussed experience with a routine fecal sampling program.

5.7.4 Use of Air Sample Data in Internal Dosimetry

Results of air sampling and continuous air monitoring implying more than 40 DAC-hours exposure should be used to initiate special bioassay to assess intakes of plutonium. Although bioassay data are the preferred method for assessing intakes and internal doses, air sample data can be used for assessing internal doses if bioassay data are unavailable or determined to be inadequate or nonrepresentative. Air sample data can be used to calculate an exposure to airborne material either in terms of DAC-hours or potential radioactivity intake as follows:

$$\text{DAC - hours} = \frac{\text{Air Concentration}}{\text{DAC}} * \text{Duration (hours)} \quad (5.2)$$

DAC = The airborne concentration for radionuclides listed in Appendix A of 10 CFR 835, taking into consideration the absorption type (F/ M/ S) expressed in µCi/mL or Bq/m^3

$$\text{Intake} = \text{Air Concentration} * \text{Breathing Rate} * \text{Time} \quad (5.3)$$

Air concentration = airborne radioactivity in units of µCi/mL or Bq/m^3

If air sample results are representative of air breathed by individuals, then doses can be calculated using the 5-rem stochastic limit for CED (E_{50}) or the 50-rem deterministic limit for committed equivalent dose ($H_{T,50}$) and the respective stochastic or deterministic DAC or ALI conversion factor, as shown below:

$$E_{50} \text{or } H_{T,50} = \frac{(\#\text{ of DAC - hours})}{2000 \text{ DAC-hours}} * \text{Dose Limit} \quad (5.4)$$

$$E_{50} \text{or } H_{T,50} = \frac{\text{Intake}}{\text{ALI}} * \text{Dose Limit} \quad (5.5)$$

If respiratory protection is worn by workers, the appropriate respirator protection factor may be applied to the above calculations (i.e., dividing the calculated result by the protection factor.)

General air sampling programs should be augmented by breathing zone sampling when air concentrations to which individuals are exposed might be highly variable. Breathing zone sampling may include both fixed-location and personal (lapel) air samplers. Personal air samples are more likely to be representative of actual exposure conditions than are samples collected at fixed locations, and can be particularly useful for assessing potential intakes involving short-term exposure to well-monitored air concentrations.

5.8 DOSE ASSESSMENT

Dose assessment involves collecting and analyzing information concerning a potential intake and developing a conclusion regarding the magnitude of intake and its associated committed doses. Dose assessments are conducted by investigating the nature of a potential intake and by analyzing bioassay measurement results or other pertinent data.

Biokinetic models are used in conjunction with bioassay data to evaluate the intake, uptake, and retention of plutonium in the organs and tissues of the body. Intake estimates can then be used to calculate committed effective and organ equivalent doses. It is essential that good professional judgment be used in evaluating potential intakes and assessing internal doses. Carbaugh (1994) has identified a number of considerations for dose assessments.

Computer codes are commonly used for assessment of intakes, dose calculation, and bioassay or body content projections. La Bone (1994a) has provided an overview of what should be considered in selecting a computer code, as well as descriptions of a number of internal dosimetry codes available in 1994. Internal dosimetry code users should understand how the code works and be aware of its limitations. Computer codes merely provide the logical result of the input they are given. Use of a particular computer code does not necessarily mean a dose estimate is correct.

As used in this section, the definition of "intake" is the total quantity of radioactive material taken into the body. Not all material taken into the body is retained. For example, in an inhalation intake, the ICRP Publication 66 respiratory tract model predicts that, for 5-µm AMAD particles, 82% of the intake will be deposited in the respiratory tract; the other 18% is immediately exhaled (ICRP, 1994a). For a wound intake, material may be initially deposited at the wound site. Once the material has been deposited, it can be taken up into systemic circulation either as an instantaneous process (e.g., direct intravenous injection of a dissolved compound) or gradually (e.g., slow absorption from a wound site or the pulmonary region of the lung). Both the instantaneous and slow absorption processes are often referred to as uptake to the systemic transfer compartment (i.e., blood). Once material has been absorbed by the blood, it can be translocated to the various systemic organs and tissues.

An understanding of this terminology is important to review of historical cases. In the past sites reported internal doses as an uptake (or projected uptake) expressed as a percentage of a maximum permissible body burden. The standard tabulated values for maximum permissible body burdens were those in ICRP Publication 2 (ICRP, 1959). Many archived historical records may have used this approach. DOE Order 5480.11 (superseded), required calculation of dose equivalent. Now, 10 CFR 835 (DOE, 2011), has codified the calculation of intakes and committed doses.

5.8.1 Methods of Estimating Intake

There are several published methods for estimating intake from bioassay data (Skrable et al., 1994a; Strenge et al., 1992; ICRP, 1988b; King, 1987; Johnson and Carver, 1981). These methods each employ an idealized mathematical model of the human body showing how materials are retained in and excreted from the body over time following the intake. IRFs are used to predict the fraction of an intake that will be present in any compartment of the body, including excreta, at any time post-intake. Intake retention functions incorporate an uptake retention model that relates uptake to bioassay data and a feed model that relates intake to uptake and bioassay data. ICRP Publication 54 (ICRP, 1988a) and Lessard et al. (1987) have published compilations of IRFs. More recently, in Potter published compilations of IRFs, consistent with the 2007 amendment to 10 CFR 835 (Potter, 2002). Selected IRFs calculated consistent with the 2007 amendment to 10 CFR 835 for the urine and fecal excretion and remaining in the whole body are shown in Tables 5.6 for absorption type M and S forms of ^{239}Pu. These functions would be similar in value to those for other long-lived forms isotopes of Pu.

$$Q_t = \text{Intake} * \text{IRF}(Q_t) \tag{5.6}$$

In its simplest form, a compartment content at any time post-intake (Q_t) can be expressed as the product of intake multiplied by the intake retention function value for compartment Q at time t post-intake, or:

Results predicted by the model can then be compared with the observed bioassay data. Such results are often referred to as expectation values.

Simple algebraic manipulation of the model allows calculation of intake from the compartment content at time t, as shown below:

$$\text{Intake} = \frac{Q_t}{\text{IRF}(Q_t)}. \tag{5.7}$$

When multiple data points are available for a compartment, the intake can be estimated using an unweighted or weighted least-squares fitting procedure, as described by Skrable et al. (1994b) and Strenge et al. (1992) or as can be found in most statistics textbooks. As an alternative, data can be fit by eye to a graphical plot; however, the apparent fit can be misleading if data has been logarithmically transformed.

Intake can also be estimated from air sample data, as described in Section 5.7.4. This method is appropriate if bioassay data are not available or insufficiently sensitive. Intake estimates based on air samples and bioassay data are also appropriate as a check on each other. Valid bioassay data showing detectable results should be given preference over intake estimates based on air sample results.

Table 5.6. Intake Retention Fractions[(a)] for ^{239}Pu

	Type M Inhalation			Type S Inhalation		
Days-Post Intake	Urine	Feces	Whole Body	Urine	Feces	Whole Body
1	2.46E-04	1.10E-01	4.95E-01	2.50E-06	1.16E-01	4.90E-01
7	2.40E-05	2.29E-03	8.34E-02	3.08E-07	2.42E-03	6.25E-02
30	9.51E-06	2.81E-04	7.29E-02	1.72E-07	3.51E-04	5.06E-02
60	8.11E-06	1.31E-04	6.68E-02	1.65E-07	1.86E-04	4.29E-02
90	7.12E-06	6.65E-05	5.98E-02	1.61E-07	1.07E-04	3.87E-02
200	5.12E-06	4.67E-06	5.74E-02	1.61E-07	3.32E-05	3.25E-02
400	3.71E-06	3.71E-06	5.74E-02	1.70E-07	2.13E-05	2.74E-02
1000	2.44E-06	1.04E-06	5.44E-02	1.77E-07	1.12E-05	1.79E-02
10000	4.16E-07	2.96E-07	4.11E-02	8.25E-08	9.53E-08	5.12E-03
20000	4.83E-07	2.11E-07	3.29E-02	5.83E-08	3.20E-08	3.13E-03

(a) Incremental (i.e., sample collected in a 24-hour period ending at the time indicated) values for excreta obtained from "Intake Retention Functions Developed from Models Used in the Determination of Dose Coefficients Developed for ICRP Publication 68 – Particulate Inhalation" (Potter, 2002). See Section 5.8.1.

5.8.2 Alternate Methods of Intake Assessment

Historically, intake as described in the foregoing section was not always calculated when assessing plutonium exposures. Estimates of uptake using methods similar to Langham (1956), Healy (1957), or Lawrence (1987) focused on assessing the magnitude of radioactivity retained in the body, rather than intake (which includes material not retained and of no dosimetric significance). These methods were (and are) dosimetrically sound in so far as estimates of deposition and uptake are concerned, but do not meet the current regulatory requirement of 10 CFR 835 (DOE, 2011) to calculate intake.

5.8.3 Estimating Dose from Intakes of Plutonium

The committed equivalent dose ($H_{T,50}$) and the CED (E_{50}) resulting from an intake of plutonium may be calculated by multiplying the estimated intake (I) by either the dose conversion factor for effective dose (DCF_{eff}) or the dose conversion factor for equivalent dose (DCF_{equ}) :

$$E_{50} = I * DCF_{eff} \qquad H_{T,50} = I * DCF_{equ} \tag{5.8}$$

Dose conversion factors consistent with the 2007 amendment to 10 CFR 835 can be obtained from the ICRP Publication 68 Database (ICRP, 1994b) or calculated directly using computer programs.

Values for simplified dose conversion factors can be obtained by dividing a dose limit by the corresponding value for the ALI. A caution shall be observed with this approach: not all tabulated valued of ALIs are the same. The ALIs and DACs are commonly rounded in most tabulations to one significant figure (e.g., as in Appendix A of 10 CFR 835). Substantial variation can occur as a result of unit conversion. For example, Appendix A of 10 CFR 835 lists the DACs for ^{246}Pu absorption types M and S as both 8E-08 uCi/ml but lists the DACs in the SI units as 3E+03 Bq/m^3 for absorption type M and 2E+03 Bq/m^3 for absorption type S. Such rounding errors can introduce significant discrepancies in dosimetry calculations. This method also raises a question about which ALI or DAC should be used if compliance monitoring is being based on comparison with secondary limits, such as the ALI or DAC rather than the primary dose limits.

Where individual-specific data are available, the models should be adjusted. However, the general lack of capability to monitor organ-specific retention for plutonium (i.e., content and clearance half-times) makes the use of default models most practical.

Ideally, one should obtain as much bioassay information as possible to determine the intake and track the retention of plutonium in the body to reduce the uncertainty associated with the daily variation in the measurements. A regression analysis should be used to fit the measurement values for estimating the initial intake and clearance half-times.

5.8.4 Evaluating ^{241}Am Ingrowth in an In Vivo Count

Ingrowth of ^{241}Am from ^{241}Pu can significantly impact bioassay monitoring projections. Unless accounted for, it can lead to suspicion of new intakes, or underestimation of clearance rates. The amount of ^{241}Pu present in a plutonium mixture depends on the irradiation history and time since irradiation. Freshly processed mixtures containing 6% by weight of ^{240}Pu may contain about 0.5% by weight of ^{241}Pu and a 12% ^{240}Pu mixture may contain 3% ^{241}Pu. Commercial spent fuel can be much higher. The ingrowth of ^{241}Am occurs following a plutonium intake over a period of years. Less transportable (Material Type S) forms of plutonium may have ^{241}Am ingrowth which gradually becomes detectable. An extreme case of this was demonstrated in a well-documented Hanford plutonium-oxide exposure which exhibited a factor-of-2 increase in ^{241}Am lung content in the 3000 days following intake (Carbaugh et al., 1991). Such an increase could not be explained using the standard 500-day class Y lung clearance half-time; finally, a 17-year biological clearance half-time was estimated. The subsequent CED equivalent was estimated to be a factor of 3 higher than if the standard 500-day half-time had been used. Similar difficulties have occurred with initial detection of ^{241}Am by routine in vivo chest counting or in long-term monitoring of residual wound content.

While many available internal dosimetry computer codes will calculate the projected ^{241}Am lung content following an intake (accounting for ingrowth in the process), none of the current codes will do curve-fitting from long-term data and at the same time adjust the data for ingrowth. Therefore, the following simplistic method was developed to assess that data.

An estimate of the ^{241}Am ingrowth can be made by assuming that, at the time of intake (t = 0), all the material that will compose the long-term component is deposited in a single compartment and that the rate of transfer of material from the compartment at any subsequent time t is proportional to the quantity of material remaining in the compartment (i.e., simple exponential transport kinetics). The following equation will then describe the buildup of ^{241}Am in that compartment following an initial deposition of ^{241}Pu and ^{241}Am and a given or assumed effective clearance rate:

$$A_{t,Am} = \lambda_{r,Am} \frac{A_{O\,Pu}}{k_{e,Am} - k_{e,Pu}} \left(e^{-k_{e,Pu}t} - e^{-k_{e,Am}t}\right) + A_{O,Am}\, e^{-k_{e,Am}t} \qquad (5.9)$$

where $A_{t,Am}$ = activity of ^{241}Am at time t

$\lambda_{r,Am}$ = radiological decay constant for ^{241}Am

$A_{0,Pu}$ = activity of ^{241}Pu at time 0

$k_{e,Am}$ = effective clearance rate of ^{241}Am

$k_{e,Pu}$ = effective clearance rate of ^{241}Pu

$A_{0,Am}$ = activity of ^{241}Am at time 0

t = elapsed time

The effective clearance rate (k_e) of any nuclide is the sum of the radiological decay constant (λ_r) and the biological clearance rate (λ_{bio}). By assuming that the biological clearance rate is constant for both parent and progeny nuclides, the equation reduces to three unknowns: the initial amount of parent, the initial amount of progeny, and the biological clearance rate. These unknowns can be dealt with by assuming a standard isotopic composition at the time of intake and then solving the equation for a biological clearance rate using an iterative process until the calculated result matches the observed result at a given time t. A computer or calculator algorithm can eliminate the need for lengthy hand calculations.

Once an optimum combination of isotopic compositions and biological clearance rate is found, internal dosimetry codes or hand calculations can be used to estimate organ and effective doses. As a check on the results, standard computer codes can be used in a bioassay projection mode to project the ^{241}Am content based on the estimated intake and biological clearance rate.

5.9 INDICATOR AND ACTION LEVELS

Indicator and action levels are essential to operation of a routine internal dosimetry program. Because a wide range of levels can be defined by various facilities and organizations, this document does not attempt to prescribe particular level titles. As used in this document, indicator and action levels are simply workplace or bioassay measurements, or associated calculated doses, at which specific actions occur.

Indicator levels based on workplace indicators for reacting to a potential intake are suggested in Table 5.7. The intent of these indicator levels is to provide guidance for field response to any potential intake of radioactive material with a potential for a dose commitment that is >100-mrem CED. It is suggested that when these levels are reached, appropriate management members of the health physics and operations organizations be informed. See Section 5.4.3 for guidance on special bioassay. Table 5.8 suggests notification levels to the occupational medicine physician for possible early medical intervention in an internal contamination event. These tables, derived from Carbaugh et al. (1994), are based on general considerations and significant experience with past intakes of radioactive material and, because they are based on field measurements, do not correspond with any exact dose commitment to the worker.

The decision to administer treatment and the treatment protocol are the joint responsibilities of the physician in charge in full coordination with the patient who has been informed of the risks and benefits of any treatment being considered. The basic principle is that the proposed intervention should do more good than harm (Gerber and Thomas, 1992).

Guidelines for the medical intervention of a radionuclide intake can be found in several publications. NCRP Report No. 65 (NCRP, 1980) and the joint publication of the Commission on European Communities (CEC) and the DOE Guidebook for the Treatment of Accidental Internal Radionuclide Contamination of Workers (Gerber and Thomas, 1992) both contain detailed guidance in intervention and medical procedures useful in mitigating radiation overexposures. The ICRP recommends in Publication 60 (ICRP, 1991a) a limit of 2-rem/y (20-mSv/y) on effective dose.

Thus, the ALIs found in ICRP Publication 61 (ICRP, 1991b) and used in the CEC/DOE Guidebook noted above are those which would provide a CED of 2-rem/y instead of current U.S. regulations of 5-rem/y.

Guidance in the CEC/DOE Guidebook can be summarized as follows:

-- When the estimated intake is below one ALI, treatment should not be considered.

-- When the estimated intake is between 1 and 10 times the ALI, treatment should be considered.

Under these situations, short-term administration will usually be appropriate, except for intake of materials poorly transported from the lung (Material Type S).

Table 5.7. Suggested Plutonium or ^{241}Am Indicator Levels for Internal Dosimetry Evaluation

Indicator	Notification Level
Nasal or mouth smears	Detectable Activity
Facial Contamination (direct measurement)	200 dpm
Skin Breaks or Blood Smears	Any skin break while handling material other than sealed sources
Head, neck contamination	2,000 dpm
Contamination in a respirator	Detectable activity inside respirator after use
Hands forearms, clothing contamination [(a)]	10,000 dpm
Airborne Radioactivity	Acute intake equivalent to 40 DAC-hours after accounting for respiratory protection factor

(a) Clothing contamination levels apply to exposure without respiratory protection, such as on inner coveralls or personal clothing.

Table 5.8. Suggested Plutonium or ^{241}Am Contamination Levels for Notification of Occupational Medicine Physician

Indicator	Medical Notification Level, dpm
Nasal or mouth smears	1,000
Facial Contamination	25,000
Skin breaks or wounds	100

-- When the estimated intake exceeds 10 times the ALI, then extended or protracted treatment should be implemented, except for materials poorly transported from the lung.

-- For poorly transported material in the lung, lung lavage (i.e. internal lung washing under anesthesia) is the only recommended treatment, and it is only a consideration for intakes exceeding 100 times the ALI.

Because the dose associated with the ALI in the CEC/DOE Guidebook is 2-rem CED and because the upper administrative level recommended by the standard, Radiological Control, is 2 rem, intervention levels of 2 rem and 20 rem might be used for guidance in the manner presented in the CEC/DOE Guidebook:

-- When the CED for an estimated intake is below 2 rem, treatment is not generally recommended.

-- When the CED for an estimated intake is between 2 rem and 20 rem, treatment should be considered. Under these situations, short-term administration will usually be appropriate.

-- When the CED for an estimated intake exceeds 20 rem, then extended or protracted treatment is strongly recommended, except for poorly transported material in the lung.

Decorporation therapy should be administered immediately following any suspected intake or accidental internal contamination in excess of established action levels. The extent and magnitude of an internal plutonium contamination usually cannot be determined quickly; however, the usefulness of therapy will diminish if plutonium is allowed to translocate to bone where DTPA is ineffective. La Bone (1994b) has provided a recent approach to evaluating urine data enhanced by chelation (DTPA) therapy.

An initial prophylactic chelation therapy may be appropriate because bioassay measurements (particularly urinalysis) cannot usually be completed within the response time required for effective chelation therapy. Urinalysis becomes very helpful following administration of chelation therapy because there is a direct correlation between DTPA, urinary excretion, and dose averted because of plutonium excreted. Bihl (1994) has shown that about 2 mrem of CED is averted for every dpm of ^{239}Pu excreted. The averted dose would be less for assessment of dose in the newer quantity, per the 2007 amendment to 10 CFR 835, of CED. This is because the dose per intake is less under the newer models. For Material Type S compared to Class Y it is lower by approximately a factor of 10 for ^{239}Pu.

This provides useful information for measuring the effectiveness of DTPA therapy and determining if it is worthwhile to initiate or to continue therapy. For example, using the pre-2007 amendment to 10 CFR 835 models, if DTPA is administered when untreated excretion is 2 dpm/d, excretion should increase to 20 to 100 dpm for a dose savings of 40- to 200-mrem/d CED. However, the dose aversion would only be 4 to 20-mrem/d CED under the models required by the 2007 amendment to 10 CFR 835. Additionally, it is probable that the efficacy of treatment will decrease with continued administration as plutonium is removed from the liver and the rate of transfer into the systemic compartment decreases.

5.10 RESPONSE TO SUSPECTED INTAKES

Experience has shown that most intakes of plutonium are accidental. Plutonium facilities and operating procedures are designed to prevent intakes. Nonetheless, it is important for management to prepare for the possibility that workers might receive an intake of plutonium--even though the probability of an incident may be very small. Prompt and appropriate action following an accidental intake of plutonium will allow for therapeutic measures to be taken to minimize the internal contamination and lessen the potential for harmful effects. The health physicist and medical staff should work closely to ensure that the proper course of action is followed.

All employees suspected of having received an intake of plutonium should be referred for special bioassay measurements. Because a fraction of an intake by inhalation may be retained in the nasal passages for a few hours after exposure to airborne radioactive materials, any level of contamination on a nasal swab indicates an intake that should be followed up by a special bioassay measurement program. However, lack of detection on nasal smears cannot be taken as evidence that an intake did not occur either because the nasal passages can be expected to clear very rapidly or, alternatively, because the worker could be a mouth-breather. Special bioassay should also be initiated if plutonium contamination is found on the worker in the vicinity of nose or mouth.

For acute intakes, direct bioassay measurements should be taken before, during, and after the period of rapid clearance of activity. Urine and fecal samples collected after known or suspected inhalation incidents should also be used to estimate the magnitude of the intake. Initial assessments of intakes from contaminated wounds are based primarily on wound count and urinalysis data.

Guidance on evaluation of intakes of plutonium is found in DOE-STD-1121-2008, Internal Dosimetry (DOE, 2008d). DOE-STD-1121-2008 recognizes the difficulty in making final assessments of plutonium intakes and cautions that it "is not appropriate to place heavy reliance on the actual magnitude of the dose in the first few days following a suspected intake." Notwithstanding this difficulty, for various reasons there is a need to be able to make a timely evaluation of the potential magnitude of plutonium intakes. DOE Order 225.1B, Accident Investigations (DOE, 2011d), has a criterion for accident investigation based on a "confirmed monitoring result (workplace or individual) indicating an intake (via inhalation, ingestion, wound or absorption) of radioactive material by a general employee equivalent to 2 or more times the annual limit on intake." The Order also states that "Confirmation must be made within 3 working days following identification of monitoring results (workplace or individual monitoring) indicating an exposure exceeding one or more of the criteria in this section."

If a significant intake is indicated, the worker should not return to further potential exposure to plutonium until the intake has been thoroughly assessed and a predictable bioassay pattern established. This is particularly important because a new intake of a very low level may confound the interpretation of bioassay measurements for previous intakes of plutonium.

The health physicist shall make important decisions for prompt action at the site of an accidental or suspected intake of plutonium or other radioactive materials. Often, these

decisions shall be based on limited data. Information that may be available for initially estimating the amount and type of intake may include the following:

-- levels of measured contamination in the work area

-- skin contamination levels, affected areas, and whether the skin is damaged or punctured

-- wound contamination levels

-- chemical form of the material involved

-- results of air monitoring

-- nasal smear activity levels

-- sputum and/or mouth contamination.

The special bioassay monitoring program is initiated following a known or suspected intake. This information is needed for dose assessment and future exposure management. The intake is confirmed if follow-up bioassay measurements indicate positive measurement results. Additional bioassay measurements may be needed to quantify the intake and provide data for determining the effective dose. The frequency of bioassay monitoring will depend on the specific case to be evaluated. Selection of the appropriate sampling frequency is based on the previously discussed performance capabilities for workplace monitoring program, consultations with internal dosimetry specialists, and the cooperation of the affected employee.

5.10.1 Planning

The management at the plutonium facility should be prepared to follow an emergency action plan for response to a plutonium intake. If a worker accidentally inhales or ingests plutonium or is injured by a plutonium-contaminated object, the action plan should be initiated immediately. A rapid response is important because any delay in implementing appropriate action could lessen the effectiveness of decorporation therapy and increase the probability for internalized plutonium to deposit on bone surfaces.

5.10.2 Medical Response Plan

The health physicist and medical staff shall establish an emergency action plan for the appropriate management of an accidental intake of plutonium. The elements of the plan should include the following:

-- Decision levels for determining when monitoring data or accident events require emergency medical response

-- responsibilities of the affected worker, health physicist, medical staff, and management or supervisory personnel

-- instructions for immediate medical care, decontamination, monitoring, and longer-term follow-up response

-- provisions for periodically reviewing, updating, and rehearsing the emergency action plan.

The sequence and priority of the emergency action plan may vary with the magnitude and type of accidental conditions and their severity. An initial early assessment of the incident should focus, first, on treatment of life-threatening physical injuries and, second, on the radioactive contamination involved. Minor injuries should be treated after decontamination.

A rapid estimate of the amount of internal contamination by plutonium or other alpha-emitters may not be possible. If a significant intake (meaning one that exceeds 10 times the ALI) is suspected, medical staff should proceed with decorporation therapy after first treating major injuries.

5.10.3 Responsibilities for Management of Internal Contamination

Responsibilities should be assigned for action in response to an accidental internal plutonium contamination. The affected worker has the responsibility to inform the health physicist, RCT, or his immediate supervisor as soon as an intake is suspected. (More broadly, all radiological workers have the responsibility to report conditions that could lead to an intake to their immediate supervisor and/or the health physics organization.) The health physicist or RCT should make an initial survey of the extent of the contamination and immediately contact his supervisor and, when action levels are exceeded, contact a member of the medical staff. He should continue to provide monitoring and radiation safety support to the medical staff and supervisors during the management of the contamination incident. Care should be taken to limit the spread of radioactive contamination.

The health physicist should immediately begin to gather data on the time and extent of the incident. Contamination survey results should be recorded. Radionuclide identity, chemical form, and solubility classification should be determined. Nasal smears should be obtained immediately if an intake by inhalation is suspected. When action levels are exceeded, all urine and feces should be collected and labeled for analysis. Decontamination should proceed with the assistance of the medical staff. Contaminated clothing and other objects should be saved for later analysis.

5.10.4 Immediate Medical Care

The medical staff should provide immediate emergency medical care for serious injuries to preserve the life and well-being of the affected worker. Minor injuries may await medical treatment until after an initial radiation survey is completed and the spread of contamination is controlled. However, the individual should be removed from the contaminated area as soon as possible. Chemical contamination and acids should be washed immediately from the skin to prevent serious burns and reactions.

Chelation

Chelation therapy, or chelation, is the process of removing unwanted metals from the body by administering an agent that binds to the metal and promotes its excretion. It is important to remove plutonium from the body because it is retained in the bones and liver for many years. Plutonium remaining in the body continues to irradiate nearby tissues. This results in increased risk of cancer. For over 60 years, chelation therapy has been practiced successfully and safely in treating lead and other heavy metal poisoning.

Chelating agents can be administered orally, intravenously, or as a mist, depending on the agent and the type of poisoning. Several chelating agents are available; each has different affinities for different metals. DTPA (diethylenetriaminepentaaceticacid) has been proven effective for the treatment of people accidentally contaminated internally with the transuranic nuclides plutonium, americium, and curium. Recently, based on additional clinical data and peer-reviewed articles, the Food and Drug Administration (FDA) has approved DTPA as a safe and effective compound to enhance elimination/excretion of radioactive materials from the body. There are two primary DTPA compounds: Ca-DTPA and Zn-DTPA. Ca-DTPA is more effective than Zn-DTPA in the first 24 hours after contamination. To avoid long-term depletion of essential metals, Ca-DTPA is administered initially, followed by Zn-DTPA if multiple doses are required.

The number of treatments is based on the results of the bioassay analyses. Most situations involve single treatments; however, a 2010 wound incident at a DOE facility involved 71 treatments. Possible side effects of such an extended chelation therapy regiment could include depletion of essential elements, which can be treated by administering supplemental minerals.

In addition, according to the Centers for Disease Control and Prevention (CDC) Web site, people who are given repeat doses of Ca-DTPA within a short period of time may have nausea, vomiting, diarrhea, chills, fever, itching, and muscle cramps. Other side effects may include headache, lightheadedness, chest pain, and a metallic taste in the mouth. Chelation therapy administered by nebulized inhalation may cause breathing difficulties in some individuals.

According to the FDA Web site at: **http://www.fda.gov/Drugs/EmergencyPreparedness/BioterrorismandDrugPreparedness/ucm130314.htm:**

- If Ca-DTPA is not available, or treatment cannot be started within the first 24 hours after contamination, treatment should begin with Zn-DTPA.
- If Zn-DTPA is not available, Ca-DTPA can be given for continued treatment, along with vitamin or mineral supplements that contain zinc.
- Ca-DTPA and Zn-DTPA can be administered by nebulizer or directly into the blood stream (i.e., intravenously). If the route of internal contamination is through inhalation alone, then nebulized chelation therapy will suffice. If the routes of contamination are multiple (e.g., inhalation and through wounds), then intravenous chelation therapy is preferred.
- The duration of treatment is dictated by the level of internal contamination and the individual's response to therapy. Levels of internal contamination should be

ascertained weekly during chelation therapy to determine when to terminate treatment.
- Zn-DTPA is the preferred treatment for the pregnant woman with internal contamination.
- FDA recommends nebulized DTPA for patients whose internal contamination is only by inhalation.
- The safety and effectiveness of the intramuscular route has not been established for
Ca-DTPA or Zn-DTPA.
- The duration of Ca-DTPA and Zn-DTPA therapy depends on the amount of internal radioactive contamination and the individual's response to therapy.
- Ca-DTPA should be used with caution in patients suffering from a severe form of a disease called hemochromatosis.

Additional information is on the CDC Web site at:
http://www.bt.cdc.gov/radiation/dtpa.asp.

Some of this information includes:

- Radioactive materials chelated to DTPA are excreted from the body in the urine; therefore, DTPA shall be used carefully in people whose kidneys do not function properly.
- Breathing treatments using DTPA may not be safe for some people with asthma. If a person with asthma requires treatment with DTPA, the drug should be injected.
- DTPA should not be used to treat people who are internally contaminated with the radioactive materials uranium or neptunium.

The big advantage of chelation for radioactive metals, such as plutonium, is the radiation dose reduction for the patient. Substantial dose reductions can be achieved if DTPA is administered within a few hours (recommended within 1 hour) of the intake of plutonium. Dose reductions from 10 percent to 90 percent have been achieved for contaminated wound or burn cases and up to 30 percent for inhalation cases.

The decision to administer chelation therapy is made by the worker in consultation with a board-certified occupational medicine physician. In communicating information to the individual concerning the risks and benefits of chelation refer to the Health Physics Society policy paper which discusses providing individual risk estimates. The position paper states, in part: "*the Health Physics Society recommends against quantitative estimation of health risks below an individual dose of 5 rem in one year or a lifetime dose of 10 rem above that received from natural sources*". More information on this position paper is found at:
http://hps.org/documents/risk_ps010-2.pdf

Chelation is generally recommended when the estimated dose exceeds 2 rem CED. If a quick dose estimate cannot be made, indicators such as airborne radioactivity exposure, nasal/mouth smears, facial contamination, skin breaks, or bioassay measurements are used. Chest or whole-body counts and wound counts are used as well.

References and Web sites for this subject, in addition to the ones listed previously, include NCRP Report Number 161, Management of Persons Contaminated with Radionuclides (NCRP, 2008), and the Radiation Emergency Assistance Center/Training Site Web site at: http://orise.orau.gov/reacts/guide/internal.htm.

5.10.5 Contaminated Wounds

Medical treatment for contaminated wounds may include flushing with saline and decorporating solutions, debridement, and surgical excision of the wound. These measures are all the responsibility of trained medical staff operating under the direction of a physician. Health physics personnel can provide valuable assistance by prompt assessment of materials removed from the wound and identification of magnitude of residual activity as decontamination proceeds. Decontamination should continue until all radioactivity has been removed or until risk of permanent physical impairment is reached.

NCRP Report Number 156, Development of a Biokinetic Model for Radionuclide-Contaminated Wounds and Procedures for Their Assessment, Dosimetry and Treatment, provides additional information on evaluation of contaminated wounds (NCRP, 2006).

The Radiation Emergency Assistance Center/Training Site Web site provides dose coefficients for 38 radionuclides based on NCRP wound model and ICRP biokinetic models:

Dose Coefficients for Intakes of Radionuclides via Contaminated Wounds

6.0 EXTERNAL DOSE CONTROL

The purpose of an external dose control program is to protect the individual radiation worker by minimizing dose to levels as low as reasonably achievable (ALARA) and preventing exposures above prescribed limits. This also implies minimizing the collective dose by summing all the individual TEDs in a specified population. This section discusses methods to minimize exposures by characterizing the radiations emitted by plutonium and effective methods to shield or otherwise reduce exposures.

The Department of Energy provides a detailed explanation of the recommendations for external dosimetry in Chapter 6 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a). Specific applicable documents for external dosimetry are listed in the reference list of that Implementation Guide. Because the requirements and recommendations are explicitly given in these documents, they will not be discussed in any great detail in this section. Rather, the emphasis will be given to items that are unique to plutonium facilities and the radiological aspects for safe handling of plutonium.

Measuring the external radiation exposure and the resultant dose for personnel handling plutonium is a difficult task because of the many radiations involved. Examples of the radioactive decay schemes and radiations emitted were presented in Section 2.0 for the various plutonium isotopes and radioactive progeny. Plutonium has a wide distribution of gamma energies; literally hundreds of different photon energies are present. Fortunately, plutonium emits few high-energy photons, so photon dose rates are low. But plutonium also emits highly penetrating neutrons from spontaneous fission and alpha-neutron reactions from compounds and alloys.

In the past, most of the dose in plutonium facilities was the result of plutonium production and fabrication operations. Most of these operations involved physical contact with freshly separated plutonium in glove boxes during fabrication and assembly operations. With the reduction in weapons production, emphasis has shifted to dismantlement and storage operations and to D&D of plutonium facilities. Much of the material in these facilities is low-exposure plutonium containing 6% ^{240}Pu that is at least 20-years-old, so a significant fraction of the ^{241}Pu has decayed into ^{241}Am. The radioactive progeny have increased gamma dose rates, making dismantlement of plutonium facilities more difficult. Although many of the examples in this section involve higher-exposure plutonium, it is expected that most dosage in plutonium facilities will originate from clean-up and storage of weapons-grade plutonium.

6.1 DOSE LIMITS

Limits of interest used for control of external radiations are specified at various depths by 10 CFR 835 (DOE, 2011) as well as the ICRP and the NCRP). The limits are given in Table 6.1 for the appropriate depths in tissue for the whole body, lens of the eye, skin and extremities.

Table 6.1. Effective Depth of Tissue for Various Organs

	Depth of Tissue mg/cm^2.
Equivalent dose to the whole body	1000
Equivalent dose to the lens of eye	300
Equivalent dose to the extremity and skin	7

6.1.1 Limiting Quantities

Recently, DOE has made significant changes in the methodology used for radiation protection. Previously, DOE used the concept of *dose equivalent*. For whole body irradiations, dose equivalent was the product of absorbed dose multiplied by the quality factor, which was evaluated by Monte Carlo calculations in a cylindrical phantom of 30-cm diameter and 60-cm height. For monoenergetic neutrons or photons normally incident on the phantom model, the dose equivalent was the highest value calculated anywhere in the phantom. Dose equivalent was non-additive because the maximum values occur at different depths in the phantom for different energies. A detailed explanation of the calculations can be found in an article by Auxier et al. (1968).

ICRP Publication 60 used revised terms for *stochastic* and *nonstochastic* for radiation effects (i.e., *stochastic* and *deterministic*) and set limits for both types of effect. Stochastic effects are defined as those for which the probability of the effect occurring (as opposed to the degree or severity of effect) is a function of radiation dose. Deterministic effects were defined as those for which the severity of the effect is a function of the dose; a threshold may exist. Limits were established such that the risk of stochastic effects occurring was equivalent to about the same risks faced by workers in "safe" industries who were not occupationally exposed to radiation in the workplace. Limits were also established for deterministic effects that prevented these effects from occurring even if the exposure occurred at the annual limit over the lifetime of the worker.

The ICRP specified in Publication 60 that radiation exposure be limited by the *effective dose*, E, which can be expressed by the relation:

$$E = \sum w_T D_{T,R} w_R \qquad (6.10)$$

where

w_T = tissue weighing factor for the relevant organ or tissue T
D_{TR} = average absorbed dose in the tissue or organ of interest
w_R = radiation weighting factor for the type of radiation R

The weighing factors are given in Table 6.2, which is taken from 10 CFR 835 (DOE, 2011). Effective dose has the benefit that it is additive, and internal and external radiations can be added numerically to drive an overall estimate of risk.

Table 6.2. Tissue Weighing Factors

Organs or tissues, T	Tissue weighting factor, w_T
Gonads	0.20
Red bone marrow	0.12
Colon	0.12
Lungs	0.12
Stomach	0.12
Bladder	0.05
Breast	0.05
Liver	0.05
Esophagus	0.05
Thyroid	0.05
Skin	0.01
Bone surfaces	0.01
Remainder[1]	0.05
Whole body[2]	1.00

[1] "Remainder" means the following additional tissues and organs and their masses, in grams, following parenthetically: adrenals (14), brain (1400), extrathoracic airways (15), small intestine (640), kidneys (310), muscle (28,000), pancreas (100), spleen (180), thymus (20), and uterus (80). The equivalent dose to the remainder tissues ($H_{remainder}$), is normally calculated as the mass-weighted mean dose to the preceding ten organs and tissues. In those cases in which the most highly irradiated remainder tissue or organ receives the highest equivalent dose of all the organs, a weighting factor of 0.025 (half of remainder) is applied to that tissue or organ and 0.025 (half of remainder) to

the mass-weighted equivalent dose in the rest of the remainder tissues and organs to give the remainder equivalent dose.

[2] For the case of uniform external irradiation of the whole body, a tissue weighting factor (w_T) equal to 1 may be used in determination of the effective dose.

The methodology of ICRP Publication 60 (ICRP, 1991a) has been incorporated into 10 CFR 835 (DOE, 2011). The annual radiation dose limits for DOE and its contractors are presented in Table 6.3.

However, DOE contractors usually establish lower annual administrative control levels, typically 500 mrem/year.

In practice, it is very difficult to measure the effective doses specified in Table 6.3 because it is necessary to know not only the type of radiation but also its energy and direction. If the flux, energy, and direction of incidence are known, it is possible to calculate effective dose using fluence to effective dose conversion coefficients, which present the effective dose as a function of energy for various irradiation geometries. Conversion coefficients for photons in various irradiation geometries, including planar sources, can be found in a report by the Zankl et al. (1994). This will provide more accurate values of effective dose as opposed to numerically setting the value of effective dose equal to equivalent dose.

Table 6.3. Radiation Dose Limits for DOE and DOE Contractors

Type of Radiation Exposure	Annual Limit
Occupational Exposures	
Stochastic Effects	5-rem total effective dose from external sources and the committed effective dose from intakes received during the year
Deterministic Effects	
Lens of eye	15-rem equivalent dose
Extremity	50-rem equivalent dose
Skin	50-rem equivalent dose
Individual organ tissue	50-rem equivalent dose
Embryo/fetus of a Declared Pregnant Worker	
Gestation period	0.5-rem equivalent dose
Planned Special Exposure	
Event plus Annual Occupational exposure	5-rem total effective dose (TED)
Minors	0.1-rem TED

6.1.2 Operational Quantities

Because of the difficulties in determining effective dose from direct measurements, the concept of *operational quantities* has been introduced to be more closely related to measurable quantities. Operational quantities include *ambient dose equivalent* used for area monitoring and *personal dose equivalent*

used for personnel dosimetry. Operational quantities should be a conservative estimator of effective dose, i.e., the values of the operational quantities should be equal or greater than the effective dose specified for the limiting quantities.

The ambient dose equivalent, $H^*(d)$, is the dose equivalent at a depth, d, in a 30-cm-diameter sphere of tissue, where a) the radiation field has the same fluence and energy distribution as the point of reference for the measurement and b) the fluence is unidirectional (i.e., the sphere can be viewed as being in an aligned radiation field). Most survey instruments are designed to measure ambient dose equivalent, and international standards are based on the ambient dose equivalent concept. The depth of interest is 1 cm of soft tissue, as specified in 10 CFR 835.2.

The personal dose equivalent, $H_p(d)$, is the dose equivalent in soft tissue at the appropriate depth, d, below a specified point on the body. Provided that personal dose equivalent is a conservative estimator of effective dose, personnel dosimeters should be calibrated in terms of personal dose equivalent. Otherwise a correction factor should be applied.

In reality, most instruments and personnel dosimeters used at DOE facilities are still calibrated in terms of dose equivalent. For example, consider the case in which personnel neutron dosimeters are calibrated on acrylic plastic phantoms at a specified distance from a calibrated neutron source. For Department of Energy Laboratory Accreditation Program for personnel dosimetry (DOELAP) testing, the dose equivalent is assessed in accordance with U. S. Department of Energy Laboratory Acceditation Program for Personnel Dosimatry, DOE STD-1095-2018 (DOE 2018a) which incorporates ANSI/HPS N13.11, Personnel Dosimetry Performance-Criteria for Testing, ANSI/HPS N13.11 (ANSI, 2009).

In most instances, the present methods based on dose equivalent over-estimate effective dose. In cases where personnel are approaching dose limits, it may be prudent to more accurately evaluate effective dose using special calibrations.

6.2 RADIATIONS IN PLUTONIUM FACILITIES

As outlined in Section 2.0 of this report, plutonium emits a wide variety of radiations, including alpha and beta particles, as well as more penetrating X-rays and gamma rays. Because of the short half-life of ^{241}Pu, the radioactive decay progeny are also important sources of radiation. This section outlines methods to calculate doses from radiations emitted by plutonium and its progeny. Examples of measured dose rates are also included.

6.2.1 Alpha and Beta Doses

Plutonium is primarily an alpha-emitter and is of great concern if inhaled, ingested or injected into the body. However, the skin is an effective barrier to alpha particles, and external contamination is only a problem if there is a wound or break in the skin.

Plutonium-241 is a beta-emitter that produces low-energy beta particles with a maximum energy of 0.022 MeV. Both alpha and beta particles are completely shielded by thin rubber gloves or other protective devices. The dose rate through

a rubber glove originates primarily from the X-rays and low-energy photons generated from plutonium and ^{241}Am, the decay progeny of ^{241}Pu.

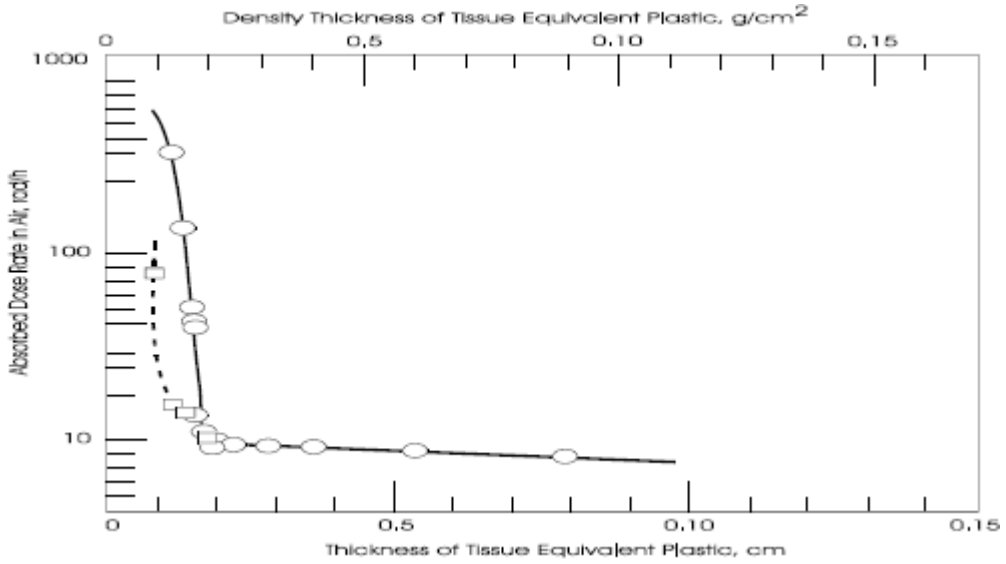

Figure 6.1. Absorbed Surface Dose Rate from Plutonium Dioxide as Measured with an Extrapolation Chamber

Figure 6.1 shows the dose rate as a function of tissue equivalent plastic absorber thickness, as measured by an extrapolation chamber in contact with a 3-in.-diameter plutonium dioxide source coated with a very thin layer of beryllium for contamination control. The plutonium was compressed to about 80% of its theoretical density and vitrified by a Dynapack process in which powder was compressed into a glassy solid by extreme pressure and heat evolved during the compression process. The plutonium oxide disk is mechanically stable and produces little smearable contamination. Even minute layers of tissue equivalent plastic reduce the dose rate significantly, as shown in Figure 6.1.

6.2.2 Gamma Doses

There can be substantial gamma doses involved in the processing and handling of plutonium, particularly in glove-box operations involving plutonium dioxide powders. Plutonium emits very few highly penetrating gamma rays; most of the radiations are L X-rays, which are very easily shielded. Because most of the photons emitted by plutonium are of low energy, plutonium sources are "infinitely thick" relative to their photon radiations, i.e., an additional thickness of plutonium does not appreciably increase the photon dose rate. A plutonium metal source of about 1-mm thickness or a plutonium oxide source about 6-mm-thick is "infinitely thick" due to self-shielding.

The age and isotopic composition are very important in determining the dose rate from plutonium because of the ingrowth of ^{241}Am from the decay of ^{241}Pu, which has a half-life of only 15 years. (The growth of plutonium daughters was discussed in detail in Section 2.1.1.) Old plutonium processing facilities can have high gamma dose rates, particularly from nearly invisible dust layers containing ^{241}Am, which has a 37% probability of emitting a 60-keV photon per alpha disintegration. A surprising amount of plutonium oxide powder can be found in dust layers on the interior surfaces of glove boxes because of the very high density of plutonium. For example, a 0.001-in.-thick layer of plutonium oxide dust on the 4-ft by 8-ft floor of a glove box can contain almost 200 grams of plutonium. Even though a glove box has additional iron or lead shielding, high gamma dose rates can persist because of the photons emitted by dust layers on the surface of gloves. Covers shall be placed over glove ports to reduce gamma dose rates around plutonium processing lines.

Doses to the extremities are usually dominated by gamma rays in typical glove-box operations. Extremity dosimeters shall be used by all personnel who perform hand contact operations with plutonium or who are involved in the manual decommissioning of plutonium facilities. Extremities are defined as the hands and forearms below the elbows and the feet and legs below the knees. In a plutonium facility, the contact doses to the hands and forearms are the most limiting cases. The extremity dose is more limiting than a whole body dose if the dose gradient is greater than 10:1 over a distance of 1 meter, the maximum distance from the fingers to the trunk of the body. In most cases, the source is not at arm's length and the dose gradient needs to be 10:1 or 20:1 for the extremity dose to be limiting (NUREG/CR-4297, Reece et al., 1985). But in highly shielded glove boxes, it is possible to have very high extremity doses from dust layers on gloves; the dose to the torso can be much lower because of shielding applied to the glove box.

6.2.2.1 Measured Gamma Dose Rates

There is a considerable amount of experimental data for measured photon dose rates from plutonium glove-box operations as recorded in progress reports issued by the Hanford Engineering Development Laboratory from the Personal Dosimetry and Shielding Program. For example, the photon dose rates were measured on an anthropomorphic Remab arm phantom inserted into gloves in a plutonium glove box. The arm phantom contains a human skeleton surrounded by tissue equivalent fluid inside a molded plastic “skin.” Thermoluminescent dosimeters (TLD-700s) were positioned at various locations along the surface of the arm phantom and inside tubes inserted into the bones.

Measurements were first made in a “clean” glove box before it was placed into service. The arm phantom was placed inside the glove and positioned in contact with a 1-quart steel can (nominal wall thickness of 10 mil or 0.25 mm), containing 1 kg of plutonium dioxide with the isotopic composition shown in Table 6.4. Measurements were made at the various locations with the arm phantom inside 20-mil Neoprene gloves (average thickness 0.021 in., 0.53 mm) and inside 37-mil (0.94-mm) lead-loaded Neoprene gloves.

The data shown in Table 6.5 are the average dose rates measured by three TLD-700s with the indicated one standard deviation in the measured values. As one would expect, the palm and fingers had the highest dose rates, approximately 300 mrad/h; the lowest dose rates of 1 mrad/h were measured at the top of the arm. Because the plutonium was "infinitely thick" and lower-energy photons were removed by the shielding provided by the steel can, the dose rates in the lead-loaded glove were only slightly lower than those in the Neoprene glove. The can of plutonium was removed, and the gloves dusted with high-exposure plutonium with an isotopic composition similar to that given in Table 6.4. The arm phantom was inserted into 20-mil Neoprene and 37-mil lead-loaded Neoprene gloves; the dose rates measured with TLDs are shown in Table 6.6.

Table 6.4. Isotopic Composition of the Plutonium Used in the Extremity Dosimetry Measurements

Isotope	Weight Percent
^{236}Pu	0.000003
^{238}Pu	0.58
^{239}Pu	72.1
^{240}Pu	19.15
^{241}Pu	6.29
^{242}Pu	1.88
^{241}Am	0.02

As expected, the highest dose rates were recorded on the hand, wrist and forearm, where the most PuO_2 dust had accumulated, and the lowest dose rates were on the upper arm and humerus. For thin dust layers, the dose rates inside the lead-loaded glove were generally much lower, typically a factor of 4 to 5 times less than the dose rates inside the Neoprene glove. The lead-loaded glove provided significantly better shielding for the 60-KeV photons from ^{241}Am and the L x-rays from plutonium, which were responsible for much of the dose. In these examples, the dose rates from the contaminated glove were about 10% of those from the 1 kg of plutonium dioxide inside the steel can. Additional experiments with 25% PuO_2- 75% normal UO_2 showed that dose rates increased as dust loadings increased with use; the dose rates on the hand and forearm increased to levels of about 30 mrem/h to 20 mrem/h, respectively.

Table 6.5. Gamma Dose Rates Along an Arm Phantom in Contact with a Steel Can Containing 1 kg of Plutonium Dioxide in an Uncontaminated Glove Box

Position	Gamma Dose Rates, mrad/h	
	Neoprene Glove	Lead-Loaded Glove
Ring Finger	330 ± 6	272 ± 25
Palm	292 ± 9	220 ± 16
Back of Hand	72 ± 2	65 ± 1
Wrist		
Inside	84 ± 6	56 ± 5
Outside	31 ± 1	24 ± 1
Forearm		
Inside	22 ± 0.4	12 ± 1
Outside	4.4 ± 0.1	3.8 ± 0.4
Elbow		
Inside	4.8 ± 0.1	2.6 ± 0.2
Outside	1.4 ± 0.1	1.8 ± 0.4
Front	2.9 ± 0.2	2.1 ± 0.1
Bottom of humerus	2.2 ± 0.1	2.5 ± 0.5
Lower mid-arm	7.1 ± 0.1	3.9 ± 0.3
Lower mid-humerus	3.8 ± 0.1	2.3 ± 0.2
Upper mid-arm	2.4 ± 0.1	2.5 ± 0.2
Upper mid-humerus	1.8 ± 0.1	1.8 ± 0.2
Top of arm	0.9 ± 0.03	2.2 ± 0.8
Top of humerus	1.1 ± 0.2	1.3 ± 0.1

The gamma energy spectra from plutonium sources are highly variable, depending on the amount of shielding present, including self-shielding. Small lightly shielded sources, such as dust layers on the interior of glove boxes, are dominated by L X-rays and the 60-keV photons from ^{241}Am, the decay progeny of ^{241}Pu.

But the gamma energy spectra are quite different in storage vaults and other facilities where the plutonium is encapsulated. In those cases, the low-energy photons have been shielded out, and the spectrum is dominated by higher photon energies. Note that plutonium metal buttons or cans of plutonium oxide prepared for storage are self-shielded, and high-energy photons from decay progeny such as ^{237}U become increasingly important.

Table 6.6. Gamma Dose Rates Measured with an Arm Phantom Placed Inside Gloves Dusted with Plutonium Dioxide Powder

Position	Gamma Dose Rates, mrad/h	
	Neoprene Glove	Lead-Loaded Glove
Palm	10.0 ± 0.4	9.5 ± 16
Back of Hand	21.8 ± 1.3	5.4 ± 0.3
Wrist		
Inside	22.6 ± 0.7	9.0 ± 0.6
Outside	22.5 ± 0.6	5.8 ± 0.4
Forearm		
Inside	34.5 ± 0.2	6.7 ± 0.4
Outside	16.7 ± 0.2	3.6 ± 0.6
Elbow		
Inside	17.5 ± 0.4	5.3 ± 0.4
Outside	11.4 ± 0.1	3.7 ± 0.4
Front	---	4.4 ± 0.3
Bottom of humerus	3.5 ± 0.2	3.6 ± 0.4
Lower mid-arm	6.7 ± 0.5	3.4 ± 0.2
Lower mid-humerus	2.1 ± 0.2	2.9 ± 0.3
Upper mid-arm	4.6 ± 0.1	2.3 ± 0.4
Upper mid-humerus	1.0 ± 0.1	1.4 ± 0.3
Top of arm	0.8 ± 0.2	3.7 ± 0.3
Top of humerus	---	1.1 + 0.3

6.2.2.2 Calculated Photon Dose Rates

It is very difficult to accurately calculate dose rates from plutonium because of the wide range of photon energies and the relatively low abundance of photons. Most of the photons are of relatively low energies, usually below 425 keV, which are easily shielded. For heavily shielded spectra, the high-energy photons from decay progeny become very important, as well as the high-energy photons from plutonium, which have very low abundances.

For this reason, there are only a few computer codes that give accurate dose rates for plutonium. Many computer codes do not calculate the photons from progeny from radioactive decay. Others do not include the high-energy photons which have very low abundances, but which become very important for massive shields. One shall check the photon libraries to make certain that the higher-energy photons are included. Also, many point kernel codes may not give accurate results for thin shields because low-energy build-up factors are not very accurate.

There are only a few codes specifically designed for plutonium dose calculations in the Radiation Safety Information Computational Center (RSIC)[1]; they include the following:

-- PUSHLD - Calculation of Gamma Radiation Dose Rates from Three- Dimensional Plutonium Sources and Shield Geometries at Various Distances, HEDL-TME 73-89, Hanford Engineering Development Laboratory (Strode, 1974).

-- BMC-MG - A Multigroup Monte Carlo Kernel Integration Neutron and Gamma-Ray Shielding Code System for Plutonium, BNWL-1855, Pacific Northwest Laboratory (Zimmerman, 1975).

-- PURSE - A Plutonium Radiation Source Code, PNCT 852-78-13, Japan Power Reactor and Nuclear Fuel Development Corp., Tokai-Mura, Japan.

The PUSHLD computer code has the advantage that the calculated results were experimentally verified to make certain that the low-energy build-up factors were correct. There are undoubtedly several other codes that could give accurate dose rates from plutonium, particularly if a radioactive decay code is used to calculate the amount of progeny as a function of time.

There are some empirical equations that can be used to calculate dose rate through simple shields, such as Neoprene, when plutonium is directly handled in a glove box. Because of the dominance of low-energy X-rays, the surface dose rates from plutonium sources can be quite high. Roesch and Faust have derived a formula for predicting the surface dose rate from plutonium through a 100-mg/cm^2 shield:

$$D_s(\text{rad/h}) = 171\ f_{238} + 0.51\ f_{239} + 2.4\ f_{240} + 8.7\ f_{241} + 0.15\ f_{242}\,(0.074\ f_{241})t \qquad (6.2)$$

where D_s = the surface dose rate of plutonium metal or oxide, rad/h

(1) RSICC, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831-6171, Telephone (865)-574-6176.

f_i = the weight fraction of the ith isotope of plutonium

t = the time since chemical separation of the plutonium, days.

This equation is only valid for a year or so after chemical separation, when the ingrowth of ^{241}Am can be represented linearly.

A similar equation has been derived for lead-loaded rubber gloves using the calculations from the computer code PUSHLD. The 80-mil lead-loaded glove is nominally 1.9-mm (0.076-in.) thick in the palm and forearm and contains the equivalent of about 1 mm of lead. The surface dose rate, D_{PbGl}, is given by the following equation:

$$D_{PbGl}(t) = 2.83\ f_{238}\ e^{-0.00789t} + 0.104\ f_{239} + 0.0315\ f_{240} + 6.35 \times 10^{-5}\ f_{242} + f_{241}\ (158.5\ e^{-0.0016t} - 152.5\ e^{-0.0457t}) \quad (6.3)$$

where $D_{pb}Gl(t)$ = surface dose rate as a function of time, rad/h

f_i = weight fraction of the ith plutonium isotope

t = time since chemical separation of the plutonium, years.

This equation includes the radiations from plutonium, as well as the ^{237}U and ^{241}Am progeny from the decay of ^{241}Pu. The expression is valid for times between 50 days and 100 years after the chemical separation of the plutonium. The formula predicts dose rates from 0% to +20% of those calculated by the computer code PUSHLD.

6.2.3 Neutron Doses

Neutron doses are significant in any process or decommissioning efforts involving kilogram quantities of plutonium or gram quantities of ^{238}Pu. Neutrons originate from three sources:

-- Spontaneous fission of even isotopes of plutonium

-- alpha-neutron reactions with low-atomic-number elements, including oxygen and fluorine in plutonium compounds and impurities in metals

-- neutron-induced fissions.

Experience has shown that only spontaneous fission and alpha-neutron reactions are important. Because of strict criticality controls, most forms of plutonium have very little neutron-induced multiplication. Induced fission seems to be a problem only in metal (1 kg or more) or in very large, high-density arrays of plutonium oxide with an additional moderator.

Plutonium-238 used for heat sources deserves special attention. Even sub-gram quantities of ^{238}Pu produce appreciable neutron doses because of the extremely high spontaneous fission rate in ^{238}Pu. Also, the high specific heat of ^{238}Pu creates handling problems; small microspheres of ^{238}Pu can melt through gloves in glove boxes and produce contamination problems.

Plutonium compounds created during the plutonium manufacturing process can produce very high neutron dose rates, especially PuF_4 created during the separation and purification of plutonium. Fluorinator glove boxes typically have the highest neutron dose rates in a plutonium processing line. Although PuO_2 is the preferred form because of its chemical stability, the oxide emits almost twice as many neutrons as pure metal. Neutrons are produced in alpha-neutron reactions with ^{17}O and ^{18}O. Some PuO_2 sources used in medical applications are prepared with enriched ^{16}O to reduce neutron dose rates, but isotopic enrichment is generally not used to reduce neutron doses from plutonium compounds.

6.2.3.1 Calculated Neutron Dose Rates

Neutron dose rates can be calculated accurately with computer codes. The MCNP code has the advantage that it can calculate both neutron and photon doses through shielding and in complex arrays. The Monte Carlo codes can also calculate the effects of neutron multiplication in systems containing large amounts of plutonium.

However, neutron dose rates can also be calculated from simple empirical formulas. Unlike gamma doses, there is very little self-shielding for neutrons in subkilogram masses of plutonium. In most instances, a canister containing plutonium can be treated as a point source at the geometric center of the plutonium. The neutron dose equivalent rate from a plutonium source can be calculated by:

$$H = 0.0097\ S/r^2 \qquad (6.4)$$

where H = dose rate, mrem/h

r = distance from the center of the source, cm

S = neutron emission rate from the plutonium source.

The total neutron emission rate, S, is the product of the mass of plutonium (in grams) times Y, the total neutron yield per gram of plutonium (neutrons/second/gram) from spontaneous fission, (α ,n) reactions with low atomic number elements in contact with the plutonium, and fission-induced neutrons. But kilogram quantities of metals or compressed oxides can have significant multiplication and increased emission rates.

6.2.3.2 Neutron Emission Yields

The neutrons produced by spontaneous fission and α ,n reactions can be estimated from the following information. Most neutrons from spontaneous fission originate from the even plutonium isotopes: ^{238}Pu, ^{240}Pu, and ^{242}Pu. Because it is the most abundant, the isotope ^{240}Pu is the most important source of spontaneous fission neutrons. Decay progeny of plutonium have very low spontaneous neutron emissions. Table 6.7 contains spontaneous fission yields for plutonium and other isotopes that may be found in plutonium facilities within the DOE complex. These data are taken from NUREG/CR-5550 (Reilly et al., 1991) and are believed to be more current than the previously published PNL values (Faust et al. 1977, Brackenbush et al., 1988). As a rule of thumb, nuclides with even numbers of protons and neutrons have the highest spontaneous fission neutron emission rates. The spontaneous fission rate for odd-even nuclides is about 1000 times less, and the rate for odd-odd nuclides is about 100,000 less. Spontaneous fission neutrons are emitted with a Maxwellian energy distribution given by the equation:

$$N(E) = (\sqrt{E})\ \mathrm{Exp}\ (E / 1.43\ \mathrm{MeV}) \qquad (6.5)$$

where N(E) is the number of neutrons as a function of the energy E in MeV.

Table 6.7.[1] Spontaneous Fission Neutron Yields

Isotope	Total Half-Life	Spontaneous Half-Life, years	Fission Yield, n/sec-gram
^{232}Th	1.41 x 10^{10} y	>1 x 10^{21}	>6 x 10^{-8}
^{232}U	71.7 y	8 x 10^{13}	1.3
^{233}U	1.59 x 10^{5} y	1.2 x 10^{17}	8.6 x 10^{-4}
^{234}U	2.45 x 10^{5} y	2.1 x 10^{16}	5.02 x 10^{-3}
^{235}U	7.04 x 10^{8} y	3.5 x 10^{17}	2.99 x 10^{-4}
^{236}U	2.34 x 10^{7} y	1.95 x 10^{16}	5.49 x 10^{-3}
^{238}U	4.47 x 10^{9} y	8.20 x 10^{15}	1.36 x 10^{-2}
^{237}Np	2.14 x 10^{6} y	1.0 x 10^{18}	1.14 x 10^{-4}
^{238}Pu	87.74 y	4.77 x 10^{10}	2.59 x 10^{3}
^{239}Pu	2.41 x 10^{4} y	5.48 x 10^{15}	2.18 x 10^{-2}
^{240}Pu	6.56 x 10^{3} y	1.16 x 10^{11}	1.02 x 10^{3}
^{241}Pu	14.35 y	2.5 x 10^{15}	5 x 10^{-2}
^{242}Pu	3.76 x 10^{5} y	6.84 x 10^{10}	1.72 x 10^{3}
^{241}Am	433.6 y	1.05 x 10^{14}	1.18
^{242}Cm	163 days	6.56 x 10^{6}	2.10 x 10^{7}
^{244}Cm	18.1 y	1.35 x 10^{7}	1.08 x 10^{7}
^{249}Bk	320 days	1.90 x 10^{9}	1.0 x 10^{5}
^{252}Cf	2.646 y	85.5	2.34 x 10^{12}

1 Adapted from NUREG/CR-5550 (Reilly et al., 1991)

Energetic alpha particles can overcome coulomb barriers in low-atomic-number elements and create an unstable nucleus that emits neutrons. Because of the high alpha activity of plutonium, this can be a significant source of neutrons. There are two nuclear reactions that are of importance:

$$\alpha + {}^{18}O \rightarrow {}^{21}Ne + n \quad (6.6)$$

$$\alpha + {}^{19}F \rightarrow {}^{22}Na + n. \quad (6.7)$$

Table 6.8 contains the alpha-neutron yields for oxides and fluorides for the most common plutonium and transuranic nuclides. Note that the neutron yields are normalized per gram of nuclide, not per gram of compound. To obtain the yields per gram of compound, multiply by 0.88 for PuO_2 and 0.76 for PuF_4. These data are taken from NUREG/CR-5550 (Reilly et al., 1991).

Table 6.9 contains the neutron yields for trace amounts of elemental impurities in plutonium metal or oxide. These data are also from NUREG/CR-5550 (Reilly et al., 1991) and are derived from thick target yields from accelerator data. The data in Table 6.9 differ from previous values in BNWL-2086 (Faust et al., 1977), and the authors have not experimentally checked the accuracy of these values. Two sets of data are included: one for alphas emitted from enriched uranium and the other for alphas emitted from ^{239}Pu. To determine the neutron yield from trace impurities, it is first necessary to determine the specific alpha activity from Table 6.8, and the neutron yield per parts per million per 10^6 alphas from Table 6.9 for either enriched uranium or plutonium. The specific neutron yield from impurities can be estimated from the following formula:

$$Y_{imp} = 10^{-12} A_{\alpha} \sum_{j}^{n} P_j \; I_j \qquad (6.8)$$

where A_{α} = alpha activity of the plutonium nuclides

P_j = specific neutron yield from the jth element (neutrons/alpha part per million) from Table 6.9

I_j = elemental impurity concentration in plutonium (parts per million).

Note that this formula is valid only if the impurities are uniformly distributed with the plutonium so that the alpha particles directly interact with the impurities. Dust layers of plutonium oxide can also produce high neutron yields. For example, plutonium oxide dust layers on HEPA filters with borosilicate glass can produce neutron emission rates 10 times higher than those for pure oxide because of alpha-neutron reactions with boron in the glass fibers and aluminum spacer plates.

The total neutron yield per gram of plutonium can be found by summing the contributions from:

-- Spontaneous fission (from Table 6.7)

-- alpha-neutron reactions in oxides or fluorides (from Table 6.8)

-- neutrons from low-atomic-number impurities (from Table 6.9).

Table 6.8. Neutron Yields from Alpha-Neutron Reactions for Oxides and Fluorides

Isotope	Alpha Decay Half-Life	Alpha Yield α/s-g	Average Alpha Energy MeV	α, n Yield in Oxides n/s-g	α, n Yield in Fluorides n/s-g
^{232}Th	1.41 x 10^{10} y	4.1 x 10^{3}	4.00	2.2 x 10^{-5}	
^{232}U	71.7 y	8.0 x 10^{11}	5.30	1.49 x 10^{4}	2.6 x 10^{6}
^{233}U	1.59 x 10^{5} y	3.5 x 10^{8}	4.82	4.8	7.0 x 10^{2}
^{234}U	2.45 x 10^{5} y	2.3 x 10^{8}	4.76	3.0	5.8 x 10^{2}
^{235}U	7.04 x 10^{8} y	7.9 x 10^{4}	4.40	7.1 x 10^{-4}	0.08
^{236}U	2.34 x 10^{7} y	2.3 x 10^{6}	4.48	2.4 x 10^{-2}	2.9
^{238}U	4.47 x 10^{9} y	1.2 x 10^{4}	4.19	8.3 x 10^{-5}	0.028
^{237}Np	2.14 x 10^{6} y	2.6 x 10^{7}	4.77	3.4 x 10^{-1}	
^{238}Pu	87.74 y	6.4 x 10^{11}	5.49	1.34 x 10^{4}	2.2 x 10^{6}
^{239}Pu	2.41 x 10^{4} y	2.3 x 10^{9}	5.1	3.81 x 10^{1}	5.6 x 10^{3}
^{240}Pu	6.56 x 10^{3} y	8.4 x 10^{9}	5.15	1.41 x 10^{2}	2.1 x 10^{4}
^{241}Pu	5.90 x 10^{5} y	9.4 x 10^{7}	4.89	1.3	1.7 x 10^{2}
^{242}Pu	3.76 x 10^{5} y	1.4 x 10^{8}	4.90	2.0	2.7 x 10^{2}
^{241}Am	433.6 y	1.3 x 10^{11}	5.48	2.69 x 10^{3}	
^{242}Cm	163 days	1.2 x 10^{14}	6.10	3.76 x 10^{6}	
^{244}Cm	18.1 y	3.0 x 10^{12}	5.80	7.73 x 10^{4}	
^{249}Bk	6.6 x 10^{4} y	8.8 x 10^{8}	5.40	1.8 x 10^{1}	
^{252}Cf	2.646 y	1.9 x 10^{13}	6.11		6.0 x 10^{5}

Multiplying the specific neutron yield (neutrons/second-gram of plutonium) by the mass of plutonium (grams) gives S, the neutron emission rate (neutrons/second). The dose rate is then calculated using Equation 6.4.

Table 6.9. Neutron Yields for Trace Impurities in Plutonium and Uranium

Element	Neutron Yield Per 10^6 Alphas at 4.7 MeV (^{234}U)	Neutron Yield Per 10^6 Alphas at 5.2 MeV (^{234}U)	Average Neutron Energy in MeV For 5.3-MeV Alphas from Pu
Li	0.16 ± 0.04	1.13 ± 0.25	0.3
Be	44. ± 4	65. ± 5	4.2
B	12.4 ± 0.6	17.5 ± 0.4	2.9
C	0.051 ± 0.002	0.078 ± 0.004	4.4
O	0.040 ± 0.001	0.059 ± 0.002	1.9
F	3.1 ± 0.3	5.9 ± 0.6	1.2
Na	0.5 ± 0.5	1.1 ± 0.5	
Mg	0.42 ± 0.03	0.89 ± 0.02	2.7
Al	0.13 ± 0.01	0.41 ± 0.01	1.0
Si	0.0028 ± 0.002	0.076 ± 0.003	1.2
Cl	0.01 + 0.01	0.07 + 0.04	

2.3.3 Radiation Weighting Factors for Neutrons

Approved Radiation Weighting Factors for neutrons are provided in 10 CFR 835.2. As used here Radiation Weighting Factor means the principal modifying factor used to calculate the equivalent dose from the absorbed dose; the absorbed dose (expressed in rad or gray) is multiplied by the appropriate Radiation Weighting Factor (wR).The Radiation Weighting Factors to be used for determining equivalent dose in rem for neutrons are as follows:

RADIATION WEIGHTING FACTORS[1], w_R

Type and energy range	Radiation weighting factor
Photons, electrons and muons, all energies	1
Neutrons, energy < 10 keV[2, 3]	5
Neutrons, energy 10 keV to 100 keV[2, 3]	10
Neutrons, energy > 100 keV to 2 MeV[2, 3]	20
Neutrons, energy > 2 MeV to 20 MeV[2, 3]	10
Neutrons, energy > 20 MeV[2, 3]	5
Protons, other than recoil protons, energy > 2 MeV	5
Alpha particles, fission fragments, heavy nuclei	20

1. All values relate to the radiation incident on the body or, for internal sources, emitted from the source.
2. When spectral data are insufficient to identify the energy of the neutrons, a radiation weighting factor of 20 shall be used.
3. When spectral data are sufficient to identify the energy of the neutrons, the following equation may be used to determine a neutron radiation weighting factor value:

$$w_R = 5 + 17 \exp\left[\frac{-(\ln(2E_n))^2}{6}\right]$$

Where E_n is the neutron energy in MeV.

6.3 RADIATION DETECTION AND EVALUATION

This section describes the response of portable instruments, personnel dosimeters, and nuclear accident dosimeters to the radiations emitted by plutonium, with a focus on photons and neutrons. Data are also included on special spectrometry instruments used to calibrate dosimeters in the field.

6.3.1 Response of Portable Survey Instruments

The energy and angular responses of almost all portable gamma survey instruments have been well characterized and published in the instruction manuals available from the manufacturers. Because of the preponderance of low-energy photons, especially the 60-keV photons emitted by ^{241}Am, particular attention should be given to the low-energy response.

It is not generally well known that neutron survey instruments have a severe energy dependence. In fact, some manufacturers claim a ±15% response per unit dose equivalent extending over an energy range of thermal to 15 MeV. The energy dependence of several commercially available neutron survey meters has been experimentally measured at the PTB in Germany (Liesecki and Cosack, 1984). Their measurements made with monoenergetic neutrons in low-scatter conditions demonstrate that a typical moderator-based neutron survey meter underestimates the dose equivalent by a factor of 2 at an energy of 14 MeV and overestimates dose equivalent by a factor of 2 to 3 at an energy of 20 keV. Survey instruments also exhibit changes in response with the direction of incidence of the neutrons due to absorption and scattering of the neutrons by the electronics package attached to the moderator/detector. This can also result in 40% variation in response, depending on the direction of incidence. Fortunately, plutonium compounds emit neutrons in the MeV range, where the problems with energy and angular responses are minimal. Accuracies of ±15% can be achieved with careful calibration with ^{252}Cf or other fission sources.

6.3.2 Personnel Dosimetry

The detailed requirements of an external dosimetry program are given in Chapter 6 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a). Explicit guidance and requirements are given and need not be repeated here. This section will focus on dosimetry problem areas specific to plutonium facilities and possible solutions.

Personnel working in plutonium facilities are exposed to both photon and neutron radiations, and plutonium processing is one of the largest contributors to neutron exposure in the United States. The response of beta-gamma personnel dosimeters is well documented and will not be discussed here.

The response of neutron dosimeters to the neutron fields encountered in the workplace shall be evaluated. All existing neutron dosimeters have a severe energy response problem and shall be carefully calibrated for the specific radiation field in which the neutron dosimeter is worn. Typically, neutron dosimeters are calibrated to either bare ^{252}Cf or D_2O-moderated ^{252}Cf sources in a low-scatter calibration facility. Then, the neutron dosimeters are worn in

plutonium facilities under high-neutron-scatter conditions, which produce a completely different energy spectrum than that in which the dosimeter was calibrated. Accordingly, a typical approach is to perform neutron energy field characterization surveys at selected areas in a facility. Based on these surveys a field correction factor is determined which corrects for the difference between the dosimeter response to the calibration source and the response to the neutron fields in the workplace. Because of the large response of TLD-albedo dosimeters to low-energy neutrons (with energies below about 20 keV), the response of the dosimeter usually depends on the scattering conditions rather than the initial neutron energy spectrum. These problems are discussed in detail in several documents, including PNL-3213, Personnel Neutron Dosimetry at Department of Energy Facilities (Brackenbush et al., 1980) and PNL-7881, Response of TLD-Albedo and Nuclear Track Dosimeters Exposed to Plutonium Sources (Brackenbush et al., 1991).

Thermoluminescent dosimeters are the most widely used neutron dosimeters in plutonium facilities. The energy response of a typical TLD-albedo dosimeter is shown in Figure 6.2. At neutron energies below about 20 keV, the energy response is almost constant. Above 20 keV, the response per unit dose drops dramatically by almost three orders of magnitude at 10 MeV. Almost all neutrons emitted by plutonium have energies in the MeV range. However, about 50% of the neutrons striking a thick concrete wall or floor are reflected back into the room at lower energies, and neutrons typically are reflected two or three times before being absorbed. Thus, the low-energy scattered neutrons are often more important in determining TLD-albedo dosimeter response than the high-energy neutrons emitted by the plutonium source. The TLD-albedo dosimeters are often calibrated in specific facilities by exposing them on phantoms at locations where the dose equivalent has been carefully determined from dose and spectrometric measurements (Brackenbush et al., 1991).

Nuclear track dosimeters are also being used for personnel dosimetry in plutonium facilities. These dosimeters have the advantage of a much more constant response per unit dose equivalent, as shown in Figure 6.3. Nuclear track dosimeters operate on the principle that a fast neutron interacts with plastic to produce a proton recoil that damages the polymer. Under special etch conditions, the damaged areas are removed to produce a distinct track, which is easily observed under a microscope. The neutron dose equivalent is then determined from the track density. Nuclear track dosimeters have a distinct threshold, usually about 100 keV.

In conclusion, the combination of TLD-albedo and nuclear track dosimeters can provide a more uniform response with energy and more accurate personnel dosimetry. This combination of dosimeters may be an appropriate solution to neutron dose monitoring in DOE facilities with significant neutron exposure.

It is important to verify and document that personnel dosimetry systems provide accurate measurements and records of the occupational radiation doses received by workers in plutonium facilities (McDonald et al., 1992). To provide a level of confidence in dosimetry services in DOE facilities, the DOELAP accreditation program was established. 10 CFR 835 402(b) (DOE, 2011) requires that personnel dosimetry programs implemented to demonstrate compliance with the

dose limits established in Subpart C shall be accredited in accordance with the requirements of the DOELAP for Personnel Dosimetry (ANSI, 2009) (DOE 2018a). NIST has also established the National Voluntary Laboratory Accreditation Program (NVLAP) for testing and accreditation of dosimeter processors serving commercial industry and medical facilities. Because the dosimetry needs at many DOE facilities, particularly those processing plutonium, are different from commercial industries, the DOE initially established a broader and more stringent accreditation program. Both DOELAP and NVLAP accreditation programs use performance tests that evaluate the accuracy and precision of personnel dosimetry measurements. The accuracy is determined by comparing the measured dose equivalent to the "conventionally true dose equivalent" derived from calibration standards directly traceable to NIST in carefully controlled conditions.

Two laboratories conduct the performance test irradiations for the DOELAP and NVLAP programs: Pacific Northwest National Laboratory of Richland, Washington, and the Radiological and Environmental Sciences Laboratory (RESL) of Idaho Falls, Idaho. Dosimeters are submitted for testing to the performance testing laboratories in specified categories. If the dosimeter passes certain accuracy and tolerance testing criteria, a team of dosimetry experts visit the processor and/or site and assess the operation of the dosimetry program, including dosimetry records and data retrieval systems, before the dosimeter processor or DOE site is accredited. DOE requirements are given in the DOE STD-1095- 2018 (DOE 2018a).

6.3.3 Extremity Dosimetry

Doses to the extremities from plutonium processing and handling can involve significant exposures to the skin of the hands and forearms. For information on performance testing of extremity dosimeters refer to DOE STD-1095- 2018 (DOE 2018a) which incorporates ANSI/HPS N13.32-2008, Performance Testing of Extremity Dosimeters (ANSI, 2008b). Doses over small areas of the skin are discussed in Chapter 6 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a) and will not be discussed here. That chapter discusses skin contamination including hot particles, and the determination of skin dose from these events.

Highly accurate measurement of the dose to the hands and forearms is especially difficult because of the low-energy photons (L x-rays and 60-keV photons from ^{241}Am). Small variations in shielding, such as differences in the thickness of gloves used in glove boxes or non-uniform distribution of plutonium oxide dust on the surface of gloves, can produce large variations in the dose rate. Examples of these variations were given previously.

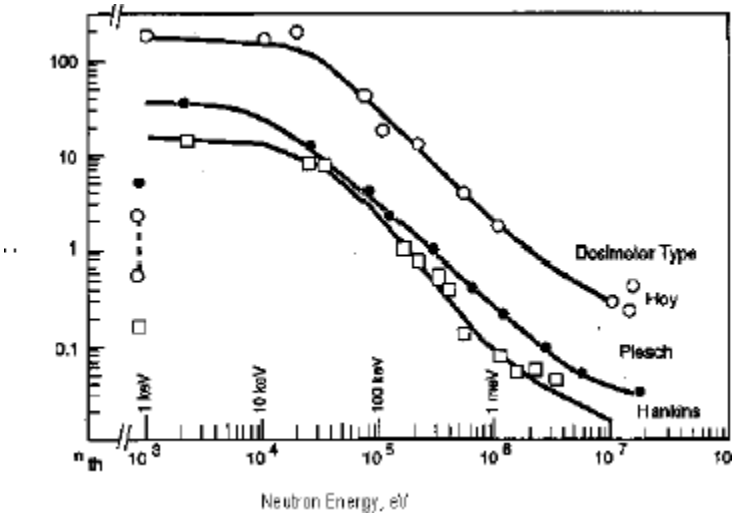

Figure 6.2. Energy Dependence of Various TLD-Albedo Dosimeters (Source: Piesch and Burgkhardt, 1978)

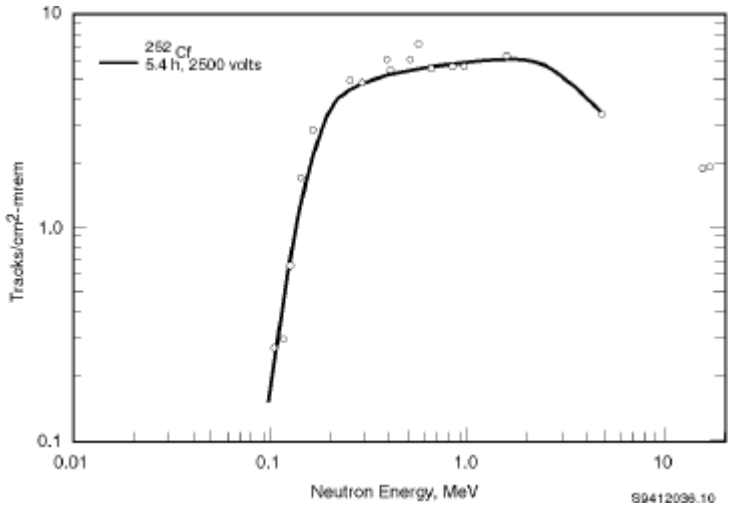

Figure 6.3. Response of Electrochemically Etched CR-39 Used in Nuclear Track Dosimeters as a Function of Neutron Energy

Extremity doses are typically determined by TLD finger rings, which are usually worn with the TLD chip facing the radiation source on the palm side of the hand. In glove-box and in D&D operations, the photon dose is usually significantly higher than any neutron dose. However, neutron dosimeters are sometimes worn to estimate extremity doses. Two types of neutron extremity dosimeters have been used: nuclear track dosimeters worn in special finger rings and specially calibrated TLD-albedo dosimeters worn on the wrist or forearm. DOE STD-1095-2018 (DOE 2018a) is currently applicable to personnel dosimeters for whole body irradiation.

There is some question about the correct radiation weighting factor to apply to extremity neutron dosimeters. Radiation weighting factors were derived from biological experiments on cancer induction, especially leukemia in blood-forming organs. There are no blood-forming organs in the extremities, so there is no biological basis for large values of radiation weighting factors for extremity exposures. However, regulatory agencies typically apply radiation weighting factors derived for whole-body exposures to the extremities, thus for compliance purposes radiation weighting factors should be applied for extremity exposures.

6.3.4 Criticality Accident Dosimetry

A criticality safety program, which includes material control, criticality alarms, and criticality accident dosimetry, is required as outlined in DOE Order 420.1C (DOE, 2012b). The requirements in 10 CFR 835.1304 require that fixed nuclear accident dosimeters (NADs) and personnel nuclear accident dosimeters (PNADs) shall be worn by all individuals entering a controlled area that contains certain quantities of fissile materials, such as those required in DOE Order 420.1C (DOE, 2012b); which requires installed criticality alarms. The criticality accident dosimetry system should follow the provisions of ANSI N13.3, Dosimetry for Criticality Accidents (ANSI, 1969); this standard, although currently withdrawn from ANSI/HPS, is being revised. Information on criticality accident dosimetry is also available from the International Atomic Energy Agency (IAEA, 1982).

The criticality accident program should contain the following items:

-- a method and procedure to conduct an initial screening of individuals involved in a nuclear accident to determine whether significant exposures to radiation occurred (10 CFR 835.1304(b)(1))

-- methods, procedures, and equipment for obtaining and analyzing biological materials (including ^{24}Na activity from blood samples and ^{32}P activity in the hair)(10 CFR 835.1304(b)(2)), as well as metal coins, jewelry, and articles of apparel that may have become activated from neutrons

-- a system of fixed dosimeters (i.e., NADs) (10 CFR 835.1304b(3)) capable of furnishing estimated radiation dose within an accuracy of ±25% and the approximate neutron spectrum at the installed locations to allow conversion from rad to rem

-- an operating range for the fixed dosimeters' neutron component 10 rad to 10,000 rad

-- measurement capabilities for the fixed dosimeters' gamma ray component of fission gamma rays in the presence of neutrons with an accuracy of ±20%, and an operating range for the gamma component operating range extending from 10 rad to 10,000 rad

-- PNADs capable of furnishing sufficient information to determine neutron and gamma dose with an accuracy of ±25% over a range of 10 rad to 1000 rad without dependence upon fixed NAD data

-- a radiological counting laboratory with the methodology, analytical procedures, and quality assurance program in place to count the activated samples from the criticality accident and provide results quickly

-- counting of activities in persons with significant exposures to assess the activation products in the body if a whole body counter is available (this is one of the more accurate methods for dose estimation)

-- a health physicist designated to coordinate and evaluate the dosimetry information and provide dose estimates shortly after the accident

-- means to obtain medical treatment for personnel who receive a high radiation dose

-- a quality assurance program in place to help assure the accuracy and validity of the dosimetry results.

As mentioned in Section 6.1, the concept of equivalent dose was used to quantify exposures to different radiations. The radiation weighting factors used to determine equivalent dose are based on stochastic effects, primarily cancer induction some years later. But the doses in criticality accidents are typically so large that acute symptoms, including death, may occur within relatively short times, and radiation weighting factors are usually not applicable. For this reason, it is usually more appropriate to determine absorbed dose rather than equivalent dose if a person receives more than about 25 rem. These absorbed dose estimates to the torso are much more important for triage and treatment considerations.

The NADs are used to determine the neutron and photon dose at various locations in the plutonium facility, as well as providing spectral and calibration data for PNADs. A typical NAD used at the Hanford Site is shown in Figure 6.4. This unit is fixed to the wall or posted at locations around plutonium storage areas where it is easily recovered in the event of a criticality. The “candle” insert contains neutron- and gamma-sensitive TLDs as well as activation foils positioned at the center of the detector. Tests at the Health Physics Research Reactor at Oak Ridge have shown that this arrangement gives accurate estimates of “deep” dose for both neutrons and gamma rays. A set of foils identical to those used in the PNAD dosimeter is positioned above the moderator. These foils provide an estimate of the average cross-section or response per unit dose, so that

the neutron dose from the foils in the PNAD can be more accurately evaluated for the incident neutron spectrum.

The PNAD dosimeter typically consists of several activation foils. In the case of the Los Alamos/Hanford design (Vasilik and Martin, 1981), the activation foils consist of ½-in.- diameter foils of bare and cadmium-covered gold, bare and cadmium-covered indium, cadmium-covered copper, and a sulfur pellet. Algorithms have been developed to unfold an approximate neutron energy spectrum from the measured neutron activation products, so that neutron doses can be calculated. Criticality dosimeters containing various activation foils are available from vendors, but some of the commercial products do not contain sufficient material to measure neutron doses as low as 10 rad, which is the recommended lower detection limit for personal criticality accident dosimeters.

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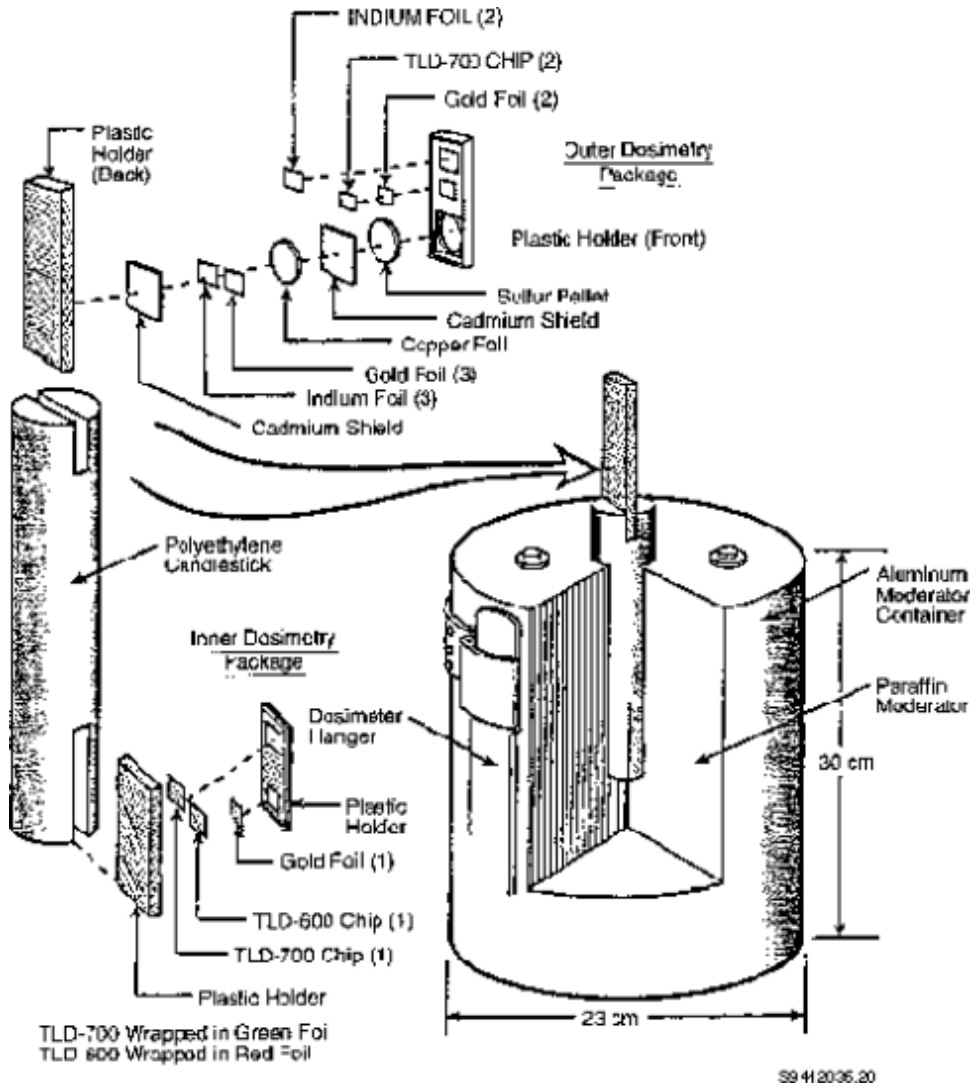

Figure 6.4 Fixed Nuclear Accident Dosimeter Used at Hanford to help Assess Doses from Criticality Accidents

6.3.5 Dose to Lens of Eye

The dose to the lens of the eye is not generally a problem in plutonium facilities because whole body exposures are generally the limiting case. Dosimeters to measure the dose to the lens of the eye are seldom used. However, equivalent doses to the extremity and skin at depths of 3 mm can be appreciable in cases where there is an abundance of low-energy photons, such as during visual inspection of machined plutonium pieces on laminar-flow tables or other situations where the plutonium is not shielded. In these cases, the eyes are generally protected by requiring safety glasses to be worn.

6.3.6 Spectrometry Measurements

Personnel neutron dosimeters used at DOE plutonium facilities include TLD-albedo and nuclear track detectors. The response per unit equivalent dose for TLD-albedo dosimeters is a sensitive function of incident neutron energy (see Figure 6.2). These dosimeters are typically calibrated under low-scatter conditions in a calibration laboratory, such as the facility at PNL used for exposing dosimeters for DOELAP accreditation. The dosimeters are calibrated to a fission spectrum from ^{252}Cf or a degraded fission spectrum from D_2O-moderated ^{252}Cf. However, the neutron energy spectrum of the workplace is significantly different from that of the calibration facility and the response per unit equivalent dose is also different, primarily because of the number of low-energy neutrons produced by scatter within process equipment, glove boxes, and the walls and floor of the facility. To achieve accurate results, the TLD-albedo dosimeter results shall be corrected for the specific neutron energy spectrum in which they are exposed. One method to achieve accuracy is to expose neutron dosimeters on a phantom in the workplace in neutron fields where the equivalent dose rate has been carefully measured using neutron spectrometers.

There are several neutron energy spectrometers available to make accurate neutron spectrum measurements and dose estimations, as outlined in the document A Field Neutron Spectrometer for Health Physics Applications (Brackenbush et al., 1992). Neutron spectrometers that are useful for dose determinations in plutonium facilities include:

-- Multisphere or Bonner sphere spectrometers

-- tissue equivalent proportional counters (to determine linear energy transfer (LET) spectra)

-- liquid scintillator spectrometers

-- proton recoil spectrometers.

6.3.6.1 Multisphere Spectrometer System

The multisphere or Bonner sphere spectrometer (Bramblett et al., 1960) is the neutron spectrometer system most often used by health physicists for neutron energy spectrum measurements, perhaps because it is simple to operate. Multisphere spectrometers are typically used for measuring neutron energy spectra over a wide energy range from thermal energies to over 20 MeV although detailed energy spectra are not obtained. With the use of an appropriate spectrum unfolding code, the multisphere system will determine the average neutron energy, dose rate, total flux, kerma, and graphical plots of differential flux versus energy and dose distribution versus energy.

The multisphere spectrometer consists of a set of polyethylene spheres of different diameters, typically 3 in. to 12 in. A thermal neutron detector, such as a ^{3}He proportional counter or a ^{6}LiI scintillator is positioned at the center of each sphere, and the count rate measured. The neutron energy spectrum can be determined from the ratio of counts from different detectors. However, the spectral unfolding algorithms do not provide mathematically unique solutions. The most appropriate solutions are obtained by making an initial guess that the spectrum consists of a fission spectrum with a 1/E "tail." Multisphere spectrometers have demonstrated accuracies of ±15% when exposed to ^{252}Cf sources with calibrations directly traceable to NIST (Brackenbush et al., 1991).

Figure 6.5 demonstrates the type of neutron energy spectra measured by the multisphere spectrometer. The plot shows the logarithmic plots of four multisphere spectrometer measurements made at a distance of 50 cm from 1 kg of plutonium for "bare" plutonium fluoride (i.e., no intervening shielding), plutonium fluoride shielded with 10 cm (4 in.) of acrylic plastic, "bare" plutonium oxide, and "bare" plutonium metal. The plutonium fluoride has the highest neutron emission rate and corresponds to the highest peak in the graph. The lowest peak corresponds to the moderated plutonium fluoride spectrum with 4 in. of acrylic plastic shielding. These measurements are typical of the neutron energy spectra in plutonium processing areas containing glove boxes.

The spectra contain a significant fraction of low-energy scattered neutrons from the glove boxes and the thick concrete floor and walls of the facility. The spectra are distinctly different from neutron emission spectra (see Section 6.2), which do not contain scattered or background neutrons.

6.3.6.2 Tissue Equivalent Proportional Counter

The tissue equivalent proportional counter (TEPC) is not often used by health physicists, but it can provide highly accurate estimates of dose. The TEPC consists of a hollow sphere or cylinder of tissue equivalent plastic filled with low-pressure equivalent gas. The pressure is so low (a few torr) that the TE gas cavity has the same mass stopping power as a 2-μm sphere of tissue at unit density. Because the TEPC actually measures the energy absorption in a known mass of tissue equivalent material, it provides an absolute measure of absorbed neutron dose. The TEPC also measures the pattern of microscopic energy distributions from any penetrating ionizing radiation. With appropriate algorithms, LET distributions, hence radiation weighting factors, can be calculated. Thus, the TEPC provides absorbed dose, radiation weighting factor, and dose from a single spectral measurement of the event size distribution from the TEPC.

The TEPC can provide highly accurate measurements of dose under laboratory conditions. However, it suffers from stability problems, and its accuracy decreases with time as impurities diffuse from the TE plastic walls and temperature changes cause gain shifts in the proportional counter. Nevertheless, the TEPC can provide reasonably accurate measurements of dose in the workplace (±15%) over extended time periods of 6 months or more, and can be used to monitor dosimeter irradiations on phantoms in the workplace.

6.3.6.3 Liquid Scintillator Spectrometer

The liquid scintillator spectrometer typically consists of a 2-in. by 2-in.cylindrical cell of hydrogenous scintillator solution in contact with a photomultiplier. Neutrons interact in the scintillator to produce proton recoils, which interact with the scintillator to produce light. With careful calibration, the incident neutron energy spectrum can be unfolded from the measured distribution of scintillation events.

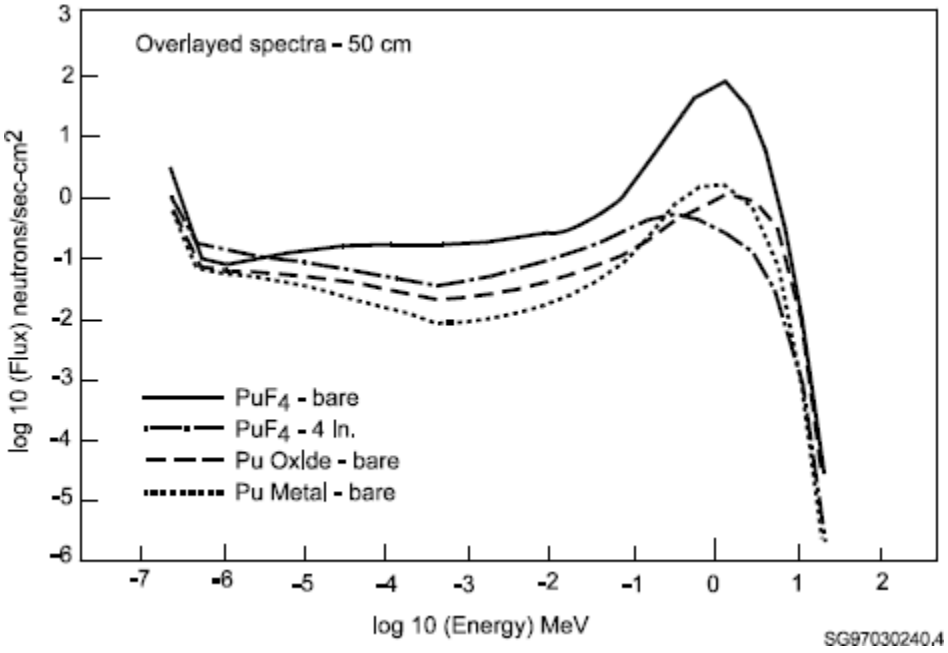

Figure 6.5. Neutron Energy Spectra as Measured by the Multisphere Spectrometer at 50 cm from Plutonium Metal, PuO_2, and PuF_4 Sources

The liquid scintillator spectrometer has the advantage that it is very sensitive and can operate at low dose rates. It is useful over an energy range extending from about 1 MeV to 20 MeV. Neutron equivalent dose can be calculated from the measured spectra using the conversion factors referenced in Chapter 6 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a). The doses calculated from liquid scintillator measurements are reasonably accurate (±10% to ±20%) for lightly moderated plutonium spectra. Because of the lower energy cut-off of liquid scintillator spectrometers, they may not provide accurate dose equivalent values outside heavily shielded facilities, such as plutonium storage vaults with thick concrete walls.

6.4 EXTERNAL DOSE REDUCTION

The traditional methods of using time, distance, and shielding are typically employed in plutonium facilities to reduce exposures to ALARA levels. However, other considerations may be just as important. Good housekeeping practices are vital to keep dose rates low. Even invisible dust layers on the interior surfaces of glove boxes can create gamma radiation fields of 10 mrem/h or more, especially through lightly shielded glove ports. The practice of pulling gloves outside for storage should not be condoned in operations that generate dust or powders. Dose rates of 30 mrem/h have been measured in facilities processing high-exposure oxide powders. A factor of 30 reduction in dose rate was achieved by merely storing the gloves inside the glove box when not in use and placing lightweight “pie plate” shields over the glove-port openings.

6.4.1 Time

Obviously, reducing the time a worker is exposed in a radiation field will reduce the dose. Any operation which involves elevated dose rates (more than a few mrem/hour) or long exposures should be reviewed for possible reductions in a worker's exposure time. For example, a worker should minimize the time spent near a fluorination operation. After the equipment has been set up, the worker should leave the area during the actual fluorination step.

6.4.2 Distance

Because of the inverse square relationship with discrete radiation sources, significant dose reductions can be achieved by increasing the distance between the worker and the plutonium source. Also, the low-energy photons emitted through glove ports and bag-out ports can be attenuated by several feet of air. Most plutonium operations involve contact work, so increasing the distance may not always be practical. But significant reductions in doses can be achieved by reducing plutonium inventories in glove boxes. It is good practice not to store plutonium samples in glove boxes, but to remove them to storage vaults or other shielded locations. In many cases, the plutonium samples can be stored in the glove box in “wells” or specially shielded areas at some distance from the work areas where the plutonium technicians spend most of their time. The best method of reducing neutron dose is simply to remove the plutonium from the glove box and minimize inventories in the glove box.

6.4.3 Shielding

The most practical method of reducing doses in plutonium operations is to apply shielding. Plutonium emits both neutrons and photons, which require different types of shielding materials to be effective. There are also additional constraints that shall be met, such as the maximum thickness of shielding that can be placed on glove boxes and still retain worker mobility. It has been found that more than about 8 cm (4 in.) of shielding on the exterior surface of a glove box greatly reduces the worker's manual dexterity and efficiency. It is also important to place the shielding close to the plutonium source and not to try to shield personnel. Because neutrons scatter around shadow shields, it is usually best to shield all

surfaces of glove boxes or storage areas. The following sections describe the shielding effectiveness of common photon and neutron shielding materials.

6.4.3.1 Photon Shielding

Because of the preponderance of low-energy photons, significant reductions in gamma doses can be achieved by even modest shielding. It is important to note that there is a significant amount of self-shielding in plutonium samples. A 1- mm-thick plutonium metal sample is "infinitely thick" and additional thicknesses will not appreciably increase the dose rate. For this reason, the photon dose is more dependent on the surface area rather than on the mass of plutonium. Invisible dust layers on gloves and interior surfaces of glove boxes can produce high exposure rates, especially if the gloves are pulled outside the glove box for storage to prevent them from being caught in machinery. Simple iron or lead shields placed over the glove ports can reduce the dose rates near the glove box by an order of magnitude. Modest gamma shields of 6 mm (0.25 in.) of lead and 13 mm (0.5 in.) of lead-loaded x-ray glass are usually sufficient to reduce photon dose rates from plutonium to acceptable levels.

Table 6.10 gives examples of how effective various gamma shielding materials are in reducing the dose rates from low-exposure (6% ^{240}Pu) and high-exposure (19% ^{240}Pu) sources. The sources consist of cylinders containing 1 kg of plutonium oxide; the dose rates are given at a distance of 2 m from the source. This example is typical of the shielding effectiveness for cans of plutonium containing kilogram quantities of plutonium oxide, as might be found in storage vaults.

In contrast, Figure 6.6 shows the reduction in photon dose rates from a small sample of plutonium oxide power weighing about 100 grams. The dose rates were measured at a distance of 3 cm from the surface of the plutonium, which was contained in polyvinyl chloride plastic bags (a total thickness of 33 mil or 0.85 mm) for radiation measurements. The isotopic composition of the plutonium was similar to that given in Table 6.11.

Photon radiation is a significant source of exposure, especially during D&D activities, when most of the plutonium has already been removed. Much of the photon exposure problem originates from thin dust layers, as described in the preceding paragraphs. High photon doses often originate from "streaming" through glove ports from dust layers on gloves. But there also can be appreciable neutron dose rates, even in supposedly "empty" glove boxes, from plutonium hold-up, especially in fluorinator glove boxes where there is a high neutron emission rate from alpha-neutron reactions. Wearing lead-loaded aprons can reduce dose rates by a factor of 2 in plutonium fuel manufacturing. High-exposure plutonium (>10% ^{240}Pu) should be handled in glove boxes with lead-loaded Neoprene gloves although some loss of mobility and dexterity may result. The photons from plutonium are easily shielded by several millimeters of lead or iron, but it requires almost 15 cm (6 in.) of

polyethylene or hydrogenous moderator to reduce neutron doses by a factor of 10. Simplistically stated, the gamma dose rate is a function of surface area, while neutron dose rate is a function of the mass of the plutonium and its chemical form.

6.4.3.2 Neutron Shielding

The neutron radiations from plutonium are much more difficult to shield than the photon radiations. As a rule of thumb, it requires about 15 cm of hydrogenous shielding to reduce the neutron dose rate by an order of magnitude.

Figure 6.7 shows the reduction in dose equivalent rate for various shielding materials for plutonium tetrafluoride sources, which have an average neutron energy of 1.3 MeV. For practical purposes, the shielding thickness for glove boxes is limited to about 4 in.; it is not possible to operate machinery through thicker shields. Figure 6.8 shows the reduction in dose equivalent rate through various slab shields for plutonium dioxide. These data were obtained from measurements of the neutron dose using a TEPC. Figures 6.7 and 6.8 show the reduction in dose equivalent rates, the reduction in equivalent dose rates will differ.

Table 6.10.[1] Photon Dose Rates at 2 Meters from Cylinders of Plutonium Containing 1 kg of Plutonium at 5 Years After Chemical Separation

Shield Material	Shield Thickness cm	Photon Dose Rate, mrad/h	
		19% ^{240}Pu Source	6% ^{240}Pu Source
Polyvinyl chloride (PVC)	0.005	19.3	1.74
	0.038	8.72	0.570
	0.084	6.29	0.391
Lead Glove	0.094	1.85	0.105
Heavy Lead Glove	0.152	0.54	0.0464
Lucite	0.612	7.03	0.447
	2.54	3.30	0.190
Steel	0.025	2.69	0.144
	0.038	2.41	0.131
	0.051	2.19	0.121
	0.317	0.42	0.0418
	0.635	0.221	0.0299
	1.33	0.134	0.0205
	2.43	0.0766	0.0119
Lead	0.635	0.0701	0.0103
	1.27	0.0380	0.00288
	2.57	0.0156	0.000391
	5.08	0.00429	0.00023
	10.16	0.000467	0.0000001
X-ray Glass	0.645	0.135	0.0251
	1.30	0.0841	0.0144
	2.60	0.0463	0.00534
Lead Apron	0.16	0.306	0.0346
Safety Glass	1.30	1.94	0.109
	2.60	1.50	0.0886

[1] Adapted from NUREG/CR-5550 (Reilly et al., 1991)

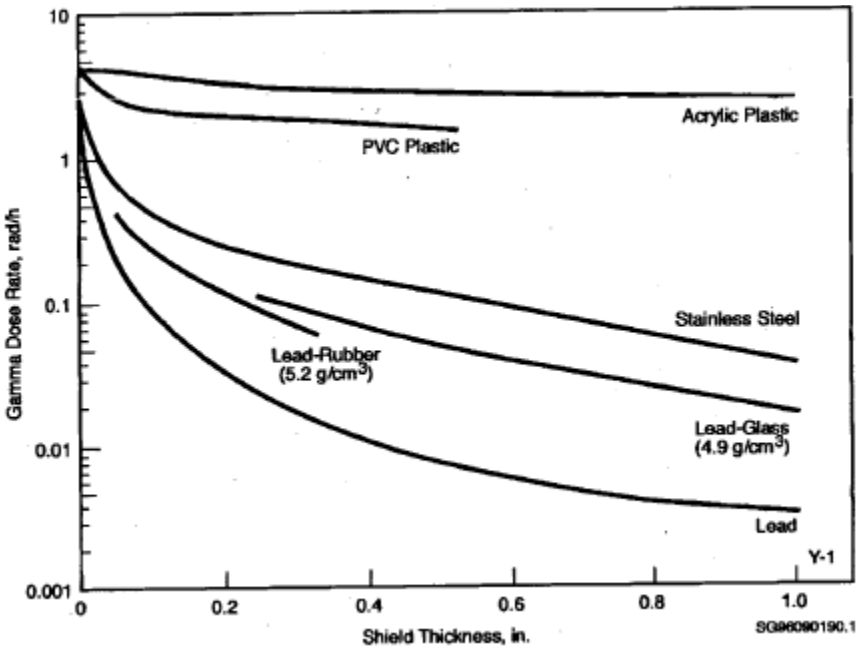

Figure 6.6. Reduction in Photon Dose Rate with Various Shielding Materials at a Distance of 3 cm from a 100-gram Disk of Plutonium Oxide

Table 6.11. Isotopic Composition of Plutonium Sources at 5 years After Chemical Separation of the Plutonium

	Weight-Percent of Isotope	
	Low-exposure Pu	High-exposure Pu
^{235}Pu	0.001	1.85
^{239}Pu	93.5	63.3
^{240}Pu	5.99	19.2
^{241}Pu	0.397	9.27
^{242}Pu	0.001	3.88
^{241}Am	0.103	2.40

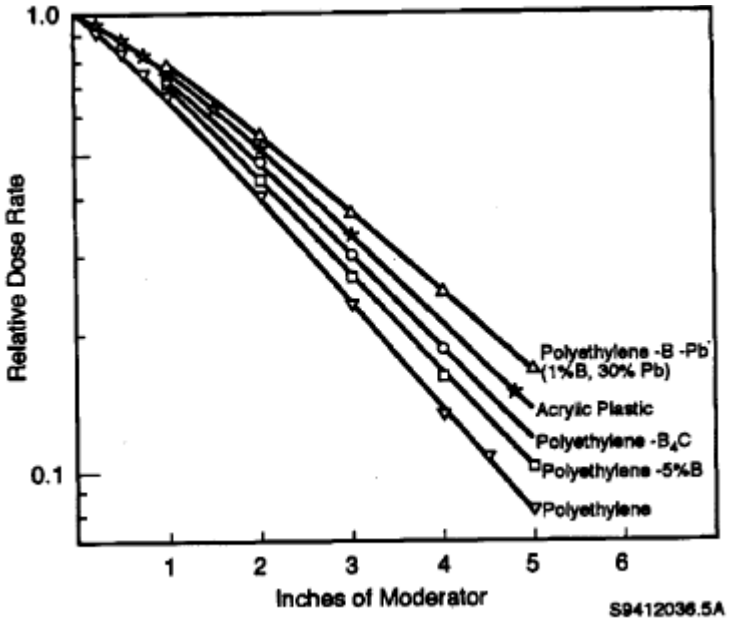

Figure 6.7. Reduction in Neutron Dose Equivalent Rate for Various Slab Shields for Plutonium Tetrafluoride Sources

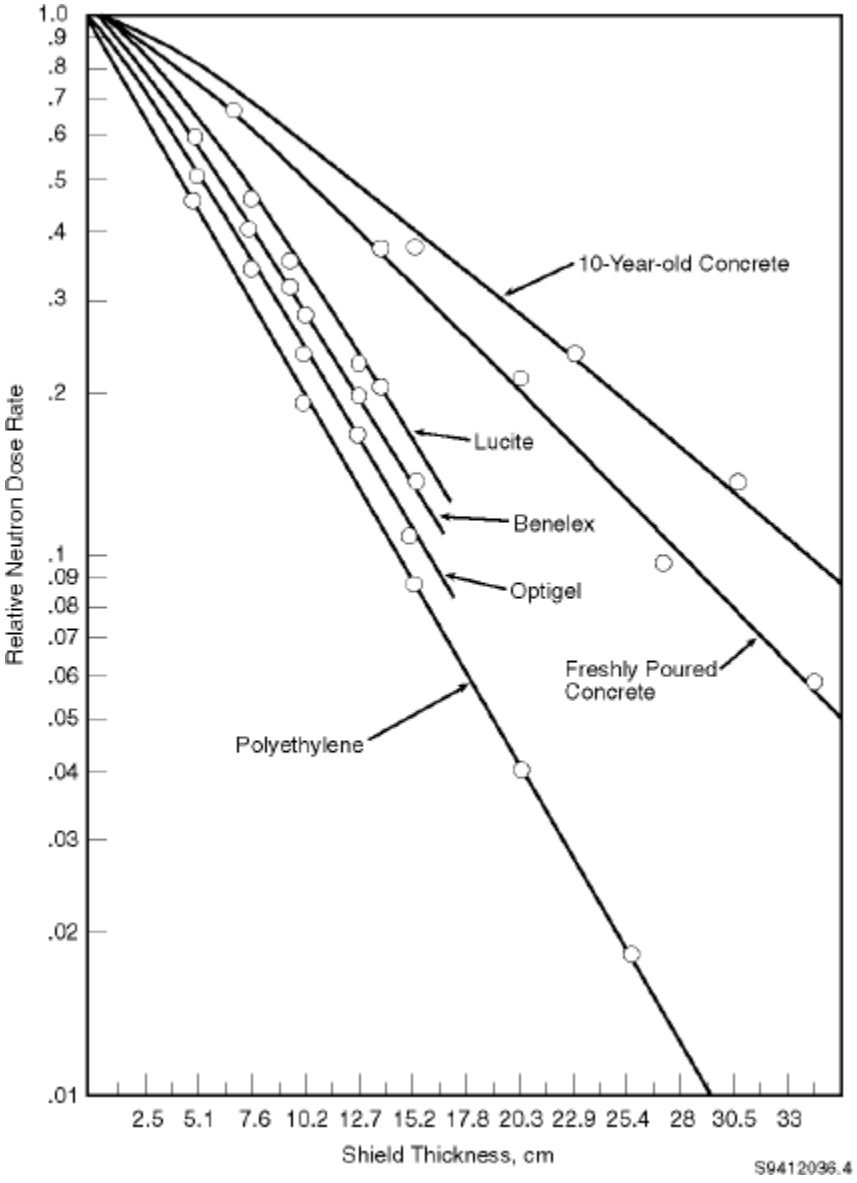

Figure 6.8. Reduction in Neutron Dose Equivalent Rate for Various Slab Shields for Plutonium Oxide Sources

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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)**

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

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)**

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.