

Y-12

OAK RIDGE Y-12 PLANT

LOCKHEED MARTIN



ASSESSMENT

OF

ENRICHED URANIUM STORAGE

SAFETY ISSUES

AT

THE OAK RIDGE Y-12 PLANT

Uranium Storage Assessment Team

August 1996

Prepared by the Oak Ridge Y-12 Plant
Oak Ridge, Tennessee 37831

managed by
Lockheed Martin Energy Systems, Inc.
for the

U.S. Department of Energy
Under contract DE-AC05-84OR21400

MASTER

MANAGED BY
LOCKHEED MARTIN ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

UCN-13672 (26 6-95)

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

RECEIVED
DEC 20 1996
OSTI

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

ASSESSMENT
OF
ENRICHED URANIUM STORAGE
SAFETY ISSUES
AT
THE OAK RIDGE Y-12 PLANT

Uranium Storage Assessment Team

August 1996

Prepared by the Oak Ridge Y-12 Plant
Oak Ridge, Tennessee 37831
managed by
Lockheed Martin Energy Systems, Inc.
for the
U.S. Department of Energy
Under contract DE-AC05-84OR21400

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

EXECUTIVE SUMMARY

All safety issues deemed relevant to the storage of enriched uranium at the Oak Ridge Y-12 Plant were enumerated and analyzed. The approach used in the assessment consisted of the following:

- Compiling the applicable government regulations, Department of Energy (DOE) Orders, miscellaneous DOE publications, and nongovernmental standards that pertain to enriched uranium (EU) storage.
- Compiling the nuclear, chemical, and physical properties of the uranium species that will, or may, be placed in storage at Y-12.
- Presenting the equations used to calculate the properties that impact safety in the storage of EU.
- Citing the primary references from the technical literature used in optimizing EU storage practices with respect to safety issues.
- Reviewing current EU storage practices at Y-12.
- Describing advanced concepts for EU storage.
- Presenting a hazard analysis that enumerates potential hazards and consequences of accidents.
- Presenting the conclusions and recommendations based on the prior sections.

The Hazards Analysis identified the following potential areas that might potentially impact the health or safety of workers or the public, or pose an environmental threat: criticality, chemical reactivity, container pressurization, release of airborne uranium, fire, and material dispersal.

Based on the issues raised in the Hazards Analysis and the nuclear, chemical, and physical properties of uranium and its compounds, a series of recommendations for the safe storage of EU were formulated. Among these are the following:

- The radiation properties of pure unirradiated isotopically EU are much less hazardous than those of transuranics such as plutonium, or of spent reactor fuels that contain highly active fission products. As a result, it is not necessary to make provisions for heat removal, to vent containers for helium pressure buildup due to alpha decay, or to provide massive shielding for high energy gamma radiation. It is, of course, necessary to design the storage configuration to preclude the possibility of criticality events.

- The two optimal chemical forms of EU for long-term storage are pure metal and the oxide, U_3O_8 . The recommended physical forms are cast hollow cylinders for the metal and dry, free-flowing powder for U_3O_8 .
- Both the metal and oxide should be stored corrosion-resistant containers such as 304-L stainless steel cans. Massive concrete arrays are recommended to maintain a criticality-safe configuration under catastrophic circumstances. Both tube vaults and modular storage vaults are satisfactory for this purpose.
- Enriched uranium-containing materials that have not been transferred to long-term storage may be held in Interim Storage (IS) for a period of up to ten years. In, IS criticality safety is maintained by use of approved devices such as bird cages, lock boxes, and carbon steel drums.

ASSESSMENT OF ENRICHED URANIUM STORAGE SAFETY ISSUES AT THE OAK RIDGE Y-12 PLANT

CONTENTS

1. INTRODUCTION	1
1.1 SCOPE	1
1.2 PROGRAM PARTICIPATION	2
1.3 IDENTIFICATION OF ORDERS, STANDARDS, AND DOCUMENTS	2
2. NUCLEAR PROPERTIES THAT RELATE TO STORAGE	3
2.1 URANIUM ISOTOPIC DISTRIBUTIONS	3
2.2 RADIOACTIVE DECAY	4
2.2.1 Half-Lives of the Isotopes	4
2.2.2 Decay Sequences	4
2.2.3 Decay from Daughter Products	5
2.3 ATTENUATION OF RADIOACTIVE EMISSIONS	7
2.3.1 Attenuation of Alpha Particles	7
2.3.2 Attenuation of Gamma Rays (and X rays)	8
2.3.3 Attenuation of Electrons (Including Beta Particles)	9
2.3.4 Generation and Attenuation of Neutrons	9
2.4 CRITICALITY CONSIDERATIONS	10
3. CHEMICAL AND PHYSICAL PROPERTIES AND STORAGE APPLICATIONS ...	11
3.1 URANIUM METAL	11
3.1.1 Storage Implications of the Corrosion of Uranium Metal	12
3.1.2 Alloys, Intermetallics, and Traced Metal	15
3.2 OXIDES	15
3.3 FLUORIDES	17
3.4 OTHER HALIDES AND OXYHALIDES	18
3.5 NITRATES	18
3.6 HYDRIDE	19
3.7 OTHER COMPOUNDS	19
3.8 BASIS FOR SELECTING SUITABLE CHEMICAL FORMS FOR LONG-TERM STORAGE	19
4. CURRENT STORAGE PRACTICES	21
4.1 TRADITIONAL DEFINITIONS OF STORAGE INTERVALS	21
4.2 PREPARATION OF URANIUM FOR STORAGE	22
4.3 IDENTIFICATION OF STORAGE AREAS AT Y-12	22

4.4	PROLONGED LOW-MAINTENANCE STORAGE (PLMS) FACILITIES . . .	23
4.4.1	Tube Vaults	23
4.4.2	Modular Storage Vaults (MSVs)	24
4.4.3	Cage Storage	24
4.5	TRANSIENT STORAGE (TS)	24
4.6	INTERIM STORAGE (IS)	24
4.7	CRITICALITY	25
4.8	INVENTORY MONITORING PROTOCOL	25
4.9	SCHEDULING	26
5.	ASSESSMENT OF ADVANCED STORAGE CONCEPTS	27
5.1	FUNCTION OF CONTAINERS	27
5.1.1	Container Design	27
5.1.2	Atmosphere Within Storage Cans	28
5.1.3	Storage Cans For Prolonged Low-Maintenance Storage (PLMS)	28
5.2	MODULAR STORAGE VAULTS (MSVs)	29
5.3	ENVIRONMENTAL CONTROL	29
5.4	STORAGE OF PARTIALLY DISMANTLED WEAPONS	29
5.5	USE OF PLASTICS IN URANIUM STORAGE	30
5.6	EVALUATION OF NEEDS	30
5.7	ON-SITE INSTRUMENTAL CAPABILITIES	30
6.	HAZARD ANALYSIS	31
6.1	HAZARD IDENTIFICATION	31
6.2	FUNDAMENTAL HAZARDS ASSOCIATED WITH MATERIAL FORMS	31
6.2.1	Criticality	31
6.2.2	Chemical Reactivity	31
6.2.3	Pressurization Due To Helium Generation	32
6.2.4	Pressurization Due to Radiolytic Reactivity	32
6.2.5	Pressurization Due To Hydrogen Evolution	32
6.2.6	Airborne Release Of Uranium	33
6.3	IMPACT OF MAJOR EXTERNAL PERTURBATIONS	34
6.3.1	Vulnerability to Natural Phenomena	34
6.3.2	Vulnerability to Aircraft Crash Scenarios	34
6.4	FIRES	34
6.4.1	Metals	34
6.4.2	Oxides	35
6.5	MATERIAL DISPERSAL	35
6.5.1	Material Dispersal In Fires	36
6.5.2	Explosion And Pressurization	36
6.5.3	Spills	36
6.6	HEALTH PHYSICS	37

7. CONCLUSIONS AND RECOMMENDATIONS FOR ENRICHED URANIUM STORAGE	
AT Y-12	38
7.1 MATERIALS ACCEPTANCE CRITERIA	38
7.1.1 Designations For Types Of EU Storage	38
7.1.2 Recommended Chemical Forms for Long-Term Storage	39
7.1.3 Recommended Physical Forms for Long-Term Storage	39
7.1.4 Acceptable Isotopic Ratios	40
7.1.5 Quality Assurance	40
7.2 TRANSIENT STORAGE (TS)	41
7.3 IN-PROCESS STORAGE (IPS)	41
7.4 INTERIM STORAGE (IS)	41
7.5 PROLONGED LOW-MAINTENANCE STORAGE (PLMS)	42
7.5.1 Storage Cans	42
7.5.2 Compartments And Storage Arrays	43
7.5.3 Long-Term Storage of Canned Subassemblies	43
7.6 STORAGE BUILDING FACILITY	43
7.6.1 Architectural Considerations	43
7.6.2 Robotics	43
7.6.3 Fire Safety	44
7.6.4 Inventory Verification	44
7.6.5 Criticality Posting	44
7.7 DATABASE MANAGEMENT	45
7.8 MATERIALS THAT DO NOT MEET ACCEPTANCE CRITERIA	45
7.8.1 Acceptable Chemical/Physical Forms for Long-Term Storage	46
7.8.2 Radiation Shielding	46
8. REFERENCES	47
Appendix	51

LIST OF TABLES

<u>Table</u>	<u>Title</u>	<u>Page</u>
1	Standard Terminology for Uranium Isotopic Assays	3
2	Count Rates Including Decay Products (Alpha and Beta)	6
3	Isotopic Blends. Alpha and Beta Activity	7
4	Cross Sections of the Uranium Isotopes to Thermal Neutrons	10
5	Fundamental Properties of Uranium Metal	11
6	Rates of Reaction of Uranium With Oxygen and Water Vapor	14
7	Thermodynamic Properties of Uranium Oxides	16
A-8	Participants from Outside of Y-12	51
A-9	Y-12 Internal Working Group	52
A-10	Federal Regulations that Pertain to Long-Term Uranium Storage Facilities	53
A-11	DOE Orders that Pertain to Long-Term Uranium Storage Facilities	54
A-12	Miscellaneous DOE Publications that Pertain to Uranium Storage Facilities	55
A-13	Nongovernmental Publications that Pertain to Long-Term Uranium Storage Facilities	55
A-14	Properties of the Uranium Isotopes	57
A-15	Decay Schemes of the Uranium Isotopes	58
A-16	Calculated Corrosion Rates for Standard Cast Uranium Cylinders	64

1. INTRODUCTION

This document is an assessment of the technical safety issues pertaining to the storage of EU at the Oak Ridge Y-12 Plant. The purpose of the assessment is to serve as the basis for defining the technical standards for storage of EU at Y-12.

A formal assessment of the Y-12 materials acceptance criteria for EU is currently being conducted by a task force cochaired by B. G. Eddy of DOE Oak Ridge Operations and S. O. Cox of Y-12 Defense Programs.⁴⁶ The mission of this technical assessment for storage is obviously dependent on results of the acceptance assessment. Clearly, the two efforts require coordination to avoid inconsistencies. In addition, both these Assessments must be consistent with the Environmental Assessment for EU storage at Y-12.¹

Both the Storage Assessment and the Criteria for Acceptance must take cognizance of the fact that a portion of the EU to be submitted for storage in the future is expected to be derived from foreign sources and to include previously irradiated uranium containing significant levels of transuranics, radioactive daughter products, and unstable uranium isotopes that do not occur in the EU stream of the DOE weapons complex. National security considerations may dictate that these materials be accepted despite the fact that they fail to conform to the Acceptance Criteria. This document will attempt to address the complexities inherent in this situation.

The eventual use of all the highly enriched uranium (HEU) being stored at Y-12 has not been established. The recommendations in this assessment will be made accordingly, without making assumptions as to what that end use will be or the length of time the material will remain in storage.

1.1 SCOPE

This Technical Assessment covers the storage of EU at the Oak Ridge Y-12 Plant. Included are (1) transient storage (TS), pertaining to the brief interval when it is first received at Y-12; (2) interim storage (IS) of material, prior to processing into the form desired for long-term storage; and (3) long-term storage, known as prolonged low-maintenance storage (PLMS). In-process storage is considered to be a manufacturing concern and is excluded from this assessment.

The results of the assessment were compiled as follows: (1) a review of fundamental properties that impact the definition of acceptable storage standards (Sections 2 and 3); (2) a review of current practices for EU storage (Section 4); (3) a discussion of advanced storage concepts (Section 5); (4) a formal hazards analysis (Section 6); (5) a compilation of relevant laws, regulations, DOE standards, non-DOE standards, and DOE orders (Appendix, Section A-1.3); and (6) the formal recommendations (Section 7) justified by the rational basis and data presented in the prior sections. The formal recommendations from the Technical Assessment document will serve as the basis for the standards for EU storage, to be compiled as a separate document.

Storage of depleted uranium (containing less than 0.713 percent ^{235}U) will not be included in this assessment but may be the subject of an assessment to be generated in the future. Also excluded from this assessment is the long-term storage of uranium in the form of canned subassemblies. This is considered to be a special case, with relevant safety issues being determined by the specific materials and the physical configuration of the particular type of weapon subassembly being placed in storage.

The focus of this assessment is on storage only. Related issues concerning handling the material are addressed only to the extent required to prepare the material for storage and to conduct routine inspections and investigations. The recommendations and eventual standards must be consistent with the proven methods, materials, and facilities that have been successfully used in the past. The standards must not be dependent upon the outcome of ongoing or future research and development programs.

1.2 PROGRAM PARTICIPATION

The names of the individuals from Y-12 and from other organizations who participated in the preparation of this report are shown in the Appendix, Section A-1.2.

1.3 IDENTIFICATION OF ORDERS, STANDARDS, AND DOCUMENTS

The DOE Orders and Standards and non-DOE standards that apply to the storage of EU are compiled in the Appendix, Section A-1.3.

2. NUCLEAR PROPERTIES THAT RELATE TO STORAGE

Nuclear properties of uranium have an impact on storage criteria in several ways: allowing the evaluation of radiation levels, guiding the formulation of acceptance criteria, permitting a numerical determination of heat generation due to radioactive decay, and making possible an evaluation of the importance of radiation-induced chemistry such as hydrogen generation due to the degradation of water or polymers.

2.1 URANIUM ISOTOPIC DISTRIBUTIONS

Naturally occurring uranium consists of a mixture of the three isotopes ^{238}U (99.275 percent abundance), ^{235}U (0.713 percent abundance), and ^{234}U (0.005 percent abundance).^{2m,3a,44} Weapons grade, HEU, known as or alloy (Oak Ridge alloy), contains approximately 93 percent ^{235}U , 6 percent ^{238}U , and 1 percent ^{234}U . Depleted uranium, which consists of the "tails" from isotopic enrichment processing, typically contains about 0.2 percent ^{235}U . Special grades of very HEU were formerly manufactured at the Portsmouth Gaseous Diffusion Plant with approximately 97.65 percent ^{235}U enrichment,^{2a} or even as high as 99 percent. Significant quantities of uranium having intermediate levels of isotopic enrichment are present in the nuclear power industry, in research reactors, and elsewhere. Table 1 summarizes the terminology and acronyms used to designate the enrichment levels of the categories of uranium that could be stored at Y-12. Unfortunately, the terminology is not universally accepted. In particular, the term HEU is often used to designate any material having a ^{235}U assay greater than 20 percent.

Table 1.
Standard Terminology for Uranium Isotopic Assays

<u>Designation</u>	<u>Acronym</u>	<u>^{235}U assay</u>
Depleted uranium	DU	<0.713%
Normal uranium	NU	0.713%
Low enriched uranium	LEU	0.713 to 20%
Moderately enriched uranium	MEU	20 to 85%
Highly enriched uranium	HEU	85 to 94%
Very highly enriched uranium	VHEU	>94%

2.2 RADIOACTIVE DECAY

2.2.1 Half-Lives of the Isotopes

In addition to the three natural isotopes, uranium has 12 artificial isotopes, not counting meso states.⁴⁴ Nine of these have half-lives so short that their existence at significant concentration levels is largely transient. However, materials recovered from nuclear reactor operations often contain high enough concentrations of the isotopes ²³²U, ²³³U, and ²³⁶U to require special handling provisions. These isotopes are formed in nuclear reactors due to neutron capture by thorium or ²³⁵U. The uranium in the nuclear weapons complex was historically isolated from irradiated material. Thus data for the isotopes ²³²U, ²³³U, and ²³⁶U are considered in this document only in order to address issues concerning acceptance criteria pertaining to materials partially contaminated by irradiated uranium, and not in anticipation that the Y-12 Site will ever be engaged in the storage of materials composed primarily of the isotopes ²³²U, ²³³U, and ²³⁶U. The current criteria for acceptance or rejection of incoming uranium by Y-12 are from the Egli Report.⁴⁵ A revised set of criteria for uranium acceptance at Y-12 is currently being prepared.⁴⁶

Half-lives and decay energies of the six uranium isotopes are given in Table A-14 of the Appendix. The number of disintegrations per second, or Becquerels, can be calculated from the formula:

$$\# \text{ Bq} = 1.322(10^{16}) \cdot M(g) / (\tau_{1/2} \cdot IW),$$

where $M(g)$ is the mass of the isotope in grams, $\tau_{1/2}$ is the half-life in years, and IW is the isotopic weight. The derivation of the formula is given in the Appendix, Section A-2.2.1. The power produced is simply the product of the number of disintegrations per second times the decay energy. Table A-14 in Appendix A-2.2.1 gives the decay rates and corresponding power levels for the six uranium isotopes. As far as storage of unirradiated EU is concerned, the more significant points of the data in Table A-14 are (1) most of the radioactivity originates with the ²³⁴U component, even though it is present in weapons grade material only to the extent of about 1 percent; and (2) the net power generation for isotopic blends containing only ²³⁵U, ²³⁸U, and small levels (1 to 2 percent or less) of ²³⁴U is very small, and provisions for heat dissipation are not needed. This is in marked contrast with the situation for the storage of more radioactive materials such as actinides and spent nuclear fuels, where removal of excess heat is necessary.

2.2.2 Decay Sequences

Apart from the nuclear properties of the uranium isotopes themselves, standards for storage of uranium must be formulated with cognizance of the types of radioactive decay products that are formed and their relevant properties. The nuclear decay sequences of the isotopes 238, 236, 235, 233, and 232 are listed in the Appendix, Section A-2.2.2 (Table A-15, parts A-E, respectively). Reference 47 is an excellent source for the energies of the alpha and beta particles and gamma rays emitted by the nuclear disintegrations and the energies of the secondary electrons that accompany

alpha decay. There is not a separate table for ^{234}U because it exists as a part of the ^{238}U decay chain. The latter portions of the decay sequences of ^{236}U and ^{232}U are identical, after they reach ^{228}Th . These decay schemes are also shown in schematic form in the Environmental Assessment.^{1a}

The ^{233}U and its associated sister isotope ^{232}U (both of which are synthetic isotopes formed by irradiation of natural thorium) present much more severe radiological hazards than any of the naturally occurring uranium isotopes.⁵ Similar to the Special Nuclear Material (SNM) version of their naturally occurring siblings, these synthetic uranium isotopes require a stable configuration to ensure nuclear safety and safeguards measures to protect and account for the material. However, ^{232}U (72-year half-life) and ^{233}U (1.6×10^5 year half-life) also require shielding features to address their higher specific alpha activities (10 million times higher and 4,375 times higher than ^{235}U , respectively). In addition to the short half-life of ^{232}U itself, the radiological problem is compounded by the short half-life of the sequential decay products ^{228}Th , ^{224}Ra , ^{220}Rn , ^{216}Po , ^{212}Pb , ^{212}Bi , and ^{208}Tl . Furthermore, they require massive biological shielding to protect personnel from the 2.6 MeV gamma emission of the ^{232}U daughter product ^{208}Tl .⁵ Because of the severe radiological hazards associated with ^{232}U , these materials must be handled in accordance with criteria specifically addressing the unique set of hazards that they present.

The isotope ^{236}U is formed by the neutron capture of ^{235}U . As noted previously, the latter portions of the decay schemes for ^{232}U and ^{236}U are identical. However, the long half-lives of ^{236}U and ^{232}Th (the product of the first decay event) cause the gamma flux of ^{236}U to be much lower than that of ^{232}U on a per gram basis during the period of time that the product of the first two decays, ^{228}Ra , is growing in.

2.2.3 Decay from Daughter Products

If the daughter products are also radioactive, they contribute to the observed radiation level. This is a significant contribution unless the half life of the daughter product is several orders of magnitude greater than the time that has elapsed since the material was last processed through chemical recovery. A reasonable estimate of the activity associated with daughter product decay is obtained by assuming the system has reached virtual steady state between formation and decay of the daughter product (known as the Secular equilibrium). In this case, the alpha and beta count rates of a material containing its daughter products is approximated by multiplying the count rate of the parent uranium isotope by the appropriate integers, $I(\alpha)$ and $I(\beta)$, corresponding to the number of alpha and beta decays associated with the initial disintegration. These integers are simply the number of nuclear decays that occur before a decay product is obtained that has a half life that is a few orders of magnitude greater than the period of time that has elapsed since the material was purified of decay products.

Referring to Tables A-15A through A-15E, it is seen that the appropriate multipliers are:

$$\begin{aligned} I(\alpha) &= 6, I(\beta) = 2 \text{ for } ^{232}\text{U}, \\ I(\alpha) &= 1 \text{ for } ^{233}\text{U}, \\ I(\alpha) &= 1 \text{ for } ^{234}\text{U}, \\ I(\alpha) &= 1, I(\beta) = 1 \text{ for } ^{235}\text{U}, \\ I(\alpha) &= 1 \text{ for } ^{236}\text{U}, \text{ and} \\ I(\alpha) &= 1, I(\beta) = 2 \text{ for } ^{238}\text{U}. \end{aligned}$$

Then the equilibrium alpha and beta count rates are found by multiplying the value from Table A-15 in the Appendix by the appropriate multipliers. Calculated values are shown in Table 2.

Table 2.
Count Rates Including Decay Products (Alpha and Beta)

Uranium Isotope	Multipliers		Count rates, Bq/g	
	Alpha	Beta	Alpha	Beta
^{232}U	6	2	$4.642(10^{12})$	$1.548(10^{12})$
^{233}U	1	0	$3.502(10^8)$	0
^{234}U	1	0	$2.305(10^8)$	0
^{235}U	1	1	$7.994(10^4)$	$7.994(10^4)$
^{236}U	1	0	$2.435(10^6)$	0
^{238}U	1	2	$1.243(10^4)$	$2.486(10^4)$

The net count rates for Orallloy, natural uranium, and depleted uranium are given in Table 3, based on taking appropriate linear combinations of values from Table 2. The count rates in Table 3 correspond to the net number of disintegrations that occur. Because many (in most cases, virtually all) of the alpha and beta particles are attenuated within the material matrix, the values in the table are much higher than those that would be measured for bulk metal, powder, or solutions. More details regarding shielding are given in Section 2.3. A generalized formula is given in reference 27b for calculating the activity of uranium having its 238, 235, and 234 isotopic concentrations fixed in the ratio predicted by diffusion theory.

Table 3.
Isotopic Blends
Alpha and Beta Activity

Material	Count Rates (Bq/g)			
	Pure Isotopes		Including Decay Products	
	Alpha	Beta	Alpha	Beta
Oralloy (a)	2.348(10 ⁶)	0	2.384(10 ⁶)	7.533(10 ⁴)
Natural U (b)	2.443(10 ⁴)	0	2.766(10 ⁴)	2.475(10 ⁴)
Depleted U (c)	1.487(10 ⁴)	0	1.486(10 ⁴)	2.497(10 ⁴)

2.3 ATTENUATION OF RADIOACTIVE EMISSIONS

This section addresses the following issues: (1) how much of the radiation from radioactive decay will escape from the uranium container, (2) how much of this radiation will be attenuated by chemisorbed water or other hydrogenous materials, and (3) what quantities of degradation products (particularly gases) will be produced by this type of interaction.

As a generalization, alpha and beta particles are readily attenuated by thin sheets of matter placed around the emitting source. On the other hand, a source of high-energy gamma rays must be shielded by thick walls of heavy (high atomic number) elements to provide sufficiently low radiation levels for personnel safety. Some gamma activity is associated with almost every nuclear disintegration, but those processes that emit very high energy gamma rays (e.g., greater than one million electron volts) require massive shielding.

2.3.1 Attenuation of Alpha Particles

Alpha particles are energetic helium nuclei; i.e., doubly charged ions having no electrons and a nucleus composed of two protons and two neutrons. As seen in Tables A-15A to A-15E, alpha emission is the initial decay mode for all the uranium isotopes and several of the subsequent steps as well. The alpha particles emitted in the radioactive decay of the uranium isotopes are fairly energetic, falling in the range 4.0 to 5.4 MeV. However, alpha particles are very readily attenuated as they pass through matter; they have a range of only 0.2 centimeter (cm) in air and much shorter values in condensed phases, such as 5(10⁻⁴) cm in water.^{48a} Thus, most of the alpha activity emitted by bulk uranium is self-absorbed before reaching the material boundary.

Alpha particles dissipate most of their energy by interaction with electrons of the absorbing medium, resulting in dissociation or excitation of the atoms and molecules in the medium.^{48a} Typically, about 35 eV is expended per ion pair produced.^{48a} Actually, about 60 to 80 percent of the ionization is a consequence of a secondary process in which energetic electrons produced by alpha interaction cause the follow-on ionization. Because the alpha particles are much heavier than the electrons with which they interact, their trajectory is nearly straight (the exception being the case where they are scattered by collision with a nucleus).

2.3.2 Attenuation of Gamma Rays (and X rays).

Unlike alpha or beta particles which dissipate their energy through a series of small interactions, gamma rays lose most of their energy through a single interaction.^{48b} Thus, some of the gamma photons that impinge on an absorber are totally absorbed while others are transmitted at their initial photon energy. The intensity of the transmitted beam is given by Beer's law,

$$I = I_0 e^{-\mu d},$$

where I is the intensity of the beam transmitted through the absorbing medium, I_0 is the intensity of the beam entering the absorber, d is the thickness of the absorber through which the beam passes, and μ is the absorption coefficient of the medium.

Three modes of interaction of gamma rays (and less energetic photons such as X rays) are photoelectric absorption, Compton scattering, and pair production. These are described individually.

Photoelectric Absorption

For gamma photons of relatively low energy, interaction with orbital electrons occurs primarily by total transfer of the photon energy to the electron. Absorption occurs most readily when the photon energy is slightly greater than the binding energy of a particular electron orbit; the probability of absorption decreases with increasing photon energy beyond that level. Ions produced by photoelectric absorption are typically in unstable, excited states that undergo electronic transitions in which electrons move from loosely bound outer orbitals to more tightly bound inner orbitals. This process may be radiationless or it may involve emission of electromagnetic radiation, known as fluorescence, which is typically ultraviolet or visible light. Materials of high atomic numbers (high-Z materials) are more effective in absorbing radiation than low-Z materials.^{48c, 49}

Compton Scattering

The term *Compton scattering* refers to the interaction of a high-energy photon (typically greater than 100 eV) with the outer electrons in the atoms of the irradiated media. Each such interaction results in a loss of a portion of the photon energy and a deflection of the photon. The dose absorbed is essentially independent of the atomic number of the atoms in the medium.^{48d, 49}

Pair Production

Very-high-energy photons can interact with the electric field of a nucleus in such a way that the photon energy is converted into mass in the form of an electron-positron pair. The rest mass of an electron is $9.109(10^{-28})$ g, so the minimum photon energy required for pair production ($E = mc^2$) is $2 \cdot 9.109(10^{-28})\text{g} \cdot [3(10^{10})\text{cm s}^{-1}]^2 = 1.640(10^{-6})\text{erg} = 1.023(10^6)\text{eV}$, or about one MeV. The photon energy in excess of this amount is shared equally between the electron and positron in the form of translational energy.^{48h, 49} The excess energy of the positron is dissipated by interactions with the irradiated medium until it is annihilated by combination with an electron, producing "annihilation radiation" of energy 0.51MeV (one-half the minimum $E = mc^2$ energy calculated above).

Pair production is not a consideration in the direct radioactive decay of the naturally occurring uranium isotopes because the gamma energies are significantly below the 1.02-MeV threshold.

2.3.3 Attenuation of Electrons (Including Beta Particles)

Free electrons are generated during radioactive decay, not only as the direct result of beta decay but also in conjunction with alpha decay and due to photoelectric absorption and Compton scattering by gamma rays (as noted in Section 2.3.2). Unlike alpha emission, beta particles are not emitted at discrete energy values. The energies of beta particles are distributed from zero up to a set of maximum levels, characteristic of the source isotope.^{48j} The penetrating power of electrons is in general greater than that of alpha particles but much lower than that of gamma radiation. Beta particles typically travel about 400 cm in air or 0.5 cm in water.^{48j} The typical energy loss per ion-pair formation by beta particles is about the same as that for alpha particles, 35eV.^{48e} Chlorides have a particularly high absorptivity for electrons, and this property is the basis for the use of electron capture detectors in gas chromatography for quantitative analysis of chlorinated organic compounds such as polychlorinated biphenyls and common insecticides.

2.3.4 Generation and Attenuation of Neutrons

Neutrons are not directly produced during the radioactive decay of any of the uranium isotopes or the sequential decays. However, neutron generation can occur as a result of the alpha-neutron (α -n) reactions, in which alpha particles interact with light isotopes such as ^9Be , ^6Li , ^7Li , ^9Be , ^{10}B , ^{11}B , ^{19}F , ^{27}Al , and ^{28}Si .^{50,51} Specific instances where this may be of importance in uranium storage include (1) storage of uranium-beryllium alloys, (2) storage of uranium-aluminum alloys, and (3) storage of UF_4 (green salt, an intermediate in uranium manufacture). It is also a consideration if a fluorinated polymer such as Polytetrafluoro ethylene (commonly called Teflon) is used to store uranium.

Neutron attenuation is qualitatively different from α , β , or γ radiation because neutrons interact almost exclusively with nuclei rather than with electrons.^{48f} Elastic scattering is the primary method of interaction between fast neutrons (say, 10 KeV to 10 MeV) and matter and is significant for

intermediate energy neutrons (from thermal range to 10KeV).^{48f} Due to classical mechanics (conservation of energy and momentum), the fraction of translational energy transferred from a neutron to a nucleus is higher for hydrogen than for heavier nuclei. Thus, in hydrogenous substances such as organic polymers, water, and cell tissue, elastic scattering (or moderating) is a dominant mechanism for energy transfer from neutrons.^{48f}

2.4 CRITICALITY CONSIDERATIONS

The most important consideration for the storage of isotopically EU is eliminating the possibility of allowing the material to reach a configuration that would result in a sustained fission reaction (criticality incident). Table 4 summarizes the cross sections of the uranium isotopes to thermal neutrons. From the point of view of criticality prevention, the most significant data in the Table are the fission cross sections, σ_f . The values show that ²³⁵U is the dominant isotope in determining the susceptibility to criticality of a mixture of the naturally occurring isotopes. The ²³³U also is seen to have a high cross section to thermal neutrons.

Table 4.
Cross Sections of the Uranium Isotopes to Thermal Neutrons (a)

Isotope	σ_γ (b)	σ_f (c)	σ_s (d)	σ_t (e)
²³² U	73.1 b	75.2 b	14.7 b	163 b
²³³ U	47.7 b	531.1 b	8.2 b	587.0 b
²³⁴ U	100.2 b	<0.65 b	12.4 b	112.4 b
²³⁵ U	98.6 b	582.2 b	13.8 b	694.6 b
²³⁶ U	5.2 b			
²³⁸ U	2.70 b	-----	8.90 b	11.60 b

a) Reference 4. All cross sections are given in units of barns, b, (10⁻²⁴) cm².

b) σ_γ = neutron radiative capture cross section.

c) σ_f = fission cross section.

d) σ_s = "free" neutron scattering cross section.

e) σ_t = total neutron cross section.

In addition to the isotopic ratios, the criticality limit of a system containing fissionable material is determined by the net mass of fissionable isotopes; the physical configuration of a single unit and the necessary spacing between units; the type, quantity, and distribution of moderators (low-mass nuclei that reduce the speed of neutrons to thermal velocities) in the system; and the efficiency of neutron reflectors around the system, if any.

3. CHEMICAL AND PHYSICAL PROPERTIES AND STORAGE APPLICATIONS

3.1 URANIUM METAL

Uranium is a very dense, lustrous metal. It is ductile and malleable.⁴⁴ Table 5 lists some of the basic physical properties of pure uranium metal. Compared with other metals commonly encountered in the workplace, uranium is relatively high melting and nonvolatile. It undergoes two solid-solid phase transformations during heating to the melting point.

Table 5.
Fundamental Properties of Uranium Metal

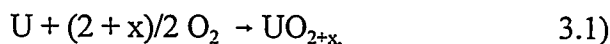
Property		Reference
Melting point	1132.8 (°C)	2c
Vapor Pressure (1720-2340K)		
Log (p/atm) = -26210/T(K) + 5.92		2c,6,7
Vapor Pressure (1480-2420K)		
Log (p/atm) = -25230/T(K) + 5.71		2c,6,8
Density (alpha phase @ 25° C)	19.07 g/cm ³	2c,6
Phase transformations		
(alpha-to-beta)	668°C	2c,6,44
(beta-to-gamma)	775°C	2c,6,44
Crystal modifications		
alpha	orthorhombic	44
beta	tetragonal	44
gamma	bcc	44
Heat Capacity (298-941K)		
Cp(J·K ⁻¹ ·mol ⁻¹) = 26.92 - 2.502(10 ⁻³)T		9,44
+ 2.656(10 ²)T ² - 7.699(10 ⁻⁴)T ⁻²		
Enthalpy of Fusion	83.4 J/g	10
Enthalpy of Vaporization	1883 J/g	10
Thermal Conductivity (70°C)	0.29 J mol ⁻¹ s ⁻¹ K ⁻¹	2c,11
Electrical Conductivity	(3.4)(10 ⁻⁴)Ω ⁻¹ cm ⁻¹	2c,6

High-purity uranium metal is routinely manufactured by metallothermic reduction of UF_4 with calcium or magnesium. Fabrication of the metal by casting and various forging techniques such as extruding and rolling are mature technologies.²⁵ When isotopically enriched material is involved, sophisticated facility designs and operating procedures are required to guarantee that a criticality incident cannot occur.

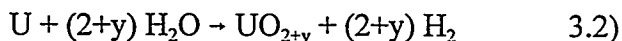
3.1.1 Storage Implications of the Corrosion of Uranium Metal

Corrosion of uranium metal during storage is a problem for two primary reasons: (1) the resulting oxide surface layer can become airborne under unfavorable circumstances, resulting in loss of fissile material and an inhalation hazard; and (2) the resulting hydrogen evolution could cause container pressurization and poses a fire and explosion hazard.

The fundamental chemistry underlying uranium corrosion has been investigated in considerable detail^{20,34} and several excellent reviews have been compiled.^{20,34a,34f,34n,34p,34q} The corrosion reaction of uranium with oxygen proceeds according to the equation:



where x is in the range 0.2 to 0.4 at temperatures below 200°C.²⁰
The corrosion reaction of uranium with water is:



where y is between 0.0 and 0.1.²⁰ No detectable hydrogen is formed in the presence of oxygen except under saturated conditions^{20,34h,34j,34k,34l,34m} (saturated conditions meaning conditions that enable water to condense on the metal surface and thereby limit the transport of oxygen from the gas phase to the metal surface). Hydrogen generation is therefore avoided by maintaining uranium metal under conditions that preclude high temperatures (above 200°) and high humidities (greater than 90 percent RH). Examples of conditions that should be avoided to prevent hydrogen evolution from uranium metal are the following:

1. storage out of doors or in damp indoor environments that permit condensation on the metal surface to occur;
2. storage of wet sludges and saw fines; and
3. storage of wet or submerged machine chips and turnings.

The rate at which hydrogen is formed due to uranium corrosion is strongly influenced by the following factors:

1. Specific surface area. The rate of corrosion is proportional to the surface area. Highly pitted surfaces corrode much more rapidly than polished metal.
2. Galvanic effects. Chloride ion in particular causes uranium to corrode at a greatly accelerated rate. Furthermore, deliquescent salts present on the surface of uranium metal will cause locally anaerobic conditions to exist, and thus permit hydrogen evolution.
3. pH effects. Increased acidity accelerates corrosion and hydrogen evolution.
4. Temperature.
5. Presence or absence of mechanical stresses.

The corrosion rate of uranium exposed to condensing-water environments is considerably higher than in relatively dry air. In order to survive long-term storage, the uranium must be kept dry. Moisture cannot be allowed to accumulate on the stored metal or in the container. Storage of unprotected uranium metal in a heated building with humidity control would prevent moisture from condensing on unprotected parts, whereas storage in sealed cans will prevent contact with atmospheric water vapor altogether. Significant corrosion will occur, however, in the presence of water vapor, especially in the absence of oxygen. Exposure to water-free oxygen results in oxide growth kinetics proportional to the square root of time.

Atmospheric corrosion of uranium metal is dependent upon several factors, and a brief discussion of these effects will follow. The mathematical models used in this discussion were developed by A.G. Ritchie, who compiled the results of numerous investigators.²⁰ These models are in good agreement with the several corrosion investigations that have been conducted over the years at Y-12 and elsewhere. Uranium corrosion can be best described quantitatively by defining four categories of environments: (1) dry air, (2) water vapor at 100 percent relative humidity (RH) with no oxygen present, (3) water vapor at 100 percent RH in typical atmospheric air (21 percent O₂), and (4) water vapor at 2 to 90 percent in the presence of oxygen. The equations for Ritchie's derived models are presented in Table 6.

Calculated corrosion rates for cast uranium cylindrical shells are given in Table A-16 of Section A-3.1.1 of the Appendix, based on the rate expressions from Table 6. The physical dimensions of the cylinders are those selected for long-term HEU storage, to be discussed later (Section 4.2.). Five generalizations should be noted from the models:

- The rates of corrosion in dry air and in normal air (with relative humidities below 90 percent) are about the same.
- Corrosion rates at 100 percent RH in normal air are greater than those in dry air or <90 percent RH air by approximately an order of magnitude.

- Corrosion rates at 100 percent RH in the absence of O₂ are approximately one additional order of magnitude higher.
- When uranium cylinders are stored at ambient temperatures under noncondensing humidities, corrosion rates are very low (ca. 0.0014 percent per year).
- Temperature has a large impact on corrosion rates.

Table 6.
Rates of Reaction of Uranium With Oxygen and Water Vapor (a)

Environment	Rate (b)	Temperature range °C	Rate at 298 K (b)
Dry air	$6.9 \times 10^8 \exp(-18,300/RT)$	40-300	$2.61(10^{-5})$
Water Vapor (100% RH, no O ₂)	$3.2 \times 10^8 \exp(-13,800/RT)$	20-100	$2.42(10^{-2})$
O ₂ -water vapor (100% RH)	$4.6 \times 10^9 \exp(-17,800/RT)$	25-100	$4.05(10^{-4})$
O ₂ -water vapor (2-90% RH)	$4.8 \times 10^{13} \exp(-25,000/RT)$	40-130	$2.21(10^{-5})$

(a) Reference 20.

(b) R in units of cal/mol K, Temperature in K, and rate in milligrams of uranium reacted per square centimeter of surface per hour.

The question of storing the uranium in a dry inert gas environment versus a dry air environment often arises. Undeniably, the storage of uranium metal in a pure inert gas leads to no corrosion. However, the rates of corrosion in dry air or in air having an RH of <90 percent are also very small (about 0.25 g per year for an 18-kilogram [kg] part). Consequently, very little is really gained by storing the part in an inert gas. Furthermore, as shown by comparison of the two models pertaining to 100 percent RH (See Appendix, Table A-16), the water corrosion of uranium, particularly at higher temperatures, is sharply retarded by the presence of oxygen. Thus, if some circumstance occurred that caused enough water to be present in the storage vessel to reach the saturation vapor pressure, the presence of oxygen would greatly retard the rate of corrosion.

The importance of preventing a condensing environment is shown by comparing the data in the two right-hand columns of Table A-16 (Appendix). The corrosion rate in 100 percent relative humidity air is considerably higher than in air at 2 to 90 percent RH. For example, at 85°F (29.4°C) the corrosion rate in 100 percent RH is approximately ten times higher than air at <90 percent humidity.

It is a useful rule of thumb that relative humidity values less than 55 percent yield significantly lower corrosion rates, due to the absence of multilayer water adsorption.¹⁹

The atmospheric corrosion rate is exponentially dependent upon temperature for all of the four models. The relative impact of temperature depends on the exponential portion of the rate expressions. Thus, Table 6 indicates that the impact of temperature is greatest for relative humidities of 2 to 90 percent in the presence of oxygen and lowest for 100 percent RH in the absence of oxygen.

The four models apply to controlled conditions, as specified in Table 6. They are also useful for estimating corrosion rates under real world conditions. For example, uranium metal parts stored in underground moist conditions for periods of approximately 30 years were observed to have corroded at a rate of about 0.001 inch (2.54×10^{-3} centimeter [cm]) per year.¹⁶ This corresponds to about 5.5×10^{-3} milligram (mg) $\text{cm}^{-2} \text{h}^{-1}$, which lies between the calculated corrosion rates for the second and third cases listed in Table 6, pure water vapor and O_2 -water vapor. However, the models must be used intelligently. In a perfectly sealed container, only the gases within the free volume of the container impact the calculations, and the rate of corrosion will approach zero asymptotically as the concentration of the rate-limiting reactive gas decreases. In the case of slow leaks, rates of diffusion of O_2 and H_2O into the vessel are expected to be the primary factors governing the net corrosion rate.

3.1.2 Alloys, Intermetallics, and Traced Metal

Uranium shows only limited solid solubility with most other metals. This is attributed to the fact that the crystal structures of both α - and β - uranium are significantly different than most other metals^{2d}. Intermetallic compounds are formed with Al, As, Au, B, Be, Bi, Cd, Co, Cu, Fe, Ga, Ge, Hg, Ir, Mn, Ni, Os, Pb, Pd, Pt, Rh, Ru, Sb, and Sn^{2d}. Uranium alloys that are known to have a significant usage within the DOE complex include those with Al, Fe, Mo, Nb, Ti, and Zr. Small quantities of many other uranium alloys are in use or in interim storage at various DOE installations, if only in the quantities needed for research purposes. The utility of the alloys are (1) pure uranium is structurally a weak metal, and alloying confers superior mechanical properties; (2) pure uranium is highly electropositive and thus vulnerable to corrosion, and some of the alloys show superior corrosion resistance; and (3) lower-melting intermetallics are a convenient form in which to collect uranium from metallothermic or electrochemical reduction operations. Over-aged U-6 percent Nb and U-0.8 percent Ti contain the intermetallics Nb_2U and U_2Ti , respectively, distributed throughout the material.

3.2 OXIDES

The uranium-oxygen system is not only the most complex actinide oxide system but is also one of the most complex oxide systems known.^{2e} A listing of binary oxides that have been reported^{2f} includes UO , U_2O_3 , UO_2 , U_4O_9 , $\text{U}_{16}\text{O}_{37}$, U_3O_7 , U_8O_{19} , U_2O_5 , U_5O_{13} , $\text{U}_{13}\text{O}_{34}$, U_8O_{21} , $\text{U}_{11}\text{O}_{29}$, U_3O_8 , $\text{U}_{12}\text{O}_{35}$, and UO_3 , but the authenticity of some of these phases is dubious. Multiple crystalline modifications of some of these phases have been reported also. The peroxide, UO_4 , appears to exist only in the hydrated state. Many of these phases, especially UO_2 , can exist in nonstoichiometric

form with variable oxygen ratios. The higher oxides, as well as many other uranium compounds, decompose to U_3O_8 above 650°C .^{28,29} This, plus the fact that oxygen lost from U_3O_8 above 800°C is replaced rapidly during cooling in air are the reasons that U_3O_8 is the form chosen for the gravimetric analysis of uranium.^{28,29}

The three oxides that can be prepared in good purity by well-established procedures are UO_2 , U_3O_8 , and UO_3 . The U_3O_8 and UO_3 are stable under ambient conditions, and UO_2 is also very stable except when it is in the form of a finely divided powder which can undergo spontaneous oxidation in normal air to form the higher oxides. The various oxides can be interconverted by exposure to oxidizing or reducing atmospheres at elevated temperatures. Thermodynamic data for the three oxides are given in Table 7. Two points of particular significance should be noted from the data: (1) the enthalpies and free energies of formation of the oxides are very high (large negative values), demonstrating the great thermodynamic stability of the compounds; and (2) the incremental changes in the free energy and enthalpy values are relatively small in proceeding from UO_2 to the higher oxides. The latter observation shows that the high chemical affinity of uranium for oxygen is manifested most strongly in reaching the tetravalent state (UO_2) and is significantly lower in proceeding to higher oxidation states.

Table 7.
Thermodynamic Properties of Uranium Oxides (a,b)

	$\Delta G^\circ_{f,298}$ kJ/mol U	$\Delta H^\circ_{f,298}$ kJ/mol U	S°_{298} j/K mol U	C_p (298°K) j/K mol U
UO_2	-1031.83	-1085.0	77.03	63.60
U_3O_8	-1123.16	-1191.6	94.18	79.31
γ - UO_3	-1145.74	-1223.8	96.11	81.67

(a) Reference 9.

(b) Note: The compiled values pertain to the quantity of compound formed from a single mole of uranium metal. Thus, the free energy of formation of one mole of U_3O_8 is three times as great as the tabulated value.

At high temperatures UO_2 shows a wide homogeneity range, from $UO_{1.63}$ to $UO_{2.25}$. Formation at ambient temperatures yields a range of $UO_{2.00}$ to $UO_{2.25}$, depending on the relative oxygen/water content of the environment. It has been shown experimentally that UO_2 can be converted to pure metal by the Saltless Direct Oxide Reduction process.

The U_3O_8 is the principal oxide formed by the corrosion of uranium metal under ambient atmospheric conditions (unrestricted access to oxygen). The UO_3 is routinely prepared as an intermediate in the manufacture of uranium metal by the thermal decomposition of uranyl nitrate hexahydrate or uranium peroxide. A serious disadvantage to storing UO_3 is the fact that it is very hygroscopic^{12,30} and forms a stable monohydrate. In water, conversion to the monohydrate takes only 24 hours.³⁰ This poses a serious difficulty in establishing the precise amount of uranium for accountability, both in terms of obtaining a truly representative sample for analysis and of preventing drifts in the net weight prior to the time the material is sealed into its storage container.

3.3 FLUORIDES

Uranium tetrafluoride is an important intermediate in the manufacture of uranium metal. It is readily prepared by precipitation from aqueous solutions of tetravalent uranium salts, but a product that is better suited for bomb reduction is obtained by the treatment of UO_2 with anhydrous hydrogen fluoride at high temperatures.^{2h,3a,14} The melting point of UF_4 is 960 °C and the crystal system is monoclinic with a theoretical density of 6.70 g/cm³. It is described as being nonvolatile, nonhygroscopic, and highly insoluble in water.²ⁿ Therefore, from the point of view of chemical and physical properties, UF_4 is a reasonable form in which to store uranium for a long period of time. However, UF_4 is observed to evolve fluoride ions, perhaps from chemisorbed hydrogen fluoride (HF), and the fluoride promotes significant corrosion.¹⁵ This is the most serious disadvantage to the long-term storage of UF_4 . The conversion of UF_4 (known as green salt) to the metal is readily accomplished by bomb reduction with calcium or magnesium; the resultant metal is more suitable for long-term storage because of its higher uranium atom density. In order to achieve consistently high levels of efficiency in the conversion of UF_4 to the metal, the UF_4 must be of high purity with respect to oxide and hydrate content. An effective means of minimizing the degradation of the salt during long-term storage would be to melt the salt in an inert container, thus obtaining a minimal specific surface area.¹⁶ If the UF_4 is seriously degraded during long-term storage, it could readily be treated to be made suitable for bomb reduction after any time lapse or condition of storage.¹⁶ The decision as to whether UF_4 is to be converted to metal for long-term storage is necessarily dependent on making an educated guess as to the end use to which the material will be put at the end of the storage interval.

A problem that is encountered in the processing of the fluorides of some actinides, such as plutonium, is the exposure of workers to neutrons due to the α -n (alpha-neutron) interaction with fluoride. The problem is much less severe when dealing with HEU containing ^{235}U at a concentration of 1 percent, simply because the net alpha flux is several orders of magnitude lower in the uranium case.

Uranium hexafluoride is a volatile solid that sublimates at atmospheric pressure [$1.013(10^5)$ Pa] at 56.5°C and has a triple point at 64.02°C with a pressure of 1134 mm Hg [$1.512(10^5)$ Pa]. The vapor pressure of UF_6 at ambient temperature is about 120 mm Hg [$1.600(10^4)$ Pa]. It is usually prepared by the direct fluorination of UF_4 . Because of its high vapor pressure, UF_6 has to be stored in gas-tight, corrosion-resistant canisters. Ordinary mild steel is satisfactory at ambient temperature if it is protected from the environment.¹⁶ The storage of isotopically depleted UF_6 in 14-ton steel

cylinders is a mature technology, and 555,000 metric tons of UF_6 are currently stored in this manner.³¹ Although the long-term safety record for this practice is good, there have been instances where leakage has occurred, primarily due to corrosion of the cylinders.³¹ The corrosion has been demonstrated to be a slow process in terms of time frames estimated at 30 years,³² 50 years,¹⁶ or 70 years.³³

The UF_6 is highly soluble and reactive in water (decomposing to UO_2F_2) and can be absorbed through the skin as UO_2F_2 .¹⁷ The most positive attribute of uranium hexafluoride as a material for long-term storage is its immediate availability for gas-phase enrichment processes or for blending down to assays suitable for reactor applications.

An established technology exists for conversion of UF_6 to UF_4 by reduction with hydrogen, and a newly redesigned facility for carrying out the process is in place at the Y-12 Plant.

The fluorides UF_3 , U_2F_9 , U_4F_{17} , and UF_5 have been described in the technical literature.^{2j,44} In addition, a large number of uranium-containing ternary fluorides have been reported. Some of these may have a combination of properties that would make them suitable for long-term storage, but none appear to be present in the DOE complex in appreciable quantities or are clearly superior to UF_4 for long-term storage.

3.4 OTHER HALIDES AND OXYHALIDES

A large volume of information has been published concerning the chlorides, bromides, and iodides of uranium and the corresponding oxyhalides.^{2k,44} Small quantities of many of these compounds exist within the DOE complex as research materials, analytical samples, or degradation products. The corrosive nature of these halides and their affinity for moisture makes them unattractive for long-term storage.

3.5 NITRATES

Uranyl nitrate hexahydrate [$\text{UO}_2(\text{NO}_3)_2 \cdot 6 \text{H}_2\text{O}$] is an important intermediate in the purification of uranium by solvent extraction. It stores well in sealed containers. The material is not an attractive candidate for long-term storage due to (1) anticipated weight fluctuations due to hydration-dehydration equilibria, which would limit the precision attainable for inventory purposes; (2) handling procedures would be complicated by the fact that the material cannot be fabricated into a tough, cohesive body; (3) the material would be corrosive toward some attractive container materials over long time spans; and (4) uranyl nitrate poses a handling problem because it can be absorbed through the skin.¹⁷

3.6 HYDRIDE

Uranium metal reacts with hydrogen to form uranium hydride (UH_3). The kinetics of the hydriding and dehydriding reactions have been studied in great detail,¹⁸ including the determination of the effects of diffusion time for hydrogen through the metal matrix and the impact of oxygen as an impurity. Uranium hydride does not corrode in pure water at an appreciable rate at ambient temperatures, but oxidizes in the presence of oxygen.¹⁶ Uranium hydride corrodes in an air-water environment at about ten times the rate of uranium metal under the same conditions.¹⁹

Due to its susceptibility to corrosion to uranium oxides plus hydrogen and the hazards associated with hydrogen evolution, uranium hydride is not a viable material for long-term storage under moist and oxidizing conditions. A knowledge of the role played by uranium hydride in the corrosion of uranium metal is valuable for purposes of designing a suitable metal storage system.

3.7 OTHER COMPOUNDS

Uranium forms a vast number of additional compounds including simple inorganic salts, salts derived from organic acids, complexes, alloys, intermetallics, and organometallics. Most of these have been prepared only in small quantities for research purposes. Although some of these compounds may be chemically stable, the methodology that would be required for maintaining a storage facility containing a large number of different chemical forms of uranium would be costly to develop and administer. Conversion to the metal or oxide for long-term storage is the preferable option.

3.8 BASIS FOR SELECTING SUITABLE CHEMICAL FORMS FOR LONG-TERM STORAGE

The chemical forms of uranium to be declared acceptable for long-term storage were selected by eliminating the following classes of materials:

- Gases and volatile condensed phases, due to the potential for loss by leakage.
- Liquids, which also might be vulnerable to leakage.
- Corrosive materials suspected of degrading the integrity of the primary container over long time periods.
- Chemical forms that are difficult to isolate in pure state and those having variable or questionable stoichiometry.
- Health hazards (beyond those intrinsic to uranium).
- Fire hazards.

- Uncommon (small potential inventory) materials. These are considered undesirable simply because the additional complications involved in accommodating these materials would outweigh the effort involved in converting them to a more common chemical form.
- Low uranium content. These are undesirable because they make an uneconomical use of space.

On the basis of these considerations and the properties described in the preceding paragraphs, two chemical forms were identified as being desirable for storage: metal and triuranium octoxide (U_3O_8). The preferred physical form for the metal is a near-full-density casting, primarily to minimize the corrosion rate and the fire hazard. The triuranium octoxide is most readily obtained as a free-flowing powder, and it should be stored in that form. In accordance with the principal that the recommendations and standards be formulated without biasing the operation in favor of anticipated end uses (Section 1), the form selected for storage should be that which is most readily obtained from the material in the as-received form.

From the discussion in Section 3.3, a reasonable justification exists for designating UF_4 as an acceptable form for long-term storage as well. The reasons for not doing so were (1) UF_4 is readily converted to the metal by bomb reduction; (2) unless due care is exercised to prevent moisture exposure, the UF_4 will degrade by hydrolysis, with a resulting diminished yield if the material is eventually converted to metal; (3) current inventories of highly enriched UF_4 in the DOE complex are rather low; (4) UF_4 is known to be rather corrosive toward most potential containment materials, probably due to the presence of a surface layer of chemisorbed HF; and (5) fluorides interact with alpha particles according to an (α, n) interaction, yielding a neutron flux that would elevate the background radiation level in the storage facility.

4. CURRENT STORAGE PRACTICES

The EU storage practices that have evolved at Y-12 have historically been driven by short-term objectives. A relatively small portion of the net DOE inventory of HEU would typically be in storage at a given time, and the mean storage time was fairly short. Therefore, the existing storage practices are a source of guidance for planning the PLMS facilities, but the practices should be modified to account for the greatly increased quantities of uranium to be accommodated and the reasonable likelihood that storage times will be much longer than were typical when Y-12 was in high-throughput production mode.

4.1 TRADITIONAL DEFINITIONS OF STORAGE INTERVALS

It is important to make a differentiation between long-term storage and other forms of storage. Prolonged Low-Maintenance Storage (PLMS) is defined as storage of material that has no currently defined near-term use under DOE or other programs. For the purpose of this assessment, the material will be assumed to be going into storage for an indefinite period of time. Because the future uses/needs for nuclear materials have not been established, it is not possible to predict which storage form will be most useful. The objectives of a PLMS facility are consolidation, low maintenance, stability, and limited access. A PLMS facility is designed for limited personnel access and low Nuclear Material Control and Accountability (NMC&A) overhead. These requirements must be reconciled to comply with commitments to comply with, or exceed, International Atomic Energy Agency (IAEA) requirements to provide "transparency," which is the term used to designate the ease and reliability with which the SNM material inventory can be verified. The goal of material containment is to ensure that the material will be in essentially the same condition when the container is opened as when the material was first stored.

Interim Storage (IS) was traditionally defined as inactive material stored for a defined use at some time in the future. Inventories requiring periodic surveillance, maintenance, and accessibility were the primary candidates for IS when the Y-12 Plant was actively manufacturing nuclear weapons components. As weapons components are returned from field sites, it is expected that a large proportion of the uranium inventory in IS will be material being held for conversion to a form suitable for PLMS. The IS facilities are more frequently accessed than PLMS facilities. The goal of material containment is to ensure that loss or diversion of nuclear material can be prevented and that incidental spills can be localized to a specific area.

In-process storage (IPS) or lag storage is best described as a working vault. Access to a working vault may be daily. Material appropriate for in-process storage is active material described as "in use." The NMC&A overhead associated with in-process storage is high because of frequency of access. In-process storage is not the focus of this technical assessment because it is primarily linked with processing operations.

Due to the change in mission for the Y-12 Plant, it seems prudent that an additional category of storage be defined, namely TS. This designation will apply to material at the time it arrives at the Y-12 Plant. While in TS the uranium is contained in the shipping container in which it was received. The duration of time during which any as-received material is held in TS should be minimized.

Some forms of material may be unsuitable for PLMS due to reactivity, worker safety, or public risk. Other material forms may be uneconomical to prepare for PLMS. Unsuitable forms of material will require stabilization or conversion to material forms suitable for PLMS. Processing capabilities that need to be retained for stabilization and conversion should be identified. Without a major production requirement, most future processing capabilities will be focused on stabilization, volume reduction, and waste minimization rather than on purification.

4.2 PREPARATION OF URANIUM FOR STORAGE

In the past, most HEU was stored as broken metal, solid metal shapes, cylindrical annuli, and oxides. Cylindrical annuli are the preferred storage form in terms of criticality safety and corrosion minimization. Current practice at Y-12 involves casting the uranium into hollow right-circular cylinders having an inner radius of approximately 1.75 inch (4.45 cm) and an outer radius of approximately 2.5 inch (6.35 cm). The typical mass of a routine casting is 18 kilogram (kg) and the maximum allowable quantity is 20 kg. For an 18-kg casting of 93 percent EU, this corresponds to a cylinder height of 14.8 cm and a volume of 956 cm³.

The process that was routinely used²⁸ for corrosion protection of newly fabricated EU weapon components involved coating the parts with a water-displacing, dry film, and non-RCRA waste "rust-preventing" oil such as Rust Veto 4214W. This coating will eventually fail, but it will provide corrosion protection for five years or more. This does not appear to be a desirable option for material to be placed in PLMS because (1) storage times may be considerably greater than five years; (2) flammable organic oils pose a small, but real, fire hazard; and (3) surface blemishes caused by corrosion are a source of serious concern on weapon components, which have stringent specifications for certification but which are of no serious consequence in the case of metal in storage.

Each part is to be assigned a job number or serial number. The containers should be designed to facilitate sampling or in-container monitoring. The gross, tare, and net weights should be recorded and entered on the batch cards and into the database. Material items must be accessible for periodic inventory unless attributes can be monitored remotely.

4.3 IDENTIFICATION OF STORAGE AREAS AT Y-12

All HEU is stored in Material Access Areas (MAAs) or other secure locations within the plant's Protected Area. An MAA is a controlled security area that segregates EU use or storage from other operations areas by physical barriers and specific access controls.^{1g}

The principal storage facilities for high-enrichment uranium (HEU) currently within the DOE complex are the vaults and cages in Y-12 storage facilities. Specialized storage areas at the plant include: (a) tube vaults, (b) modular storage vaults, (c) in-process storage vaults, (d) overnight storage vaults, (e) rack storage vaults for small container storage (typically bird cages), and (f) the fuel element storage vaults.

4.4 PROLONGED LOW-MAINTENANCE STORAGE (PLMS) FACILITIES

4.4.1 Tube Vaults

The configuration and design criteria for the tube vaults are dictated by nuclear criticality safety requirements. The vault walls, floor, and ceiling are constructed of massive high-density, reinforced concrete. The two opposite walls of each tube vault have arrays of 5 (vertical) \times 20 (horizontal) tubes, or 200 tubes per vault. A tube is a horizontal square prism hole in the vault wall, 8 inches tall by 8 inches wide by 12 feet deep. The tubes are steel lined. Ten uranium storage cans can be arrayed horizontally on a tray inside each tube, so a tube vault has a capacity for 2,000 cans. Access to each tube is blocked by a vertical steel bar. Verification that the seal has not been broken and isotopic analysis by nondestructive analysis (NDA) of a small sample is adequate for routine inventories. There are no fire sprinklers inside the vaults to avoid possible criticality ramifications.

Each tube holds a 12-foot-long tray that can hold up to ten 1-gallon steel cans. The contents of each can is limited to a maximum of 20 kg of HEU. Therefore, the holding capacity of a tube vault is $200 \text{ trays} \times 10 \text{ can/tray} \times 20 \text{ kg/can} = 40,000 \text{ kg}$. The trays are mounted on rollers, such that they can be rolled out of the tubes and secured in a tray holder supported by a fork lift. The nominal dimensions of the trays are 27 $\frac{1}{2}$ inch height by 6 $\frac{1}{2}$ inch width, but the widths are somewhat less in some cases, due to constriction that occurs when cross-members are welded into the trays to separate the individual can storage slots. The gross, tare, and net weights and the requisition number and batch number are recorded on cards attached to the cans. A job card is attached to the end of each tray.

Activity in each vault is monitored by a closed-circuit television monitor and a motion detection monitor. Two gamma-radiation monitors are mounted on the wall for personnel protection. Both HEU and VHEU are stored in the vaults. The door of the vault is steel and is secured by a key lock, a combination lock, and a plastic seal. The two-plus-two person rule is in effect for any personnel working in the vaults, and no single individual is given both a combination and a key. Safety features include the plant Emergency Notification System (ENS) speakers, duress alarms, criticality alarms, and physical/mechanical features to prevent entrapment.

All storage procedures for HEU are covered by CSAs (Criticality Safety Approvals). Any activities not explicitly covered by existing Plant Procedures are evaluated by a Unreviewed Safety Question Determination (USQD) in accordance with DOE Order 5480.21. The HEU in storage vaults is expected to survive a total building collapse because the vaults are made of steel-reinforced concrete and are structurally self-supporting.^{1c}

4.4.2 Modular Storage Vaults (MSVs)

Consideration of the MSVs is reserved for the Advanced Concepts discussion in Section 5.2.

4.4.3 Cage Storage

In addition to the vaults, EU may be stored in cages which are isolated rooms within MAAs. The cages are protected by combination locks and by an alarm system that is activated by any penetration of walls, floors, ceilings, or openings or by motion within the room.^{1g} Cages are typically 16 feet square, constructed from a wire mesh screen on steel supports. Unlike the tube vaults, the cages are equipped with fire sprinklers. Cages are used to store both high and low isotopic enrichment material; but, in general, the material has been categorized as less attractive than the pure, high-equity material stored in the vaults. The material is low equity because it is of a low enrichment level or because it is mixed with nonuranium materials, which prevent it from being converted to usable products.

4.5 TRANSIENT STORAGE (TS)

Although the Y-12 Plant has established a carefully defined set of procedures for receipt of uranium from outside sources and shipping of finished parts to outside points, the term *Transient Storage* has not been in prior use. Introduction of the term is justified because of the projected high volume of EU to be received at the site within a few years.

Most EU is shipped to and from the Y-12 Plant by Safe-Secure Transport (SST). Details are given in the Environmental Assessment.^{1k} Commercial carriers may be used if designated guidelines are met (see reference 1k and references cited therein). All HEU is packaged in containers meeting the Department of Transportation (DOT) regulations. At the point of receipt, the material is designated as being in TS. A transfer check is made to confirm the container item count and identity, and the integrity of the tamper-indicating devices is verified. Confirmatory measurements are made to verify that the specified quantity of EU is in fact present.^{1j} The material is then transferred to a Y-12-owned storage container and placed in IS. The shipping containers are returned to the owner.

4.6 INTERIM STORAGE (IS)

The IS concept was discussed briefly in Section 4.1. The current Y-12 Plant goal is to limit IS of weapons grade EU to ten years or less. This provides time for decision making and implementation of a long-term storage strategy. Much of the EU in IS is stored in cells known as bird cages. The bird cages are specially fabricated to maintain the EU within its container in a criticality-safe configuration.^{1g}

The Y-12 containers for IS are built to DOT standards (49 CFR 173 and 178). These include (but are not limited to) the following conditions:

- Thermal Exposure (10 CFR 71.73) of 30 minutes at 1475°F (800°C)
- Cold (10 CFR 71.71) of -40°F (-40°C) in still air and shade
- Immersion (10 CFR 71.73) in 3 feet of water for eight hours, and
- Water spray (10 CFR 71.71) of 2 inches per hour for at least one hour

4.7 CRITICALITY

Criticality avoidance will take precedence over other considerations in the design and operation of an EU storage facility. In addition to providing for the HEU to be stored in an array that is initially criticality safe, the facility must be engineered to avoid the occurrence of sustained fission under upset conditions such as fires, dust explosions, flooding, earthquakes, and tornadoes. In some respects, the existing Y-12 warehouse facility appears to be a reasonable prototype for a long-term storage facility. However, it must be borne in mind that the present warehouse was not initially designed for long-term storage but as an interim holding facility for EU.

Operations with fissionable uranium at Y-12 are carried out in compliance with DOE Order 5480.24 (*Nuclear Criticality Safety*). Two key elements of the order are the double contingency principle and preferential reliance on geometrical controls. The double contingency principle requires that "process designs shall incorporate sufficient factors of safety to require at least two unlikely, independent, and concurrent changes in process conditions before a criticality accident is possible." Preferential reliance on geometrical control requires that "as a first priority, reliance shall be placed on equipment design in which dimensions of the contained fissionable material and spacing between equipment are limited via passive engineering controls." The use of active engineering controls or administrative controls is acceptable only in cases where passive engineering controls are not achievable.

4.8 INVENTORY MONITORING PROTOCOL

Periodic physical inventories are necessary in order to assure that book values and records are up to date and that they accurately reflect current material holdings when materials are being handled routinely. The conduct of physical inventories to verify the presence, location, and quantity of nuclear materials is a fundamental principle of safeguards. An important part of physical inventory of discrete items is the verification of tamper indicating device (TID) integrity. It is necessary to show that TIDs are present in their assigned locations, are intact, and that the intact TID has served its function.

4.9 SCHEDULING

Current practice requires a bimonthly (every two months) inventory of special nuclear materials. This practice typically consists of locating all discrete items (either individually or by verification of the integrity of the storage system) and verification of TIDs on discrete items. Some portion of the inventory population is then selected to be remeasured to confirm/verify material contents. Alternate inventory frequency requirements also may be approved for inventories containing large numbers of items, items stored in high radiation fields, critical assemblies, and other situations where alternate control mechanisms provide assurance that unreported changes in inventories would be detected. Conditions never exist in which an inventory or a periodic verification of nuclear material may be waived. Therefore, the issue regarding physical inventories is always one of frequency and method.

The recent changes in the weapons complex have initiated activities related to providing guidance for extended inventory frequency, based on meeting various levels of control. With an eye on long-term storage, DOE-Headquarters has provided methods to achieve these extended inventory frequencies. The methodology provides six months to perhaps five years between required inventories. These extensions are based on providing enhanced storage facilities, access and containment monitoring, and item monitoring systems. Provisions are also detailed approving systems capable of qualifying as continuous physical inventory. In theory, no inventory or verification would ever be required if it could be demonstrated that necessary high-quality data could be continuously collected relative to each item of nuclear material. Demonstration of technology capable of achieving continuous inventory is still forthcoming.

5. ASSESSMENT OF ADVANCED STORAGE CONCEPTS

5.1 FUNCTION OF CONTAINERS

The principal functions of the EU container are to avoid dispersal of the uranium material during normal handling and to control corrosion during storage.

5.1.1 Container Design

The Oak Ridge Y-12 Plant has maintained an inventory of both highly enriched and depleted radioactive materials for over 40 years. The task of designing, fabricating, and testing containers for transportation and storage of these materials belongs to the Packaging Systems Management (PSM) organization at Y-12. This process is outlined in the following paragraphs.

Upon recognition of the need for a storage container a System Requirements Document is prepared. This document provides the requirements for the container; e.g., radiation level, material form, quantity of material, safeguard, handling, and anticipated storage configuration among other considerations. The Package Designer with support from a team of plant experts in the fields of nuclear criticality, radiation protection, and mechanical design prepares a detailed design package.

The detailed design package includes fabrication drawings, specifications, data sheets, supporting calculations, and analyses for the proposed container. Criticality avoidance is the first and most important design consideration in container design when dealing with HEU. The Criticality Safety Organization within Y-12 provides detailed guidance on quantity of material that can be placed in a container as well as a safe geometry for a given material and the handling techniques to be used. This guidance is supported with detailed analysis and calculations. Storage configuration must also be considered in the design from both a criticality and radiation protection standpoint; i.e., what is the necessary spacing between containers and how will the spacing be maintained. Radiation shielding of the materials is important for the protection of those working around the storage area. As Low As Reasonably Achievable (ALARA) is the standard that is required by DOE directive. The mechanical design of the container and array configuration must consider containment of the contents in the event of an anticipated accident as well as compatibility of the contents with the container materials. Upon completion the detailed design package is subjected to a review process which includes an independent review as well as a review by operations personnel.

Upon completion of a successful review, a quality assurance plan is developed for the container. The quality assurance plan details the quality evidence required by applicable quality assurance standards and procedures.

5.1.2 Atmosphere Within Storage Cans

The fundamental corrosion chemistry of uranium is discussed in Section 3.1.1. From that discussion, it is evident that environmental control of the internal volume of the container must play a significant role in the container design. Conceptually, the best procedure for minimizing corrosion would be to evacuate the container and backfill with a dry inert gas or to evacuate the container after closing. The method of sealing the container also becomes a key factor if this option is exercised. Because the container might be in storage for very long periods of time, the option expected to minimize net corrosion through minimizing in-leakage would be to seal the container with a weld. This would preclude the concern that over time any type of sealing gasket, whether metal or polymeric, would have to be replaced. The cost of repackaging several thousand containers would not be trivial and the additional cost of welding might eventually be justified. On the other hand, if a modest amount of surface corrosion is deemed acceptable (such as the corrosion rates calculated in Section 3.1.1 for cool, dry conditions), nonwelded assemblies become a tenable alternative. A formal cost analysis could be performed, but because of the uncertainties in storage times and ultimate uses, the results of such an analysis would necessarily be ambiguous. While acknowledging that under some scenarios a fully welded container would be more cost-effective, it is felt that crimp-sealing is the option more likely to serve the needs of the EU storage program.

5.1.3 Storage Cans For Prolonged Low-Maintenance Storage (PLMS)

The storage container for EU metal and U_3O_8 is a cylindrical steel can. The can material is 304L stainless steel meeting the standard ASTM-A-240.³⁹ The low-carbon steel is required to avoid vulnerability to corrosion in the welded seam of the can. The wall thickness is 30 gage, corresponding to a wall thickness of about 0.012 inch or 0.030 cm. The outer diameter and height of the can are $6\frac{1}{8}$ inch (15.56 cm) and $8\frac{3}{4}$ inch (22.22 cm), respectively. The internal volume of the cans is 255.1 in^3 ($4,180 \text{ cm}^3$). The volume of the hollow cylindrical castings was shown previously to be 956 cm^3 . The free volume of the cans in standard metal storage mode is thus $4,180 - 956 = 3,224 \text{ cm}^3$.

The mechanical strength of the can is enhanced by four concave symmetrically spaced beads in the can wall. The bead width and inner-contour radius are 0.08 inch and 0.04 inch, respectively. The top and bottom lids are attached to the wall by crimp seals. The integrity of the crimp seal is enhanced by use of a sealant applied to the perimeter of the lids prior to crimping. A commercially available, water-based sealant consisting of an emulsion of natural rubber, pigment, and modifiers is currently used.

The cans are to be obtained from a commercial vendor with the bottom lids already sealed and the sealant applied to the top lids. The supplier of the sealant recommends that the lids be sealed within six months of the time of application of the sealant. All the cans are leak-tested by the vendor with the bottoms crimped. The leak tests are performed at six psi with the system either submerged in water or covered with detergent. Appearance of bubbles is a cause for rejection. After the charge

is placed in the can at Y-12, the top lids are sealed with a commercial crimping apparatus. The consequences of can-leakage or improper handling are discussed quantitatively in Section A-5.1.3. of the Appendix.

5.2 MODULAR STORAGE VAULTS (MSVs)

Anticipated returns of uranium from the nuclear weapons stockpile and of reactor grade materials will require that the storage capacity be expanded. The Y-12 Plant has capacity available within existing buildings to store the entire DOE inventory of EU. Rather than build a large number of additional tube vaults, the concept of MSVs is being considered.^{1e,21} The MSVs that would be used are structurally equivalent to the existing tube vaults.^{1e} Some prototypes have been obtained and are currently being evaluated at Y-12. The storage arrays will be built up by vertically stacking concrete modules, each of which holds twenty cans in a five by four array. The cavities which accommodate the cans are lined with eight-inch schedule 40 pipe having a wall thickness of 0.32 inch. The individual modules have rectangular dimensions of 78 inches × 76 inches, are 20 inches thick, and weigh 9,697 pounds empty. Seismic safety analysis documentation for the modular storage vaults allows for the stacking of eight MSVs,³⁸ but in current practice they are stacked only five high. A concrete lid will be placed on top of each stack. Cable-lock seals will be installed on each of the vertical arrays. Construction costs for the MSV approach are only ten percent of typical tube vault facility costs.

5.3 ENVIRONMENTAL CONTROL

Degradation of uranium metal in long-term storage can be further prevented by maintaining the environment within the entire storage facility. In this way, corrosion of stored metal would be minimal even if the container does not remain air tight. Equally important, maintaining a noncondensing environment within the facility will increase the effective service life of the storage cans. The importance of the latter consideration is dependent upon how long the material is eventually maintained in PLMS. Under short-term scenarios, significant degradation of the stainless steel cans is not expected to be a significant consideration.

5.4 STORAGE OF PARTIALLY DISMANTLED WEAPONS

One option for dismantling of the nuclear stockpile is to only partially dismantle the retired weapons and to place the components in long-term storage. This option introduces several additional complexities that are considered to be beyond the scope of the current report. A separate assessment document covering the storage of partially dismantled weapons will be prepared in the future.

5.5 USE OF PLASTICS IN URANIUM STORAGE

Plastics (or polymers) are convenient vessels for use in storing chemicals of all sorts. In particular, polyethylene is a useful material for bottles because it is reasonably rugged, does not shatter on impact, and is inexpensive. Uranium-containing materials from other sites are often shipped to Y-12 in polyethylene containers. Polyvinylchloride is useful in the form of a plastic film that is easily heat-sealed. Natural rubber, which is primarily polymer of methylbutadiene, is a constituent of the sealant used for the crimp-sealing of storage cans for PLMS (Section 5.1). There are some problems that have to be addressed if these and other polymers are to be used in uranium storage, including flammability, long-term degradation, vulnerability to radiation damage, and neutron reflecting and moderating properties that affect nuclear criticality. Because of the complexity of the issues, the use of plastics in uranium storage is being addressed in a separate document currently in preparation.⁵²

5.6 EVALUATION OF NEEDS

Successful operation of the PLMS Facility requires that (1) the materials for storage are acceptable as-received in terms of their chemical and isotopic content; (2) the physical form is acceptable for storage; (3) the relevant data have been recorded and entered into the database; (4) databases should not be technology-dependent for long-term utility (i.e., frequent software upgrades are not desirable; and (5) the storage configuration meets the appropriate specifications. In addition, a systematic methodology must be developed to verify the authenticity of the database on both a routine inspection basis and on demand.

5.7 ON-SITE INSTRUMENTAL CAPABILITIES

Specific items needed for the storage facility include weight scales, bar code readers, energy discriminating radiation detectors, and glovebox facilities for opening and inspecting containers. An FTIR (Fourier Transform Infrared Spectrometer) inspection capability is recommended for the identification of any elastomers, lubricants, or other organic matter that might be associated with materials received for long-term storage. An economic analysis is needed to determine the level of NDA (nondestructive analysis) capability that will be required for the facility itself. Parts might be shuttled to existing instrumentation in production areas for acceptance certification and inventory verification.

6. HAZARD ANALYSIS

A rigorous hazard analysis is the key element in establishing the credibility of this assessment. The fundamental properties of uranium metal and triuranium octoxide that underlie the hazards are discussed in Sections 2 and 3. The intent of the hazard analysis is thoroughness and will include the data and logical basis on which some perceived hazards can be eliminated from consideration.

6.1 HAZARD IDENTIFICATION

The scope of this analysis is focused on the long-term storage of uranium metal as cast full-density cylindrical shells and triuranium octoxide in the form of loose powder. The hazards of other oxides and of finely divided uranium metal are discussed for two reasons: (1) to address the hazards involved when these materials are placed in TS, IS, and in-process storage; and (2) to show thoroughness and justify the logic that excluded these materials from being designated as acceptable forms for long-term storage.

6.2 FUNDAMENTAL HAZARDS ASSOCIATED WITH MATERIAL FORMS

6.2.1 Criticality

As noted in Section 4.7, criticality avoidance will take precedence over other considerations in the design and operation of an EU storage facility. The Environmental Assessment addressed possible scenarios that could lead to criticality excursions. The PLMS in tube vaults or MSVs was shown to be sufficiently rugged to withstand even building collapse incidents without a significant likelihood of allowing the stored material to reach a critical configuration.^{1f} The IS arrays are less robust and there is some possibility of experiencing a criticality excursion exists in a case of building collapse.

6.2.2 Chemical Reactivity

The chemical forms of uranium recommended for PLMS were given in Section 3.8 as cast, fully dense metal and loose U_3O_8 powder. The latter is thermodynamically stable with respect to reaction with air or water. Uranium metal is thermodynamically unstable with respect to reaction with either air or water, but reactions of both types are very slow under ambient conditions (see Section 3.1.1).

The wider variety of materials that might be placed in IS pose more complex problems. Metals having a high specific surface area, such as machine turnings and saw fines, show enhanced rates of corrosion and the formation of UH_3 during corrosion further enhances the vulnerability to ignition. The oxides UO_3 and U_3O_8 are nonreactive, but very finely divided UO_2 can be pyrophoric (see discussion, Section 6.4.2).

Uranyl nitrate crystals and solutions are corrosive toward many container materials. Long-term storage scenarios involving uranyl nitrate should be avoided by converting the material to metal or U_3O_8 for PLMS. The same generalizations apply to the chlorides and sulfates.

Uranium tetrafluoride is intrinsically inert toward air and water, but often contains chemisorbed HF which is highly corrosive. Uranium hexafluoride has an appreciable vapor pressure under ambient conditions. Thus, both fluorides show hazardous characteristics that mitigate against their being maintained in IS for extended periods of time.

The generalizations concerning IS are also pertinent to TS. Materials in IPS are covered by the Procedures of the appropriate operations organization and will not be addressed as part of this assessment.

6.2.3 Pressurization Due To Helium Generation

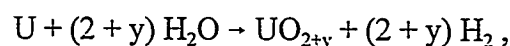
The rate of container pressurization due to alpha decay of the naturally occurring uranium isotopes is demonstrably very small. Because each alpha decay of the naturally occurring uranium isotopes generates one helium atom, the rate of helium formation is numerically equal to the disintegration rate, as given in Table A-14 of the Appendix (Section A-2.2.1). For a 18-kg oralloy casting containing 1 percent ^{234}U , the net fractional pressurization in a standard storage can is only $1.51(10^5)$ year⁻¹. The calculation is given in the Appendix, Section A-6.2.3.

6.2.4 Pressurization Due to Radiolytic Reactivity

The half-lives and decay sequences of the uranium isotopes are given in the Appendix, Section A-2.3. For the naturally occurring isotopes ^{238}U and ^{235}U , the maximum pressurization due to radiolysis of gas precursors is minimal for any reasonable time frame of storage. For ^{234}U and the artificial isotope ^{236}U , the extent of radiolytic pressurization is also small under most foreseeable circumstances. On the other hand, serious pressurization may be anticipated in a short time frame in the case of hydrated compounds of the uranium isotopes ^{233}U , and ^{232}U . Calculations are given in the Appendix, Section A-6.2.4.

6.2.5 Pressurization Due To Hydrogen Evolution

Container pressurization by hydrogen is an improbable occurrence in the long-term storage of HEU metal containing only the naturally occurring isotopes. As noted in Section 3.1.1, hydrogen is formed during the corrosion of uranium by water vapor according to the reaction:



where y lies between 0.0 and 0.1. However, measurable hydrogen is not evolved in the presence of oxygen except under saturated conditions as noted in Section 3.1.1.

Even in an oxygen-free environment or under condensing conditions, significant pressurization is to be anticipated only if the amount of water within the storage vessel is large with respect to the free volume. Moisture that contacts the metal due to leakage of moist air into the storage can will react to generate a small positive pressure of hydrogen, but the positive pressure within the can will then vent through the same leak. Therefore, a net pressurization is anticipated only if (1) some moisture is trapped inside the storage can at the time it is sealed, or (2) some sort of self-sealing occurs in the vessel wall after a significant quantity of water vapor has diffused in. Sample calculations are given in the Appendix, Section A-6.2.5, of the net possible hydrogen pressure buildup and the rate of hydrogen pressure buildup under these scenarios. With reasonable care, these are very improbable occurrences.

6.2.6 Airborne Release Of Uranium

Circumstances that might lead to the airborne release of finely divided uranium-containing materials from uranium in storage are fire, criticality excursion, or violent destruction of a vessel in which a finely divided uranium compound is stored. Fire and criticality considerations are discussed in Sections 6.4 through 6.4.2 and 6.2.1, respectively. The best means of preventing the third scenario (dispersal of powdered material during violent container failure) is by prohibiting the storage of any finely divided form of uranium compound that is capable of causing an explosion. Thus finely divided uranium metal, uranium hydride, and pyrophoric uranium dioxide are unacceptable forms for long-term storage. The storage facility itself should be designed in such a way as to protect the public from an airborne release of uranium. Two key elements in achieving this goal are providing adequate air filtration for the facility ventilation system and constructing the facility in such a way as to assure that its physical integrity will be maintained in the event of foreseeable mishaps.

The primary exposure mechanisms for HEU are through inhalation or ingestion. Assuring that containers used to store uranium are free from contamination will reduce the potential for exposure to both uranium and radiation-emitting decay products and contaminants. This assurance can be obtained by the following steps:

1. Conducting health physics monitoring on shipping containers for contamination when they arrive at Y-12 for storage,
2. Storing in containers in configurations that are demonstrably engineered to survive design basis accidents without release of contents,
3. Inspecting container integrity on a routine basis to detect any leakage at an early stage, and
4. Maintaining a routine health physics monitoring program for removable contamination in the facility.

6.3 IMPACT OF MAJOR EXTERNAL PERTURBATIONS

6.3.1 Vulnerability to Natural Phenomena

The susceptibility of the Y-12 Plant to severe natural catastrophes such as earthquakes, tornadoes, and floods has been evaluated in the Y-12 Environmental Assessment.^{1b,1c,1d} The Oak Ridge Reservation experiences moderate seismic activity, with one or two detectable earthquakes per year having magnitudes less than three on the Richter scale. The largest recorded earthquake in the area, which occurred November 30, 1973, was 4.6 on the Richter scale; the ground acceleration was less than 0.01 g.^{1b,35,36} By contrast, the criterion for the design basis earthquake used in the Y-12 Environmental Assessment was 0.18 g.^{1c}

Wind speeds on the Oak Ridge Reservation are less than 11.9 km/h (7.4 miles per hour [mph]) 75 percent of the time, and wind speeds exceeding 30 km/h are rare.^{1b} Tornadoes are very rare, but a tornado having wind speeds estimated to be 64 to 209 kilometer/h (40 to 130 mph) did strike the perimeter of the Y-12 Plant on February 21, 1993.^{1b, 37} No structural damage occurred to the facilities and the ensuing power outage did not result in the release of hazardous material.^{1b}

The consequences of the collapse of an uranium processing facility were evaluated in a Defense Programs Safety Survey Report.⁴³ The results encompass the consequences of a postulated collapse of any other building on the site. The HEU in storage vaults is expected to survive a building collapse because the vaults are made of steel-reinforced concrete and are structurally self-supporting. Birdcages could be crushed by falling debris and the possibility exists that a critical array could be formed.⁴³

6.3.2 Vulnerability to Aircraft Crash Scenarios

The probabilities of an aircraft crash impacting the proposed Y-12 EU storage areas has been calculated in the Environmental Assessment.^{1f,1g,42} Calculations were based on the methodology prescribed by the U.S. Nuclear Regulatory Commission.⁴¹ The building having the highest calculated frequency had a value of $2.1(10^{-7}) \text{ year}^{-1}$.

6.4 FIRES

This section considers the flammabilities of the stored uranium materials in isolation. Consequences of a fire occurring in the vicinity of stored uranium are deferred to Section 6.5.1.

6.4.1 Metals

The ignition temperature of uranium is dependent upon the state of subdivision of the material.²² Large billets of uranium are routinely preheated to temperatures in excess of 630°C and transferred in air to forging facilities. Metallic uranium in massive forms presents little fire hazard, but it will

burn if exposed to a severe, prolonged external fire. Large pieces of uranium (cherry size and larger) will stop burning when allowed to cool in air from a red heat.¹⁶ Thus, uranium metal in the form of full-density cast or forged bulk material is accurately described as slightly, or slowly, combustible.²³

By contrast, finely divided uranium metal powder and finely divided uranium hydride are pyrophoric.^{23,24} The product of combustion is primarily U_3O_8 .²² Uranium machine turnings are observed to spark frequently when handled dry, and chip fires occur frequently when uranium metal is being machined, despite the use of copious quantities of water-based machine coolants. These "hot chips" are routinely quenched by moving them below the liquid coolant level. Uranium turnings and fines stored out of doors in a closed container under water or water-soluble oil will corrode and eventually ignite during hot weather if the water is allowed to evaporate.²³ Clean uranium turnings or chips oxidize readily in air. If confined in a container without liquid or air movement, they can ignite spontaneously. The presence of moisture in the gas phase over exposed chips increases this possibility.^{23,25}

The standard precautions regarding avoiding contact of highly electropositive metals with chemical oxidizers or other reactive chemicals apply to uranium metal.

6.4.2 Oxides

Uranium dioxide (UO_2) is known to be pyrophoric in the finely divided state, such as the product obtained by the hydrogen reduction of uranyl oxalate, ammonium diuranate, or a higher oxide.^{12,13} Such a pyrophoric oxide, containing 2 to 9 percent UH_3 , is also obtained by the anaerobic corrosion of uranium metal by water.²⁵ On the other hand, sintered high-density UO_2 is highly resistant to oxidation.

6.5 MATERIAL DISPERSAL

The health and safety properties of uranium are well known and have been comprehensively addressed in Reference 27. They will not be repeated in detail in this document. However, certain parameters which may specifically relate to the storage of uranium deserve mention here.

The isotopic distribution, the chemical form, and the physical form determine the primary hazards from uranium. The isotopic distribution determines the intensity of the gamma radiation and the overall specific activity. The chemical form determines the solubility and therefore the fate of the uranium within the body. The physical form determines the exposure mechanism; powders are more likely to be inhaled than bulk solids but less likely than gases.

6.5.1 Material Dispersal In Fires

Burning of uranium occurs as solid state combustion; no flames are seen unless a contaminant is present in or on the metal. As a combustible metal, uranium is less readily ignited than magnesium but more readily than aluminum, which forms a tenacious oxide protective layer.¹⁶ Solid state combustion is typical of combustible metals and is seen as a glowing, ember-type phenomenon without flames. Burning metal typically reaches higher temperatures than burning flammable liquids because heat transfer away from the fire does not occur through the evolution of gaseous combustion products.

Uranium fires release products of combustion in the smoke plume of fires with gas-producing potential. The amount and rate of combustion products released depends on the amount and form of the metal and contaminants involved. Small amounts of chips and other finely divided forms of uranium may undergo rapid oxidation but not at a rate fast enough to be considered burning. Estimates of plume travel and quantity must consider the form and amount of uranium as well as the type event, rapid oxidation or true combustion. The heated residual nitrogen from the surface of a burning uranium mass can carry some fine particulates.

For uranium metal or octoxide stored in the configurations recommended for PLMS, only the most extreme sort of circumstances can be visualized wherein fires could occur that would result in significant material dispersal. In IS or TS, the possibility of a serious fire occurring within the facility is more plausible. In this case, the use of containers meeting appropriate DOT regulatory requirements is the means of preventing dispersal. Storage of canned subassemblies constitutes a point of particularly high vulnerability because of the acute flammability and very high enthalpy of oxidation of some weapons components. As noted previously (Section 1.1), this topic will be addressed in a separate document.

6.5.2 Explosion And Pressurization

Uranium, like all reactive metals, presents a potential explosion hazard because smaller pieces react slowly with moisture (i.e., in air) to liberate hydrogen. Larger pieces also react, but oxidation ceases due to oxide-crusting of the surface and the ability of larger pieces to dissipate heat. It is possible to accumulate enough hydrogen in a closed area or space to present a possibility of hydrogen ignition when exposed to an ignition source. The term *explosion* is perhaps inaccurate because the invisible flame front would not travel at the speed of sound (detonation) unless well confined. If hydrogen is not formed, the ignition of uranium metal is expected to cause a deflagration, not a detonation.

6.5.3 Spills

Major spills are improbable for PLMS material for the following reasons: (1) the concrete storage array will protect the storage cans, even under very stressful conditions; (2) the storage cans are to be removed from the concrete storage arrays only infrequently, such as for IAEA inspections, routine inspections, inventories, or investigations; (3) the stainless steel storage cans are robust and would

not be breached as a result of dropping or other mishandling. Furthermore, for metal storage, the castings would remain intact even if the system were subjected to a mechanical insult sufficient to destroy the storage can, and the only material subject to spillage would be the corrosion layer.

6.6 HEALTH PHYSICS

As noted in Section 2.1, the primary factor governing the amount of radiation emitted from uranium containing only the naturally occurring isotopes is the fraction of isotope ^{234}U . However, additional protection for operating personnel may be needed if the uranium is irradiated material containing fission products, if it contains a high level of nuclear decay products, or if it has been contaminated by contact with transuranics such as plutonium.

The significance of penetrating radiation (i.e., X- and gamma radiation) increases severely when the material contains significant amounts of the isotopes ^{232}U , ^{233}U , and ^{236}U as noted in Section 2.3. Massive shielding is required to attenuate the high-energy gamma radiation emitted during the decay of the ^{208}Tl at the end of the decay sequence. Procedures should limit the amount of time spent by personnel in areas where HEU is stored in very large quantities. Adequate ventilation is needed to prevent radon levels from becoming elevated.

Requirements for protection from the hazards of uranium in storage are highly dependent upon these parameters. Uranium with a high gamma intensity, particularly reprocessed uranium that may contain uncommonly high levels of ^{232}U , may require greater shielding than enrichments with lower gamma intensities. Residual fission products in reprocessed uranium may introduce additional radiological characteristics. Solubility is a factor in regulatory limits for airborne uranium concentrations. Physical form is a factor in the types of monitoring programs required for a storage facility.

7. CONCLUSIONS AND RECOMMENDATIONS FOR ENRICHED URANIUM STORAGE AT Y-12

The following conclusions and recommendations are for storage of EU metal and its compounds and alloys, and will only include handling to the extent that it is required to prepare the material for storage and to conduct routine inspections and investigations. The recommendations are not inclusive of all possible processing and handling scenarios. The recommendations are consistent with the proven methods, materials, and facilities that have been successfully used in the past. They are not dependent upon the outcome of ongoing or future research and development programs.

7.1 MATERIALS ACCEPTANCE CRITERIA

As noted in Section 1, the formal Y-12 Acceptance Criteria are currently being formulated by an ad hoc committee.⁴⁶ The criteria adopted will be the basis for acceptance of EU at the site. Until the revised acceptance criteria are approved, the basis for materials acceptance will continue to be the Egli document.⁴⁵ The standards for acceptance of uranium given by the Egli document were the following:

- the relative hazard potential of alpha emitters other than uranium is limited to ten percent
- the beta ratio, defined as the activity of the sample divided by the activity of an unirradiated uranium standard, must be less than 1.25
- Total fission product gamma activity must be less than 0.20 microcurie (7,400 Bq) per g of uranium

7.1.1 Designations For Types Of EU Storage

At the point of receipt at the Y-12 Plant, any EU-containing materials entering the plant will be defined as being in TS.

As expeditiously as possible, the materials in TS should be converted to IS status. This places the materials under the configuration defined and controlled by plant documentation. Under no circumstances should the materials remain in the TS category for more than one year.

All materials containing isotopically EU with a ²³⁵U enrichment of 20 percent or greater should be placed in long-term storage (PLMS) as soon as practicable. Exceptions are the following:

- material that can be classified as in-use or in-process
- material that is assigned to interim storage because of an anticipated programmatic use
- low-equity material that has no foreseeable future use and can be managed in a legal and environmentally acceptable manner without endangering the public

- irradiated uranium and other materials containing transuranic elements, fission products, and/or radioactive decay products in such concentrations as to require their being maintained under more stringent controls than are appropriate for HEU

7.1.2 Recommended Chemical Forms for Long-Term Storage

Based on the properties of the various uranium compounds listed in Section 3.0, the hazards described in Section 6.0, and other considerations outlined in Section 3.8., only uranium metal and triuranium octoxide (U_3O_8) should be designated as acceptable forms for long-term storage. Storage of UF_4 prepared by duct melting into a fused casting would probably be an acceptable practice because any chemisorbed volatiles such as HF or UF_6 would be expelled during the melting process. However, this process is not a proven technology and should not be deemed acceptable until a sound experimental program has been conducted to prove its adequacy. Details were given in Section 3.8.

Uranium alloys and intermetallics can be stored under the same standards that pertain to pure metal, provided that a sound technical basis exists for certifying that said material is comparable or superior to the metal in terms of corrosion resistance, flammability, and health/environmental impact.

7.1.3 Recommended Physical Forms for Long-Term Storage

The preferred physical form of uranium metal for long-term storage, as noted in Section 4.2, is a hollow right-circular cylinder having an inner radius of $1\frac{3}{4}$ inch (4.45 cm) and an outer radius of 2.5 inch (6.35 cm). The preferred size of a routine casting is 18 kg, and 20 kg is the maximum permissible mass. For an 18-kg casting, this corresponds to a cylinder height of 14.8 cm and a volume of 956 cm^3 . Other physical forms of uranium metal might be considered for long-term storage if their specific surface is sufficiently low that the material is not subject to rapid corrosion or ignition. Metal forms that have a high specific surfaces such as machine turnings, chips, and saw fines are definitely unacceptable for long-term storage and should be converted to metal or U_3O_8 . It is recommended that a maximum specific surface of $1.00 \text{ cm}^2/\text{g}$ be adopted as the standard for acceptance of uranium metal for long term storage. Calculations in the Appendix, Section A-7.1.3 show the basis for this recommendation. Note that the $1.00 \text{ cm}^2/\text{g}$ criterion is also being considered for use for plutonium storage.⁴⁰

As noted in Section 4.2, uranium metal parts manufactured as weapons components were routinely coated with corrosion-inhibiting oils immediately after being fabricated. This practice is not recommended for uranium cylinders going into long-term storage because:

- the oil films provide protection against corrosion for only a period of a few years, and
- the additional fire hazard associated with the oil film, while slight, is considered undesirable.

Triuranium Octoxide (U_3O_8) may be stored as a loose powder. This is a less-favorable form than the metal because of the possibility that some of the material could become airborne during handling.

Use of an elastomer bag, such as polyvinylchloride, is acceptable as a means of reducing potential inhalation risk to personnel who prepare the material for storage. Polyvinylchloride is preferable to polyethylene for this application because the latter is flammable, although this is a very minimal risk because the polymer will be sealed within the storage can in contact with the nonflammable U_3O_8 . Calcining to 800°C in air prior to canning is recommended to convert impurity oxides to the U_3O_8 and to drive off adsorbed moisture.

7.1.4 Acceptable Isotopic Ratios

The majority of the EU to be accepted for storage at Y-12 is expected to be weapons grade, HEU or or alloy, with the approximate isotopic ratio 93 percent ^{235}U , 6 percent ^{238}U , 1 percent ^{234}U . However, the mission of the storage areas will be to accommodate all EU that meets the acceptance criteria, meaning ^{235}U enrichments above 20 percent. Within the DOE complex, many intermediate assay uranium storage values are being diluted to 20 percent to avoid stringent storage standards in existing treaties.¹⁶ All limits associated with 93 percent enriched material will be applicable to lower enrichments, including criticality constraints on maximum allowable weights per container.

7.1.5 Quality Assurance

The EU prepared for prolonged storage will be packaged under a documented quality control program to assure that the nuclear material meets the acceptance criteria and that the storage methodology meets the standard's requirements. The storage facilities will be operated in accordance with Federal Regulation 10-CFR 830.120. Occurrence reporting will be in accordance with DOE 5000.3B (See Appendix Section A-1.3, Tables A-10 and A-11).

7.2 TRANSIENT STORAGE (TS)

Upon arrival at the Y-12 Plant, any EU-containing material should be designated as being in TS. The time the material remains in this status should be minimized. While in TS, the following determinations are made:

- The shipping containers meet DOT specifications and do not show visual signs of damage or distortion.
- The items within the shipping containers are those specified by the shipping agreement.
- The received material meets Y-12 Plant acceptance criteria.⁴⁶
- The quantity of EU specified by the shipping agreement is, in fact, present.

Most received materials will at this point be transferred to storage containers owned by Y-12 and placed in IS. In some instances, materials that are considered to be unsuitable for IS should be transferred to the Enriched Uranium Operations Organization for conversion to a form that is acceptable for either IS or PLMS.

7.3 IN-PROCESS STORAGE (IPS)

In-process storage is a function of the Y-12 Enriched Uranium Operations Organization or other operating divisions. As such, it is not considered part of this storage assessment.

7.4 INTERIM STORAGE (IS)

The current goal of the Y-12 Plant is to limit the duration of storage of a material in IS to ten years or less. As a generalization, priority should be given to conversion of materials to forms accepted for PLMS if the material is in a form that poses health, environmental, or safety risks due to its chemical or physical state. Materials in IS should be contained in storage containers that meet the specifications for DOT Type A or Type B shipping containers (refer to Sections 4.5 and 4.6). Alternative configurations based on the use of approved safety devices such as bird cages, lock boxes, and carbon steel drums are also acceptable if they have been declared acceptable by a Criticality Safety Approval. However, it is an acceptable practice to use shipping containers whose certifications have expired or to use containers manufactured according to DOT specifications for use strictly for IS which have not been certified as shipping containers. The physical configurations in which the storage containers are arrayed must be established by operating procedures derived from formal Criticality Safety Approvals.

7.5 PROLONGED LOW-MAINTENANCE STORAGE (PLMS)

As noted in Section 7.2, provisions will be required for converting the as-received, EU-containing materials into forms acceptable for long-term storage, namely the metal (or alloy) or U_3O_8 . This will normally be the function of the Enriched Uranium Operations Organization at Y-12. Details of the unit operations normally used for chemical recovery of EU, purification, conversion to metal, and casting are given in reference 1h. It is recommended that the PLMS facility be physically isolated from any processing operations.

7.5.1 Storage Cans

Materials should be stored in standardized containers in order to facilitate efficient inventory confirmation and monitoring. Providing for the protection of uranium metal and its common alloys consists primarily of selecting a robust material that will protect the metal from corrosion by moisture or air. The oxide U_3O_8 is very inert, requiring little protection. Thus the main function of the storage can is to enclose the oxide powder to prevent its dispersal. The container design described in Section 5.1.3 is well-suited for these objectives. Dimensions were selected to allow storage of optimum quantities with respect to critically safe configurations.

To summarize:

- The contents of each storage can must be marked on the exterior of the container and entered into the database, as described in Section 7.7.
- Storage containers for uranium metal shall be designed for easy content retrieval and to facilitate inspections and inventory requirements.
- Plastics, oils, other combustible materials, and substances that may initiate corrosion must not be in physical contact with stored metal. However, use of polyvinyl chloride bags within the cans is allowable in the case of the oxide.
- Storage cans for PLMS of uranium should be made of 304-L stainless steel.
- The container's gaseous environment may consist of an inert gas or dry air. If an inert gas is selected, oxygen should be added to the extent of 1 ± 0.5 percent to inhibit corrosion by water vapor. The moisture content may be waived if an inspection certifies that the ambient conditions at the time the storage can is sealed are sufficient to preclude condensation.
- The storage can should be designed to maintain mechanical integrity, including its seal, during normal handling. However, the storage can is not expected to provide protection against all perils such as major fire and earthquakes. Design of the facility and of the concrete storage array are expected to cover these considerations.

7.5.2 Compartments And Storage Arrays

The two types of storage arrays for HEU, tube vaults and MSVs are described in Sections 4.4.1 and 5.2, respectively. Both types of storage are considered to be adequate for PLMS, but economic considerations strongly favor the use of MSVs. The geometrical array of the concrete storage array must provide a rigid lattice that will preclude the possibility of criticality excursions under foreseeable circumstances.

7.5.3 Long-Term Storage of Canned Subassemblies

As noted in Section 1.1, the long-term storage of partially dismantled weapons is considered to be a special case and is not included in the Enriched Uranium Storage Assessment.

7.6 STORAGE BUILDING FACILITY

7.6.1 Architectural Considerations

New storage facilities or significant modifications to existing storage facilities are designed to meet the requirements of DOE Order 6430.1A. Current storage facilities should be evaluated for compliance with DOE Order 6430.1A. A backfit analysis should be performed to determine what upgrades may be necessary to the facilities for compliance with the order. The design of the storage facilities provides for the integrity of the system with respect to criticality excursions to survive under any foreseeable circumstances. The facility must be designed to minimize the possibility of fire. The facility must be designed to prevent the escape of airborne uranium-containing particulate matter to the surrounding areas under any foreseeable circumstances. The facilities should have pitched (not flat) roofs. The facility heating and ventilation systems should be adequate to prevent conditions leading to condensation of moisture, as described in Section 3.1.1.

7.6.2 Robotics

Although the use of robotics has been impressively demonstrated for the storage of plutonium at the Rocky Flats Plant, it is doubtful that a need exists for this sort of facility in the case of long-term storage of HEU. The extremely toxic nature of plutonium and the high radioactivity makes the use of robotics attractive for this application. The toxicity and radiation hazards associated with EU are lower by orders of magnitude than those for plutonium. Therefore, the use of robotics is considered to be unwarranted for the uranium facility based on safety considerations alone.

7.6.3 Fire Safety

Because criticality safety is considered to be the dominant concern in the design and operation of the long-term uranium storage facility, the EU storage vaults should not be equipped with fire sprinklers or other sources of water. Undeniably, it would be proper and safe to use sprinklers in a fissile facility that was designed with that in mind. However, it is generally best to eliminate all combustible materials from the facility and thus avoid the need. Consequently, the design of the facility, the form of material stored, the mode of storage, and the operating procedures must be adequate to preclude the occurrence of a fire. This can be achieved by the following precautionary steps: (1) the facility itself is constructed to meet the National Fire Protection Association codes, using nonflammable materials, meaning principally concrete; (2) uranium metal is cast into a physical shape that gives a low surface area-to-mass ratio; (3) corrosion is minimized by storing the cast metal in leak-tight cans filled with dry air or argon containing a small amount of oxygen to preclude hydride formation; (4) uranium is not accepted for storage in chemical forms that are flammable or that tend to corrode the containment materials; and (5) uranium metal is removed from its containment only after being transferred to an inert gas-filled glovebox facility.

7.6.4 Inventory Verification

Every effort should be made to store EU in a manner that takes full advantage of extended inventory guidelines.

- Formidable barriers in storage systems
- Enhanced vault protection systems (interior motion detection, etc.)
- Continuous item observation and monitoring (TIDs, video image, closed circuit television, etc.)
- Minimal access to storage systems

As technology matures to an acceptable level for long-term applications, storage systems should be configured to allow continuous physical inventory. This could, for example, mean a continuous monitoring of mass and a nuclear attribute on individual items. However, the cost of implementing these state-of-the-art technologies is high and should be analyzed for cost-effectiveness.

7.6.5 Criticality Posting

Due consideration should be given to criticality safety limit posting. Emphasis should be placed on permanence. For example, signs etched in noncorrodible metal are more appropriate than painted metal signs. Computer-controlled laser etching systems would be capable of creating detailed text on such signs, as well as marking the metal canisters or metal labels can be attached to the canisters, with detailed information on each lot of material.

7.7 Database Management

As a result of this assessment, a database should be maintained in a permanent repository to serve as a master list of relevant information for the stored uranium materials. As a minimum, the database should contain:

1. identification of the material by a serial number;
2. material type;
3. chemical analysis for key elements;
4. a general elemental analysis by an approved method such as spark source mass spectrography or inductively coupled plasma emission spectroscopy (ICP);
5. isotopic analysis;
6. gross, tare, and net weights;
7. radiation analysis;
8. material history, including processing information for Y-12 processed material;
9. log of inspection tests performed and dates of inspection;
10. inspection results;
11. container information;
12. exact, current location in the storage facility; and
13. tamper-indicating device serial numbers.

Each container should be marked to include the job number (serial number), gross weight, net weight, tare weight, chemical analysis, and isotopic analysis. Laser-etched bar codes are an attractive option for ensuring that the information on the containers will be retrievable over a long period of time. Analyses may represent stream average values rather than specific part values provided that the basis for the analytical value is entered in the data base.

Provisions for electronic data storage must not be technology-dependent. It must not be dependent on tapes, disks, computers, etc., that will become obsolete after fifty years or longer. Standard software should be selected using a commercial database, hopefully customized to the application. The operating system should be selected in terms of (1) security considerations, (2) redundancy and adequate provisions for backups, and (3) ease of use. Standard commercial hardware is appropriate. Optical media may be preferable to magnetic media to avoid possible magnetic interference.

A hardcopy backup is recommended to permit data retrieval under conditions of extraordinary duress. Punched-card data organization systems might be used for this purpose. Of course, the card material used in such a system would have to be of a type that is immune to the passage of long periods of time.

7.8 MATERIALS THAT DO NOT MEET ACCEPTANCE CRITERIA

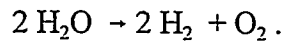
Materials which fail to meet the Acceptance Criteria⁴⁶ should be placed in storage at Y-12 only at the explicit instruction of DOE. Materials failing to meet the acceptance criteria that are transferred to Y-12 by DOE edict will not be stored under EU standards and criteria unless written approval is

obtained from the Y-12 Health Physics and Criticality Safety Organizations, certifying that the EU standards and criteria provide acceptable levels of protection from ionizing radiation and criticality excursions.

7.8.1 Acceptable Chemical/Physical Forms for Long-Term Storage.

As in the case of the naturally occurring isotopes, the most satisfactory chemical form for long-term storage is the pure metal or an alloy that has comparable or superior properties to the metal in terms of fire and corrosion resistance. The oxide U_3O_8 is an acceptable alternative.

Moisture poses a much more serious problem in the storage of artificial uranium isotopes than with the naturally occurring isotopes. In particular, the high radiation fields (alpha, beta, and gamma) that accompany ^{232}U decay cause rapid dissociation of water.



The resulting mixture of hydrogen and oxygen creates an explosion hazard as well as a possible source of container pressurization. As noted in Section 3.3, UO_3 is much more hygroscopic than U_3O_8 . However, the calcining of UO_3 to U_3O_8 involves exposure of personnel and equipment to high radiation levels if the material being processed contains a high concentration of decay products, and this consideration may override the advantages associated with conversion to U_3O_8 . If uranium is placed in long-term storage as UO_3 , the buildup of hydrogen in the cans should be prevented by some technique such as continuous purging with an inert gas, gettering, or incorporating a pressure-relief valve in the can.

7.8.2 Radiation Shielding

Storage facilities for ^{236}U , ^{233}U , and ^{232}U must provide radiation shielding for personnel protection. Calculated gamma fluxes must be based on long-term projections including the concentration of the species undergoing the most hazardous nuclear transformation, specifically that of ^{208}Tl . While the use of robotics does not appear to be justified for EU containing only the naturally occurring isotopes, storage facilities for the isotopes ^{236}U , ^{233}U , and ^{232}U might incorporate robotics to good advantage due to the highly radioactive nature of the decay products of these isotopes.

8. REFERENCES

1. DOE/EA-0929, *Environmental Assessment for the Proposed Interim Storage of Enriched Uranium above the Maximum Historical Level at the Y-12 Plant, Oak Ridge, Tennessee*, September, 1994. (a) Pp. D-4, D-5; (b) pp. 5-3 and 5-4; (c) pp. 6-22 and 6-23; (d) Appendix G; (e) pp. 3-11 to 3-13; p. 6-23; (f) pp. 3-4 to 3-7; (g) p. 3-8; (h) pp. 3-16 to 3-24; (j) p. 3-25; (k) pp. 3-31 to 3-36.
2. J. J. Katz, G.T. Seaborg, and L.R. Morss, *The Chemistry of the Actinide Elements*, Second Edition, Chapman and Hall, London and New York, 1986; and references cited therein. (a) p. 177, (b) p. 172, (c) p. 228, (d) pp. 233-244, (e) p. 256, (f) p. 257, (g) p. 266, (h) p. 304, (j) pp. 299-308, (k) pp. 298-335, (m) p. 171, (n) p. 306.
3. R. C. Weast and M. J. Astle, *CRC Handbook of Chemistry and Physics*, 62nd Ed., CRC Press, Inc., Boca Raton, Florida. (a) pp. B-329 and B-330; (b) p. F-172; (c) pp. B256-B339.
4. S. F. Mughabghab and D.I. Garber, *Neutron Cross Sections: Volume I, Resonance Parameters*, Published by the National Neutron Cross Section Center, Brookhaven National Laboratories, 1973.
5. A. M. Krichinsky, Chemical Technology Division, Oak Ridge National Laboratory, Personal Communication.
6. F. H. Oetting, M. H. Rand, and R. J. Ackerman, *The Chemical Thermodynamics of Actinide Elements and Compounds, part 1, The Actinide Elements*, IAEA, Vienna, STI/PUB/421/1.
7. A. Pattoret, J. Drowart, and S. Smoes, *Trans. Farad. Soc.*, 1964, 65, 98.
8. R. J. Ackerman and E.G. Rauh, *J. Phys. Chem.*, 1967, 73, 769.
9. I. Grenthe et al., *Chemical Thermodynamics of Uranium*, Elsevier Science Publishing Co., Inc., New York, 1992.
10. M. A. Filyand and E. I. Semenova, *Handbook of the Rare Elements III*, Boston Technical Publishers, Inc., Cambridge, Mass., 1970.
11. J. L. Weeks, *Trans. AIME, J. Metals* 1955, 203, 192.
12. Lisa Loden, Y-12 Enriched Uranium Operations, Personal Communication.
13. F. S. Patton, J. M. Googin, and W. L. Griffith, *Enriched Uranium Processing*, Int. Ser. Monogr. Nucl. En., Div. IX, Vol. 2, Pergamon Press, Oxford (1963).
14. R. D. Baker, B. R. Hayward, G. Hull, B. Raich, and A. R. Weiss, *Preparation of Uranium Metal by the Bomb Method*, LA-472 (1946).

15. J. Kreykes, Y-12 Enriched Uranium Operations, Personal Communication.
16. J. M. Googin, Martin Marietta Energy Systems, Inc., Senior Corporate Fellow, Personal Communication. (Deceased)
17. N. I. Sax and R. J. Lewis, Sr., *Dangerous Properties of Industrial Materials*, Van Nostrand Reinhold, New York, 1989.
18. J. B. Condon and E.A. Larson, *J. Chem. Phys.*, 1973, 59, 855.
19. J. S. Bullock, Martin Marietta Energy Systems, Inc., Y-12 Development Division, Personal Communication.
20. A. G. Ritchie, "A Review of the Rates of Reaction of Uranium with Oxygen and Water Vapor at Temperatures up to 300°C", *Journal of Nuclear Materials* 102, 170 (1981).
21. S. D. Moses, D. A. Tollefson, J. P. Belk, and E. Stumpfl, *Modular Storage System for Plutonium Storage*, Y/LF-472, Martin Marietta Energy Systems, Inc., Oak Ridge Y-12 Plant, September 17, 1993.
22. *The Merck Index*, Tenth Edition, Published by Merck & Co., Inc., Rahway, N.J., 1983.
23. Martin Marietta Energy Systems, Inc., Material Safety Data Sheet, and references cited therein.
24. L. Bretherick, *Hazards in the Chemical Laboratory*, Fourth Ed., Published by The Royal Society of Chemistry, Burlington House, London, 1986.
25. J. J. Burke, D. A. Colling, A. E. Gorum, and J. Greenspan, *Physical Metallurgy of Uranium Alloys*, Brook Hill Publishing Co., Chestnut Hill, Mass., 1976.
26. J. Belle, *Uranium Dioxide: Properties and Nuclear Applications*, Published by Naval Reactors, Division of Reactor Development, United States Atomic Energy Commission, 1961.
27. B. L. Rich, et al., *Health Physics Manual of Good Practices for Uranium Facilities*, EGG-2530, June 1988. (a) p. 2-5, (b) p. 2-7.
28. J. Dillon, Y-12 Development Division, Private Communication.
29. O. A. Vita, C. R. Walker, and E. Litteral, *Anal. Chim. Acta*, 64, 249-57 (1973)
30. G. Brauer, Ed., *Handbook of Preparative Inorganic Chemistry*, Academic Press, New York and London, 1965.

31. T. J. Hertzler and D. D. Nishimoto, Science Applications International Corporation, Idaho Falls, Idaho, "Depleted Uranium Management Alternatives," Predecisional Draft, December, 1993.
32. (Draft), "Cost Study for the D&D of the GDPs-Depleted Uranium Management and Conversion," K/D-5940-DF, September 1991.
33. T. R. Lemons et al., *The Ultimate Disposition of Depleted Uranium*, K/ETO-44, December 1990.
34. See also: (a) C. A. Colmenares, *Prog. Solid State Chem.*, 9 (1975) 139; (b) R. L. Jackson, J. B. Condon, and L. M. Steckel, Y-2078, Union Carbide, Oak Ridge Y-12 Plant (1977); (c) R. E. Sladky, Y-1829, U.S. Atomic Energy Commission, Oak Ridge Y-12 Plant (1972); (e) J. B. Condon, *J. Less Common Metals* 73 (1980) 105; f) S. Orman, *Oxidation of Uranium and Uranium Alloys in Physical Metallurgy of Uranium Alloys*, J. J. Burke, Ed., Brook Hill Publishing Company, 1976; (g) M. McD. Baker, L. N. Less, and S. Orman, *Trans. Farad. Soc.* 62 (1966) 2513; (h) M. McD. Baker, J. W. Bleloch, L. N. Less, and S. Orman, AWRE Report 040/64 (1964); (j) M. McD. Baker, L. N. Less, and S. Orman, *Trans Farad Soc.*, 62 (1966) 2525; (k) V. J. Corcoran, C. Johnston, W. J. Metcalfe, and J. Thorpe, AWRE Report 042/65 (1965); (l) T. Kondo, E. D. Verink, F. H. Beck, and M. G. Fontana, *Corrosion* 20 (1964) 314; (m) T. Kondo, F. H. Beck, and M. G. Fontana, *Corrosion (NACE)* 30 (1974) 330; (n) J. V. Cathcart, "Gaseous Oxidation of Uranium Alloys," in *Physical Metallurgy of Uranium Alloys*, Ed. J. J. Burke, (Brook Hill Publishing Company, 1976); (p) J. E. Antill, *Corrosion*, Vol. 5, 2nd Ed., Ed. J. J. Shrier (Butterworths, London, 1976), p. 73; and (q) W. D. Wilinson, *Uranium Corrosion and Alloys*, Volume II, *Uranium Metallurgy* (Wiley-Interscience, 1962).
35. *Seismic Hazard Evaluation for Department of Energy, Oak Ridge Reservation*, Oak Ridge, Tennessee, Y/EN 4683, 1992.
36. C. A. Powell et al., *Science*, 254 (1994) 686.
37. M. W. Knazovich et al., *Tornado Special Study Report*, Martin Marietta Energy Systems, Inc., ES/ESH-35, (1993).
38. *Unreviewed Safety Question Determination*, U/Eng/USQD-034, Martin Marietta Energy Systems, Inc., July 1993.
39. *Standard Specification for Heat-Resisting Chromium and Chromium-Nickel Stainless Steel Plate, Sheet, and Strip for Pressure Vessels*. ASTM A-240-93b, June, 1994. See Table A-13.
40. *U.S. Department of Energy Criteria for Safe Storage of Plutonium Metals and Oxides*, DOE-STD-3013-94, October 1994.

41. NRD (Nuclear Regulation Commission) Standard Review Plan 3.5.1.6., *Aircraft Hazards*, Revision 2 (1981).
42. R. S. Siegler and L. J. Luttrell, *The Annual Probability of an Aircraft Crash on the U.S. Department of Energy Reservation in Oak Ridge, Tennessee*, ORNL/ENG/TM-36, (1992).
43. *Defense Program Safety Survey Report*, Volume III, DOE/DP/70056-H1(UCNI), D. Pinkston, Ed. (1993).
44. *Kirk-Othmer Encyclopedia of Chemical Technology*, Third Edition, Vol. 23, pp. 510-47, John Wiley and Sons.
45. D. Egli et al., *The Report of the Joint Task Force on Uranium Recycle Materials Processing*, DOE/OR-859, September 1985.
46. "Criteria for Acceptance of Enriched Uranium at the Y-12 Plant," to be published.
47. RadDecay, Version 3, available from Grove Engineering, Inc., 15215 Shady Grove Rd., Rockville, Maryland, 20850.
48. J. E. Wilson, *Radiation Chemistry of Monomers, Polymers, and Plastics* Marcel Dekker, 1974. (a) pp. 14-16, (b) pp. 26-30, (c) pp. 80-82, (d) pp 82-84, (e) pp. 19 26, (f) pp. 30-37, (g) pp. 369-84, (h) pp. 84-85, (j) pp. 220-24.
49. R. J. Berry and N. W. Holm, Introduction to Basic Concepts and Principles in Radiation Dosimetry, P. 4 in *Manual on Radiation Dosimetry* (same authors, Marcel Dekker, Incorporated, 1970).
50. G. Friedlander and J. W. Kennedy, *Introduction to Radiochemistry*, Wiley and Sons, 1949.
51. J. K. Bair and J. Gomez del Campo, *Nucl Sci & Engr.*, 71, 18 (1979).
52. "Position Paper. Oak Ridge Y-12 Plant. Storage of Uranium in Plastics." To be published.

Appendix

The numbering system in this appendix corresponds to the sections of the text to which they pertain.

A-1.2 PROGRAM PARTICIPATION

The names of non-Y-12 individuals who participated in the preparation of this report and of the standards document are shown in Table A-8. Table A-9 lists the Y-12 employees who were involved.

Table A-8.
Enriched Uranium Storage Program
Participants from Outside of Y-12

Name	Company	Organization	Phone
Phillip Carpenter	DOE Y-12	Site Office	576-8238
Charles Coones ^a	DOE Y-12	Site Office	576-0482
Gary Galloway	SAIC	Oak Ridge Office	574-3754
Robert Graham	SAIC	Oak Ridge Office	481-2388
Scott Gibbs	LANL	Uranium Lead Lab. Program Office	(505) 665-8999
Leslie Jardine	LLNL	C-21	(510) 423-5032
Jeff Kass	LLNL	Lead Lab. Program Manager	(510) 422-4831
Alan Krichinski	ORNL	MMESI	574-6940
Lynn McCurry ^a	WASTREN, INC	Y-12 Site Office	241-3986
Damon Snow ^b	WINCO, INC	DOE Idaho Operations	(208) 526-0111
David Wall	DOE Y-12	Site Office	576-1989

^aConsultant to the DOE Y-12 Site Office.

^bConsultant to DOE-Idaho.

Table A-9.
Enriched Uranium Storage Program
Y-12 Internal Working Group

Name	Organization^a	Phone
Jim Anderson	Ind. Saf.	6-5608
Gary Bean	Ind. Saf.	6-8195
Jon Bullock	Dev. Op.	4-1735
Tom Christman	Ind. Saf.	4-1635
S. W. Combs	HSEA	4-2585
Shirley Cox	DP	1-4365
Jack Dillon	Dev. Op. ^b	4-1808
Walt Duerksen	Dev. Op.	4-1703
Gary Ellis	HSEA	4-3530
Tom Gafford	DP	1-4366
David Gass	HSEA	4-8070
John Googin	Dev. Op. ^c	6-5605
Bill Heineken	Ind. Saf.	6-3803
J. B. Hunt	HSEA	4-3547
Jon Kreykes	EUO	4-2099
Lisa Loden	EUO	4-2122
C. O. Miller	SSO	4-5861
T. R. Miller	DP	1-4367
Steve McClanahan	DP	6-3686
A. F. Moore	EUO	4-2087
Stan Moses	Engr., Y-12	4-2543
B. Niemann	Engr., Y-12	4-4059
Bill Orick	D&SO	4-3716
Glenn Pfenningwerth	EUO	4-1715
Steve Robertson	Engr., Y-12	4-9712
J. D. Stout	DP	4-2119
Ed Stumpf	DP	4-2547
R. G. Vornehm	HSEA ^b	6-2289
Gene Walker	D&SO	4-3720
Bryce Wilkinson	DP	6-3845
Steve Wilson	Ind. Saf.	4-0485

^a Ind. Saf. = Industrial Safety; Dev. Op. = Development Operations; HSEA = Health, Safety, and Environmental Affairs; DP = Defense Programs; EUO = Enriched Uranium Operations; SSO = Site Shift Operations and Emergency Preparedness; ENGR., Y12 = Engineering at Y-12; D&SO = Disassembly and Special Operations; PM = Program Management; Ind. Saf. = Industrial Safety.

^b Retired

^c Deceased

SECTION A-1.3 APPLICABLE FEDERAL REGULATIONS, DOE ORDERS, DOE DOCUMENTS, AND NONGOVERNMENTAL PUBLICATIONS

Table A-10 shows the Federal Regulations that apply to the design and operation of uranium storage facilities. Table A-11 lists the DOE Orders that pertain. Table A-12 lists some miscellaneous DOE publications pertaining to long-term EU storage. Table A-13 lists the nongovernmental publications that are relevant to this publication.

Table A-10.
Federal Regulations that Pertain to Long-Term
Enriched Uranium Storage Facilities

10 CFR 71, *Packaging and Transportation of Radioactive Material*

10 CFR 830.120, *Quality Assurance*

10 CFR 835, *Occupational Radiation Protection*

29 CFR 1910, *Occupational Safety and Health Standards*

40 CFR 61, Subpart H, *Emission Standards*

40 CFR 191, Subpart A, *Environmental Radiation*

49 CFR 173, *Shippers - General Requirements for Shipments and Packagings*

49 CFR 178, *Shipping Container Specifications*

TABLE A-11.
DOE Orders that Pertain to
Enriched Uranium Storage Facilities

- DOE 5000.3B, *Occurrence Reporting and Processing of Operations Information*, July 1993.
- DOE 5400.1, *General Environmental Protection Program*, June 1990.
- DOE 5400.5, *Radiation Protection of the Public and Environment*, January 1993.
- DOE 5480.3, *Safety Requirements for Packaging and Transportation of Hazardous Materials*, July 1985.
- DOE 5480.7A, *Fire Protection*, February 1993.
- DOE 5480.19, *Conduct of Operations Requirements for DOE Facilities*, May 1992.
- DOE 5480.20, *Personnel Selection, Qualification, Training, and Staffing at DOE Facilities*, June 1991.
- DOE 5480.21, *Unreviewed Safety Questions*, December 1991.
- DOE 5480.23, *Nuclear Safety Analysis Reports*, March 1994.
- DOE 5480.24, *Nuclear Criticality Safety*, August 1992.
- DOE 5500.1B, *Emergency Management System*, February 1992.
- DOE 5500.2B, *Emergency Categories, Classes and Notification and Reporting Requirements*, February 1992.
- DOE 5500.3A, *Planning and Preparedness for Operational Emergencies*, February 1992.
- DOE 5500.4A, *Public Affairs Policy and Planning Requirements for Emergencies*, June 1992.
- DOE 5500.10, *Emergency Readiness Assurance Program*, February 1992.
- DOE 5630.11B, *Safeguards and Security Program*, August 1994.
- DOE 5633.3B, *Control and Accountability of Nuclear Materials*, September 1994.
- DOE 6430.1A, *General Design Criteria* (especially section 1306, Unirradiated Uranium Storage Facilities), October 1989.

Table A-12.
Miscellaneous DOE Publications that Pertain to
Enriched Uranium Storage Facilities

DOE/EH-0929, *Environmental Assessment for the Proposed Interim Storage of Enriched Uranium Above the Maximum Historical Storage Level at the Y-12 Plant, Oak Ridge, Tennessee*, September 1994.

DOE/EH-0256T Revision 1, *Radiological Control Manual*, April 1994.

DOE-NE-STD-1001-91, *Guide to Good Practices for Training and Qualification of Instructors*, November 1991.

DOE-NE-STD-1002-91, *Guide to Good Practices for Training and Qualification of Chemical Operators*, November 1991.

DOE-STD-1012-92, *Guide to Good Practices for On-the-Job Training*, July 1992.

DOE-STD-1029-92, *Writer's Guide for Technical Procedures*, December 1992.

TABLE A-13.
Nongovernmental Publications that Pertain to
Enriched Uranium Storage Facilities

3.3 Nongovernmental Publications

ACI-211.1, *Standard Practice for Selecting Proportions for Normal, Heavyweight, and Mass Concrete*, American Concrete Institute, 1991.

ACI 349, *Code Requirements for Nuclear Safety Related Concrete Structures*, American Concrete Institute, 1990.

ANS 8.7, *Guide to Nuclear Criticality Safety in the Storage of Fissile Materials*, 1975 (Reaffirmed 1987).

ASTM-A-240-94, *Standard Specification for Heat-Resisting Chromium and Chromium-Nickel Stainless Steel Plate, Sheet, and Strip for Pressure Vessels*, 1994.

A-2.2.1

The predominant mode of initial decay for all six uranium isotopes is alpha, gamma emission. The half-lives and decay energies are given in Table A-14. The basis for calculating the rate constants, disintegration rates, and power outputs shown in the table are given in the following.

For any first order decay process applied to a quantity N of material:

$$dN/dt = -kN, \text{ or } dN/N = -kdt \quad (\text{A-2.1})$$

$$\text{Integrating, } \ln N = -kt + C \quad (\text{A-2.2})$$

When $t = 0$, $N = N_0$ by definition of N_0 , so $C = \ln N_0$

$$\text{Then } \ln N = \ln N_0 - kt, \text{ or } \ln N/N_0 = -kt \quad (\text{A-2.3})$$

When $t = \tau_{1/2}$ (the half life), $N = N_0/2$, so

$$\ln [N_0/(N_0/2)] = \ln 2 = k\tau_{1/2} \quad (\text{A-2.4})$$

$$\text{or } k = \ln 2 / \tau_{1/2} \quad (\text{A-2.5})$$

This equation is the basis for calculating the first order rate constants in Table A-14.

$$\text{and } dN/dt = -(N \ln 2) / \tau_{1/2} \quad (\text{A-2.6})$$

where $\tau_{1/2}$ must be in same units as t .

The number of counts per second, or Becquerels (Bq) is $-dN/dt$ if N is in atoms and t is in seconds. Then for one mole of pure isotope:

$$\text{Counts per second} = \# \text{ Bq} = (\ln 2 / \tau_{1/2}) \cdot 6.023 (10^{23}) \quad (\text{A-2.7})$$

or for M g of pure isotope of isotopic weight (IW) and the half-life expressed in seconds:

$$\# \text{ Bq} = (\ln 2 / \tau_{1/2}) \cdot 6.023 (10^{23}) \cdot (M/IW) \quad (\text{A-2.8})$$

For half lives expressed in years,

$$\# \text{ Bq} = (\ln 2 / \tau_{1/2}) \cdot 6.023 (10^{23}) \cdot (M(g)/IW) / (365.25 \cdot 24 \cdot 60 \cdot 60) \quad (\text{A-2.9})$$

Numerically,

$$\# \text{ Bq} = 1.322(10^{16}) \cdot M(g) / (\tau_{1/2} \cdot IW), \quad \tau_{1/2} \text{ in years.} \quad (\text{A-2.10})$$

This equation is the basis for the calculation of the number of counts per g per second in Table A-14. Count rates are often expressed in units of the Curie (Ci):

$$1 \text{ Ci} = 3.7(10^{10}) \text{ disint/sec} = 3.7(10^{10}) \text{ Bq} \quad (\text{A-2.11})$$

$$\text{Count rate} = 3.5755(10^5) \cdot M(\text{g}) / (\tau_{1/2} \cdot \text{IW}) \text{ Ci} \quad (\text{A-2.12})$$

The power generated per g is found by multiplying the number of disintegrations per g per second by the decay energy.

Table A-14.
Properties of Uranium Isotopes

<u>Isotope</u>	<u>Half-Life (Years)^a</u>	<u>Decay Energy (MeV)^b</u>	<u>First Order Rate Constant, k, s^{-1c}</u>
²³² U	73.6	5.414	$2.984(10^{-10})$
²³³ U	$1.62(10^5)$	4.909	$1.356(10^{-13})$
²³⁴ U	$2.45(10^5)$	4.856	$8.964(10^{-14})$
²³⁵ U	$7.037(10^8)$	4.681	$3.121(10^{-17})$
²³⁶ U	$2.39(10^7)$	4.573	$9.190(10^{-16})$
²³⁸ U	$4.468(10^9)$	4.268	$4.916(10^{-18})$

<u>Isotope</u>	<u>Number of atoms per g^d</u>	<u>Disintegrations per g·s^e</u>	<u>Power watts/g^f</u>
²³² U	$2.596(10^{21})$	$7.746(10^{11})$	5.204
²³³ U	$2.585(10^{21})$	$3.505(10^8)$	$2.756(10^{-4})$
²³⁴ U	$2.574(10^{21})$	$2.308(10^8)$	$1.795(10^{-4})$
²³⁵ U	$2.563(10^{21})$	$8.000(10^4)$	$5.995(10^{-8})$
²³⁶ U	$2.552(10^{21})$	$2.345(10^6)$	$1.718(10^{-6})$
²³⁸ U	$2.531(10^{21})$	$1.244(10^4)$	$8.507(10^{-9})$

^aReference (2b)

^bReference (3a)

^c $dN/dt = -kN$. $k = \ln 2 / \{t_{1/2}(\text{year}) [60 \times 60 \times 24 \times 365.25]\text{s/year}\}$.

^d $6.023(10^{23})/\text{isotopic weight}$.

^e $k \cdot \# \text{ atoms/g}$

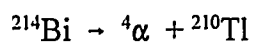
^fDisintegrations $\text{s}^{-1}\text{g}^{-1} \cdot \text{decay energy (MeV)} \cdot 1.602(10^{-13}) \text{ joule/MeV}$.

A-2.3. Decay Sequences

TABLE A-15.
Decay Schemes of the Uranium Isotopes (A)
Table A-15a: ²³⁸U Decay Sequence

<u>Process</u>	<u>$t_{1/2}$</u>
$^{238}\text{U} \rightarrow ^4\alpha + ^{234}\text{Th}$	4.51(10 ⁹) y
$^{234}\text{Th} \rightarrow ^0\beta + ^{234}\text{Pa (m)}$	24.1 d
$^{234}\text{Pa (m)} \rightarrow ^{234}\text{Pa}$	1.17 m
$^{234}\text{Pa} \rightarrow ^0\beta + ^{234}\text{U}$	6.75 h
$^{234}\text{U} \rightarrow ^4\alpha + ^{230}\text{Th}$	2.47 (10 ⁵) y
$^{230}\text{Th} \rightarrow ^4\alpha + ^{226}\text{Ra}$	8.0 (10 ⁴) y
$^{226}\text{Ra} \rightarrow ^4\alpha + ^{222}\text{Rn}$	1600 y
$^{222}\text{Rn} \rightarrow ^4\alpha + ^{218}\text{Po}$	3.823 d
$^{218}\text{Po} \rightarrow ^4\alpha + ^{214}\text{Pb}$	3.05 m
$^{214}\text{Pb} \rightarrow ^0\beta + ^{214}\text{Bi}$	26.8 m
$^{214}\text{Bi} \rightarrow (\alpha + \beta \text{ decay})$	19.7 m

Alpha Decay Subsystem For ^{214}Bi



Beta Decay Subsystem For ^{214}Bi

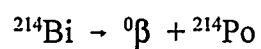
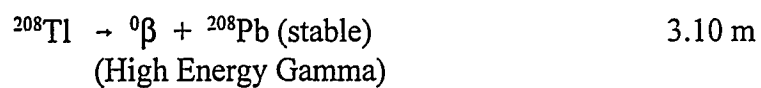
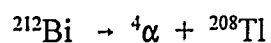


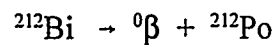
Table A-15b ^{236}U Decay Sequence

<u>Process</u>	<u>$T_{1/2}$</u>
$^{236}\text{U} \rightarrow {}^4\alpha + ^{232}\text{Th}$	$2.39(10^7) \text{ y}$
$^{232}\text{Th} \rightarrow {}^4\alpha + ^{228}\text{Ra}$	$1.41(10^{10}) \text{ y}$
$^{228}\text{Ra} \rightarrow {}^0\beta + ^{228}\text{Ac}$	5.77 y
$^{228}\text{Ac} \rightarrow {}^0\beta + ^{228}\text{Th}$	6.13 h
$^{228}\text{Th} \rightarrow {}^4\alpha + ^{224}\text{Ra}$	1.913 y
$^{224}\text{Ra} \rightarrow {}^4\alpha + ^{220}\text{Rn}$	3.64 d
$^{220}\text{Rn} \rightarrow {}^4\alpha + ^{216}\text{Po}$	55 s
$^{216}\text{Po} \rightarrow {}^4\alpha + ^{212}\text{Pb}$	0.15 s
$^{212}\text{Pb} \rightarrow {}^0\beta + ^{212}\text{Bi}$	10.64 h
$^{212}\text{Bi} \rightarrow (\alpha \text{ \& } \beta \text{ decay})$	60.6 m

Alpha Decay Subsystem



Beta Decay Subsystem



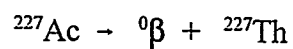
^aData from Reference 3c.

^b $\tau_{1/2}$ = the half life.

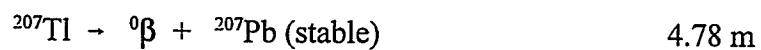
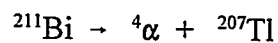
Table A-15c ²³⁵U Decay Sequence

<u>Process</u>	<u>$\tau_{1/2}$</u>
$^{235}\text{U} \rightarrow ^4\alpha + ^{231}\text{Th}$	$7.1(10^8) \text{ y}$
$^{231}\text{Th} \rightarrow ^0\beta + ^{231}\text{Pa}$	25.5 h
$^{231}\text{Pa} \rightarrow ^4\alpha + ^{227}\text{Ac}$	$3.25(10^4) \text{ y}$
$^{227}\text{Ac} \rightarrow (\alpha \text{ and } \beta \text{ decay})$	21.6 y

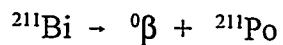
Beta Decay Subsystem (98.6 percent) for ^{227}Ac



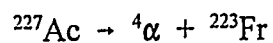
Alpha Decay Secondary Subsystem (99.72 percent) for ^{211}Bi



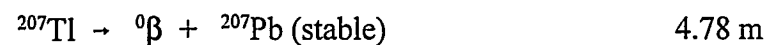
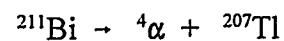
Beta Decay Secondary Subsystem (0.28 percent) for ^{211}Bi



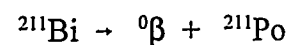
Alpha Decay Subsystem (1.4 percent) for ^{227}Ac



Alpha Decay Seconeary Subsystem (99.72 percent) for ^{211}Bi



Beta Decay Secondary Subsystem (0.28 percent) for ^{211}Bi



^aData from reference 3c.

^b $\tau_{1/2}$ = the half-life.

Table A-15d ^{233}U Decay Sequence

<u>Process</u>	<u>$T_{1/2}$</u>
$^{233}\text{U} \rightarrow ^4\alpha + ^{229}\text{Th}$	$1.62(10^5) \text{ y}$
$^{229}\text{Th} \rightarrow ^4\alpha + ^{225}\text{Ra}$	7340 y
$^{225}\text{Ra} \rightarrow ^0\beta + ^{225}\text{Ac}$	14.8 d
$^{225}\text{Ac} \rightarrow ^4\alpha + ^{221}\text{Fr}$	10.0 d
$^{221}\text{Fr} \rightarrow ^4\alpha + ^{217}\text{At}$	4.8 d
$^{217}\text{At} \rightarrow ^4\alpha + ^{213}\text{Bi}$	0.032 s
$^{213}\text{Bi} \rightarrow (\text{alpha and beta decay})$	47 m
Beta Decay System (97.8 percent)	
$^{213}\text{Bi} \rightarrow ^0\beta + ^{213}\text{Po}$	
$^{213}\text{Po} \rightarrow ^4\alpha + ^{209}\text{Pb}$	$4.2(10^{-6}) \text{ s}$
$^{209}\text{Pb} \rightarrow ^0\beta + ^{209}\text{Bi} (\text{stable})$	3.30 h
Alpha Decay System (2.2 percent)	
$^{213}\text{Bi} \rightarrow ^4\alpha + ^{209}\text{Tl}$	
$^{209}\text{Tl} \rightarrow ^0\beta + ^{209}\text{Pb}$	2.2 m
$^{209}\text{Pb} \rightarrow ^0\beta + ^{209}\text{Bi} (\text{stable})$	3.30 h

^aData from Reference 3c.

^b $T_{1/2}$ = the half-life.

Table A-15e ^{232}U Decay Sequence

<u>Process</u>	$T_{1/2}$
$^{232}\text{U} \rightarrow ^4\alpha + ^{228}\text{Th}$	73.6 y
$^{228}\text{Th} \rightarrow ^4\alpha + ^{224}\text{Ra}$	1.913 y
$^{224}\text{Ra} \rightarrow ^4\alpha + ^{220}\text{Rn}$	3.64 d
$^{220}\text{Rn} \rightarrow ^4\alpha + ^{216}\text{Po}$	55 s
$^{216}\text{Po} \rightarrow ^4\alpha + ^{212}\text{Pb}$	0.15 s
$^{212}\text{Pb} \rightarrow ^0\beta + ^{212}\text{Bi}$	10.64 h
$^{212}\text{Bi} \rightarrow (\alpha \text{ \& } \beta \text{ decay})$	60.6 m

Alpha Decay Subsystem for ^{212}Bi

$^{212}\text{Bi} \rightarrow ^4\alpha + ^{208}\text{Tl}$	
$^{208}\text{Tl} \rightarrow ^0\beta + ^{208}\text{Pb} \text{ (stable)}$ (High Energy Gamma)	3.10 m

Beta Decay Subsystem for ^{212}Bi

$^{212}\text{Bi} \rightarrow ^0\beta + ^{212}\text{Po}$	
$^{212}\text{Po} \rightarrow ^4\alpha + ^{208}\text{Pb} \text{ (stable)}$	$3.04(10^{-7}) \text{ s}$

^aData from Reference 3c.

^b $\tau_{1/2}$ = the half life.

A-3.1.1 Calculated Corrosion Rates For Hollow Uranium Cylinders

Rates of uranium corrosion were calculated for right-circular uranium cylinders having outer radii of 6.35 cm and inner radii of 4.445 cm, and a mass of 18.00 kg, as described in Section 4.2. The surface area was taken to be 1,333 cm². Rate constants were from the equations given in Table 6 of the text (Section 3.1.1). The conversion of units from corrosion rate constants in mg(U)·cm⁻²·Hr⁻¹ to annual corrosion rate of a standard cylinder in g(U)·year⁻¹ was based on the formula:

$$\text{Rate (g/year)} = \text{Rate [mg(U)·cm}^{-2}\text{Hr}^{-1}] \cdot 10^{-3} \text{g/mg} \cdot 1131^{-2} \cdot [24 \cdot 365.25] \text{Hr/year},$$

$$\text{or Rate (g/yr)} = \text{Rate [mg(U)]cm}^{-2}\text{hr}^{-1} \cdot 9.9319(10^3).$$

Table A-16.
Calculated Corrosion Rates
Standard Cast Uranium Cylinder^a

Temperature (°C)	Net Annual Mass of Uranium Corroded (g)			
	Dry Air	Water Vapor (no ₂) RH = 100%	Water Vapor plus ₂ RH = 100%	Water Vapor plus ₂ RH = 2-90%
15	0.0906	108.7	1.447	0.052
20	0.156	164.0	2.458	0.110
25	0.264	244.0	4.103	0.226
30	0.441	358.2	7.73	0.453
35	0.721	519.4	10.88	0.889
40	1.161	744	17.29	1.706
45	1.844	1055	27.11	3.206
50	2.886	1478	41.91	5.911

^a Rates were calculated by using the rate constants from Table 7, Section 3.1.1. The surface area of an 18-kg part was taken as 1,133 cm² (see Appendix Section A-5.1.3.). Calculated values are the net weight of uranium, in g, that corrodes per year, per part.

A-5.1.3 Consequences of Can Leakage or Improper Handling

Baseline Cases 1 and 2. No Container Leakage

In the trivial case of a perfectly sealed can containing a pure inert gas, no corrosion will occur.

If a perfectly sealed can contains dry air, the volume of oxygen is the product of the free volume in the can (see text, Section 5.1.3) multiplied by the volume fraction of oxygen in air (20.95 percent according to reference 3b), or

$$\text{Vol (O}_2\text{)} = 3,224 \text{ cm}^3 \cdot 0.2095 = 675.4 \text{ cm}^3.$$

For a perfect gas at one atmosphere net pressure and 298°C, this corresponds to 0.0276 moles O₂. If the corrosion product is UO₂, this same number of moles of uranium (which is equivalent to 6.49 g uranium) will eventually be converted to oxide. Because 18-kg of 93 percent EU contains 76.53 moles of uranium, the fraction of metal eventually converted to oxide is $0.0276/76.53 = 0.00036$.

The initial rate of corrosion can be calculated by the expression given for a dry air environment in Table 6, Section 3.1.1:

$$\text{Rate} = 6.9 \cdot 10^8 \cdot \exp(-18300/RT) \text{ (mg U corroded/cm}^2 \text{ h)}$$

at 298K (25°C), the rate is $2.61(10^{-5}) \text{ mg U/cm}^2 \text{ h}$.

The specific surface area of the hollow right circular cylinders described in Section 4.2 can be calculated as follows:

$$\begin{aligned} \text{Surface area} &= 2 \pi \cdot 6.35 \cdot 14.8 \quad (\text{outer contour}) \\ &+ 2 \pi \cdot 4.45 \cdot 14.8 \quad (\text{inner contour}) \\ &+ 2 \pi \cdot [6.35^2 - 4.45^2] \quad (\text{two planar ends}) \\ &= 1,133 \text{ cm}^2 \end{aligned}$$

Therefore the initial rate of corrosion is $2.61(10^{-5}) \text{ mg U/cm}^2 \text{ h} \cdot 1,133 \text{ cm}^2 = 2.9610^{-2} \text{ mg/h}$. This rate of corrosion is to be expected to prevail until most of the oxygen is depleted from the can, at which point the net amount of uranium converted to oxide asymptotically approaches the value of 6.49 g of uranium calculated above.

Special Case 1. Pressure-driven dry air in-leakage, infinite time.

The limiting amount of dry air corrosion possible for a uranium casting in a standard storage can containing a very small leak is calculated as follows. The leak is taken to be small enough that significant in-leakage occurs only under a positive pressure differential between the can and the surrounding atmosphere. Since uranium does not react with nitrogen under ambient conditions, the decrease in pressure is solely dependent on the amount of oxygen consumed by the corrosion process. The fraction of oxygen in dry air is 20.95 percent^{3b}. As this amount of oxygen (designated x) is removed by corrosion, it is replaced by the same quantity of air which has the concentration x^2 inside the can prior to its being removed in turn. The volume fraction of oxygen that eventually reacts is

$$x + x^2 + x^3 + x^4 + \dots = \sum_{i=1}^{\infty} x^i,$$

which is an infinite series given by $x/(1-x)$. The sum of the infinite series is calculated to be 0.2650. This is the fraction by volume of the gas molecules within the can that would be converted to oxide if no moisture were involved and dry air continued to diffuse into the can until the residual pressure from nitrogen was equal to the external pressure. The product of the free volume of the can (3,224 cm³ from Section 5.1.3) multiplied by the value of the above series expansion gives the volume of oxygen converted to oxide, or 854 cm³. For an ideal gas at one atmosphere pressure and 298°C, 854 cm³ corresponds to 0.0350 moles. If the oxide produced is assumed to be UO₂, this same number of moles is converted to oxide. The mass 18-kg of ²³⁵U contains 76.53 moles of uranium, so the fraction of metal converted to UO₂ under these conditions is $0.035/76.53 = 0.00046$.

A-6.2.3 Pressurization Due To Helium Generation

Details for the calculations of the number of Becquerels per g were given in Section A-2.2.1. The value calculated for ²³⁴U was 2.308(10⁸) Bq/g, or 2.308(10⁸) alphas per g per second. Then for 18 kg of oralloy containing 1 percent ²³⁴U, the rate is:

$$2.308(10^8) \text{ alpha g}^{-1} \text{ s}^{-1} \cdot 18 \text{ kg(oy)} \cdot 0.01 \text{ g}^{238}\text{U/g(oy)} \cdot 10^3 \text{ g/kg} = 4.15(10^{10}) \text{ alpha/s},$$

which corresponds to 1.311(10¹⁸) helium atoms per year. The expression (oy) in the equation refers to oralloy. Dividing by Avogadro's number gives 2.177(10⁻⁶) moles of helium produced per year. Assuming ideal gas behavior, a mole of helium at standard temperature and pressure occupies 22,400 cm³. Then the rate of pressurization is 4.876(10⁻²) cm³·atm/year. Since the free volume within the storage cans is 3,224 cm³ (Section 5.1.3), net fractional pressurization due to helium evolution is only 1.51 10⁻⁵ year⁻¹.

A-6.2.4 Pressurization due to Radiolytic Activity

The decay schemes of the artificial uranium isotopes are shown in Section A-2.3. of the Appendix. To summarize the significance of the data (1) ²³²U is extremely radioactive, and the nuclei in its decay sequence are even more short-lived. The presence of even a small quantity of ²³²U would be sufficient to cause very rapid radiolytic decay of any water of hydration; (2) ²³³U has a half-life comparable with that of ²³⁴U, and its decay products likewise are comparable with those of ²³⁴U.

From a pragmatic point of view, the more serious concern in storing ^{233}U is the high likelihood that it contains a significant quantity of ^{232}U ; and (3) ^{236}U has a long half-life and its immediate decay product, ^{232}Th , is more stable than any of the uranium isotopes. It becomes a radiolysis hazard only after long-term storage has allowed the ^{232}Th concentration to reach appreciable levels.

The net potential quantity of hydrogen that can be liberated due to radiolytic decomposition can be calculated as follows. If 18.00 kg of partially hydrated UO_3 were stored in a can having a net internal volume of $4,180 \text{ cm}^3$ (see Section 5.1.3), the volume occupied by the powder would be $18,000 \text{ g} / 7.30 \text{ g/cm}^3 = 2,466 \text{ cm}^3$. The residual free volume would be $4,180 \text{ cm}^3 - 2,466 \text{ cm}^3 = 1,714 \text{ cm}^3$. Assuming ideal gas behavior, $PV = nRT$, with $P = 1 \text{ atm}$, $V = 1,714 \text{ cm}^3$, $R = 0.08206 \text{ L}\cdot\text{atm./mole deg}$, and $T = 298 \text{ K}$ (25°C), the number of moles of gas that would generate one atmosphere pressures 0.0701. The quantity of gas that could be generated by the radiation-induced decay of water is determined by assuming that all of the water present is dissociated to H_2 and O_2 . Thus, one mole of water yields one mole of H_2 and one half mole of O_2 . Consequently, the amount of water in 18 kg of UO_3 that would yield one atmosphere pressure is $2/3 \cdot 0.0701 = 0.0467 \text{ mole}$. The molecular weight of water is 18, so the weight of water allowed in the 18 kg of UO_3 is $0.0467 \cdot 18 = 0.841 \text{ g}$. This corresponds to only 47 ppm water in 18 kg of UO_3 .

The rate at which the water of hydration undergoes radiolysis is dependent on the radiolytic field. In Section (A-6.2.3) it was shown that alpha decay in oralloy occurs at the rate of $4.151(10^{10})$ decays per second. From ref 47, ^{234}U emits alphas at four energy levels clustered around 4.7 MeV. As noted in Section 2.3.1 and reference 48a, about 35 eV is expended per ion pair produced by alpha attenuation. Then the rate at which ion pairs are formed is:

$$4.151(10^{10})\alpha/\text{s} \cdot 4.7(10^6) \text{ eV}/\alpha / (35 \text{ eV/ion pair}) = 5.57(10^{15}) \text{ ion pair/s},$$

which corresponds to $1.83(10^{22})$ ion pairs per year, or 0.0303 moles of ion pairs per year.

Continuing the calculation to determine how the rate of ion pair formation translates into hydrogen formation rate would involve a major modelling exercise. Two limiting boundary cases can be defined.

1. In the limit where each ion pair results in liberation of a molecule of hydrogen plus half a molecule of oxygen, the number of moles of gas needed to yield a net can pressurization of one atmosphere is approximated by dividing the free volume [$1,714 \text{ cm}^3$, calculated earlier in this section] by the volume of a mole of ideal gas, $22,400 \text{ cm}^3$, giving 0.144 mole. Under this scenario, the time required to generate one atmosphere of pressurization is $[0.144 \text{ mole gas}] / [(0.0303 \text{ mole ion pairs/year}) \cdot (1.5 \text{ mole gas/mole ion pairs})]$ or 3.16 years.
2. In the limit where the number of ion pairs formed is equal to the number of molecules in the can, an average molecule (whether uranium oxide or water) has been involved in one instance of ion pair formation. The number of moles in the can is $18 \text{ kg } (\text{UO}_3) / (.283 \text{ kg/g mole}) = 63.6 \text{ moles}$. Then the time required to fulfill this condition is $[63.6 \text{ mole gas}] / [(0.0303 \text{ mole ion pairs/year}) \cdot (1.5 \text{ mole gas/mole ion pairs})] = 1,400 \text{ years}$.

In the absence of either a modelling study that refines the uncertainty between boundary cases (1) and (2) or the acquisition of reliable experimental data, it is considered prudent to avoid the consequences of excessive pressurization by prohibiting the long-term storage of chemical forms that retain water such as UO_3 .

A-6.2.5 Pressurization Due to Hydrogen Evolution

Case 1. Net Pressure Buildup for a Uranium Casting in a Standard Storage Can Due to Enclosed Moisture.

From Section 5.1.3, the free volume of the PLMS storage cans is $3,224 \text{ cm}^3$. The number of moles of hydrogen required to cause a net pressurization of 1.0 atmosphere can be estimated from the ideal gas equation:

$$n = PV/RT = 1.0 \text{ atm} \cdot 3.224 \text{ L} / (0.082 \text{ L} \cdot \text{atm} / \text{mole deg}) \cdot 298 = 0.132 \text{ mole}.$$

Since one mole of water can liberate one mole of hydrogen, the amount of water (molecular weight = 18) needed to cause a can pressurization of 1.0 atmosphere is $18 \cdot 0.132 = 2.37 \text{ g}$. Alternately stated, the net possible pressure buildup due to inadvertent entrapment of water in a standard storage can is $1.0 \text{ atm} / 2.37 \text{ g} = 0.42 \text{ atmosphere per g of water}$. The figure is diminished to the extent that uranium hydride formation occurs.

Case 2. Rate of Hydrogen Pressurization Due to Enclosed Moisture (Absence of Oxygen Assumed)

From Section 3.1.1, the rate of corrosion of uranium in water vapor in the absence of oxygen is given by:

$$\text{Rate (mg U reacted cm}^{-2} \text{ h}^{-1}) = 3.2 \cdot 10^8 \exp(-13,800/RT),$$

which yields $2.42 (10^{-2}) \text{ mg (U) cm}^{-2} \text{ h}^{-1}$ at 298K. For a surface area of 1133.05 cm^2 (see Appendix, Section A-5.1.3), the rate is 27.4 mg (U)/h . For oralloy with an atomic weight of 235.2, the molar rate of corrosion is $27.4/235.2 = 1.16(10^{-4}) \text{ mole(U)/h}$. The ratio of moles H_2 generated per mole U corroded is taken to be 2.05, which is the median value from the range indicated in Section 3.1.1. Then the rate of hydrogen evolution is

$$1.16(10^{-4}) \text{ mole (U)/h} \cdot 2.05 \text{ mole(H}_2\text{/mole (U))} = 2.39(10^{-4}) \text{ mole H}_2\text{/h}.$$

Using the perfect gas approximation, the hydrogen evolution rate in the storage cans with a free volume of $3,224 \text{ cm}^3$ is

$$2.39(10^{-4}) \text{ mole H}_2\text{/h} \cdot 0.082 (\text{L} \cdot \text{atm/mole} \cdot \text{deg}) \cdot 298 \text{ deg} / 3.224 \text{ L} = 1.81 (10^{-3}) \text{ atm/h}.$$

A-7.1.3 Recommended Chemical/Physical Forms for Long-Term Storage

The following three sample calculations show selected material shapes to determine whether they would qualify as acceptable on the basis of having a specific surface area less than $1.00 \text{ cm}^2/\text{g}$.

1. The specific surface area of the hollow, right-circular cylinders described at the beginning of Section 7.1.3 can be calculated based on the surface area of 1133.05 cm^2 (calculated in Appendix Section A-5.1.3, case 2) and the mass of 18,000 g, or $1133.05/18000 = 0.063 \text{ cm}^2/\text{g}$.
2. A uranium metal cube, 1.00 cm on a side, has a volume of 1.00 cm^3 and a mass of about 19.0 g/cm^3 (depending on isotopic content). The surface area is 6.00 cm^2 . The specific surface area is then $6.00/19.0 = 0.316 \text{ cm}^2/\text{g}$.
3. A square ($1 \text{ cm} \times 1 \text{ cm}$) segment of a planar uranium metal foil of 0.100 cm thickness has a surface area of 2.4 cm^2 , a volume of 0.100 cm^3 , and a mass of 1.90 g. The specific surface area is $2.4/1.90 = 1.26 \text{ cm}^2/\text{g}$. Thus, a waiver should not be granted to allow long-term storage of a 0.100-cm-thick uranium foil.