Y/LB-15,913

Y-12

OAK RIDGE
Y-12
PLANT

MARTIN MARIETTA

POSITION PAPER

OAK RIDGE Y-12 PLANT

STORAGE OF URANIUM

IN PLASTICS

W.K. Duerksen

July 1995

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

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

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PREFACE

As a result of the end of the Cold War, the United States nuclear weapon stockpile is being reduced from approximately 20,000 warheads to fewer than 10,000 by the end of the century.[1] The Oak Ridge Y-12 Plant is the Department of Energy (DOE) site charged with the responsibility of providing safe, secure storage for the uranium recovered from these weapons. In addition to weapons material, Y-12 has traditionally processed and stored uranium from nonweapon programs and presumably will continue to do so. The purpose of this document is to evaluate the suitability of plastics for use in the containment of uranium.

1. Y-12 URANIUM STORAGE MISSION

1.1 GENERAL STATEMENT

The Y-12 Plant is not authorized by DOE or by the state of Tennessee to treat or store generalized radioactive waste. From the point of view of materials acceptance, this means that Y-12 is not allowed to accept materials from outside sources that it is not capable of treating (i.e., purifying) with existing process equipment and technology. Thus, uranium that is contaminated with significant concentrations of transuranics (actinide elements of higher atomic number than 92), radioactive decay products, and fission products can not be accepted at Y-12 for storage or processing.

1.2 CATEGORIZATION BY ISOTOPIC ENRICHMENT

Table 1 lists the terminology that is commonly used to describe uranium at various isotopic enrichment levels. Unfortunately, the terminology is not universally accepted and confusion often exists when the isotopic enrichment is not explicitly stated. The most common enrichment level recovered from dismantled nuclear weapons is 93 percent. Metal of this enrichment was traditionally called Oak Ridge alloy (oralloy).

1.3 CATEGORIZATION BY POINT OF ORIGIN

The Y-12 Plant has traditionally been the recipient of enriched uranium (EU) that is declared excess at other DOE sites.[1] Reference 1 lists 23 DOE sites that currently have surplus inventories of EU. Details given indicate that the inventory of highly enriched uranium (HEU) stored at Y-12 as of September 1994 was 168.9 metric tons. It was estimated that the upper bound for material received within the next ten years from outside sources other than Pantex is 98.4 metric tons.[1] The quantity to be received from Pantex is classified. Approximately five metric tons of HEU from foreign sources may be stored at Y-12 due to implementation of the United States' nonproliferation policy or related national security considerations.[1] The quantity of low enriched uranium to be received at Y-12 is open to speculation, but the upper bound was set at over 7,100 metric tons.[1]

Table 1.
Standard Terminology for Uranium Isotopic Assays

Designation	Acronym	^{235}U Assay
Depleted uranium	DU	<0.713%
Natural uranium	NU	0.713%
Low enrichment 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%

In addition to uranium at Y-12 and at other DOE sites, small quantities of DOE-owned EU are situated at diverse institutions including laboratories and universities. These materials are shipped to Y-12 under the auspices of the DOE Central Scrap Management Office (CSMO). The net quantity of uranium received from the private sources is small compared with the total site inventory, but the logistics of storage or conversion are complicated due to the diversity of the materials, the sometimes uncertain history (i.e., the possibility that a portion of the material may have been irradiated), and a lack of standardization in the packaging practices of the shippers. Many of these materials are shipped to Y-12 in plastic containers.

Another category of material that requires special attention is enriched material transferred to Y-12 from nations comprising the former Soviet Union. Materials of this type present special challenges because: (1) they are likely to contain higher quantities of transuranics and fission products than are present in domestic materials, and (2) they are often in chemical forms that have not traditionally been processed at Y-12.

1.4 CATEGORIZATION BY CHEMICAL AND PHYSICAL STATE

The chemical forms of HEU that are considered for prolonged long-term storage are metal, alloys that are comparable or superior to metal in terms of corrosion resistance and ignitability, and the oxide U_3O_8. However, the range of materials that are in interim storage or in-process storage is very great and includes such diverse materials as alloys, various oxides, oxalate, diuranate, nitrates, halides, sulfates, organic solutions, loaded ion-exchange resins, and others. They are also present in a variety of physical forms varying from full-density castings to loose powders to solutions.

1.5 STORAGE CONFIGURATIONS

The acceptable forms of HEU for prolonged low-maintenance storage (PLMS) are full-density cast metal or metal alloy and oxide (U_3O_8) powder. The HEU metal is prepared for long-term storage by casting into hollow cylinders, which are sealed in stainless steel cans. The canned cylinders are stored in concrete arrays, or vaults, that are engineered to preclude the possibility of allowing the material to reach a critical configuration. Uranium oxide (U_3O_8) is stored as a loose powder in the same type of container.

The PLMS configuration represents the best attainable means of storing large quantities of EU in terms of the following criteria:

1. preventing the occurrence of a criticality event under even the most severe circumstances;
2. safeguarding the health and safety of the public;
3. safeguarding the environment;
4. maximizing security, subject to maintaining a required level of transparency for international inspection teams and for inventories;
5. safeguarding the health and safety of the workers;
6. maintaining the material in the state in which it was initially placed in storage; and
7. cost minimization.

Plastics are not the materials of choice for PLMS. The possibility of fire can be reduced to virtually zero if the entire PLMS facility and its contents are limited to concrete, steel, uranium oxide, and cast uranium metal. This strategy makes it tenable to build the facility without fire sprinklers and thus preclude the possibility of a criticality excursion due to neutron moderation and reflection by water.

The storage situations where plastics are desirable are interim storage and in-process storage. There are currently large quantities of material in these classifications due to high backlogs, high anticipated volumes of incoming material, low-equity materials, unusual isotopic compositions, materials being stored for CSMO, and uncommon chemical forms. The maximum time to be allowed for maintaining EU in interim storage is ten years. The highest priority in designing and maintaining the interim and in-process storage facilities is given to maintaining nuclear criticality safety. All storage practices are required to be covered by a Criticality Safety Approval (CSA). Storage arrays commonly called "bird cages" are frequently specified in the CSAs. These are rigid lattices designed to maintain the EU in a noncritical geometry under design basis accident scenarios.

Storage of depleted uranium does not require the stringent controls that apply to EU because it is not fissionable and has little economic value. Depleted uranium metal and its alloys are stored in the form of massive cast billets. Depleted uranium machine turnings are oxidized to U_3O_8 in a dedicated facility known as the Uranium Chip Oxidizer Facility (UCOF). Oxides are stored as a loose powder in two large concrete vaults. Moist, finely divided uranium metal powder such as saw fines or sludges cannot be safely oxidized in the UCOF because of the explosion hazard associated with entrained water and cutting fluids. This material is blended with the oxide and transferred to the oxide vaults.

2. PROPERTIES OF POLYMERS (PLASTICS)

2.1 POLYMERS

The two types of plastics currently being used extensively in the Y-12 uranium storage areas are polyethylene and polyvinylchloride (PVC). Polyethylene bottles are well suited for transfer or storage of chemicals because they are flexible enough to deform without breaking under mechanical stress. Packaging of chemicals in polyethylene bottles is a well-established industrial practice, justified by the very long-term service life of polyethhylene containers as long as they are protected from long-term, direct exposure to sunlight. Polyvinylchloride film is a convenient material for providing a protective wrap and is readily heat-sealed.

2.2 PROPERTIES OF POLYETHYLENE

Polyethylene is formed by the polymerization of ethylene under carefully specified conditions of pressure, temperature, and catalysis.[2,15a] The chemical reaction is:

$$n\ (CH_2 = CH_2) \rightarrow (-CH_2\text{-}CH_2\text{-})_n$$

The specific properties of polyethylene vary according to the methodology used in its manufacture. The molecular weight typically ranges from 1,500 to 100,000; its density ranges from 0.91 to 0.97. The flash point and auto ignition temperatures are 341°C and 349°C, respectively.[3] It is nonvolatile and resistant to chemical attack under most circumstances. It is slightly permeable to atmospheric gases including water vapor.[15b] Polyethylene decomposes gradually when exposed to ultraviolet light.

2.3 PROPERTIES OF POLYVINYLCHLORIDE

Polyvinylchloride is formed by the polymerization of the vinyl chloride monomer:

$$n\ (CH_2 = CHCl) \rightarrow (-CH_2\text{-}CHCl\text{-})_n$$

It is nonflammable.[3] Stabilizers are incorporated into the polymer to prevent discoloration due to exposure to heat or light.[3] The PVC bags can conveniently be sealed by heating.

Radiolysis of chlorinated aliphatics tends to cause breaking of the C-Cl bond, simply because the C-Cl bond is weaker than either C-C or C-H. The liberated chlorine atom reacts rapidly with the substrate to yield HCl.[4j]

2.4 DEGRADATION OF POLYMERS

A discussion of the degradation of polymers due to alpha, beta, gamma, and neutron irradiation is deferred to subsequent sections. This section deals with the degradation of polyethylene and polyvinylchloride under normal environmental conditions.

In an outdoor environment, plastics degrade primarily due to photochemically induced oxidation.[5a] Other contributing factors are mechanical abrasion by wind, dust, and rain; but these are not serious considerations for materials in an indoor environment such as pertains to EU storage. Biodegradation of polyethylene is generally not a serious consideration. Alkanes of molecular weights of 451 and below (i.e., 32 carbons or less) show evidence of biodegradation when tested by standard ASTM methods, but larger polymer molecules (which make up the greater part of most industrial polyethylene) are unaffected.[5b]

Photochemical oxidation is initiated when a photon of ultraviolet light is absorbed in the polyethylene matrix and generates a radical (a species containing an unpaired electron), designated R·. Oxidation then proceeds according to the repetitive sequence:

$$R\cdot + O_2 \rightarrow ROO\cdot \qquad (2.1)$$

$$ROO\cdot + RH \rightarrow ROOH + R\cdot, \qquad (2.2)$$

where the R· from reaction (2.2) then reinitiates reaction (2.1). This reactive sequence continues until it is terminated by a reaction in which two radicals combine. Ordinarily, ultraviolet light having a wave length of about 300 nm initiates most of the radical formation that occurs in polyethylene. Photons of significantly lower wavelength (higher energy) than 300 nm are filtered-out of sunlight on passage through the earth's upper atmosphere, whereas photons of significantly higher wavelength (lower energy) than 300 nm simply do not have sufficient energy to cause radical formation.

Mechanical degradation of polyethylene containers by photochemical oxidation while in a typical EU storage environment is expected to be minimal for the following reasons:

1. the exposure to ultraviolet light in an indoor storage facility is much lower than that in sunlight;
2. even under outdoor conditions, polyethylene lasts for several years before being significantly degraded;
3. many commercial grades of polyethylene contain ultraviolet absorbers or pigments to minimize radical formation (although recently, some polyethylene food containers have been manufactured containing photoactivators that are deliberately added to accelerate photochemical oxidation as an environmental measure); and
4. the maximum permissible term of interim storage for EU at Y-12 is defined as ten years.

2.5 SAMPLE CALCULATION: PHOTOCHEMICAL DEGRADATION

As noted in the prior section, the wavelength at which polyethylene shows maximum sensitivity to ultraviolet light is 300 mμ.[5c] Typically, 1.21 percent of the radiant energy in sunlight at the earth's surface lies at wavelengths of 300 mμ or lower.[10b] The total annual solar flux at the latitude of Oak Ridge is typically about 209.4 kcal·cm^{-2}·year^{-1}.[10c] Then the annual flux of solar ultraviolet light of wavelength < 300 mμ is:

209.4 kcal cm^{-2} year^{-1} ·0.0121 = 2.533 kcal cm^{-2} year^{-1}

which equals 1.060(10^{11}) erg cm^{-2} year^{-1}.

The energy of a photon having a wavelength of 300 mμ is $h\nu = hc/\lambda$,
or:

E = 6.626(10^{-27})erg sec · 3.00(10^{10})cm s^{-1} / 3.0(10^{-5})cm = 6.626(10^{-12}) erg.

Then:

Number of photons ($E > h\nu^*$)/cm^2 year = 1.060(10^{11})/6.625(10^{-12}) = 1.6(10^{22}) activations/cm^2 year.

Where ν^* is the frequency corresponding to the wave length 300 mμ, or 1.0(10^{15}) s^{-1}.

This number can be compared with the number of C_2H_4 units (or mers) in a volume element 1 cm ×1 cm × 0.1 cm, or 0.1 cm^3. With a density of about 0.95 g/cm^3, the weight of the volume element is 0.095 g. The molecular weight of the mer is 28 g/mole, so the volume element contains 0.095/28 = 0.0034 moles of the mer, or 2.04 (10^{21}) mers. Therefore, the sunlight falling at normal incidence on a 1 × 1-cm strip of polyethylene that was 0.10 cm thick for one year would contain nearly ten times more photons capable of initiating radical formation than the number of mers in the strip.

3. BASIC URANIUM RADIATION PHYSICS

A consideration of radiation physics is needed to answer the following questions: (1) what radiation fields will plastics be exposed to when they are used for uranium storage? (2) will these radiation fields degrade the plastics? (3) will potentially harmful gases be produced by the interaction of the radiation fields with the plastics? And (4) what plastics should and should not be used for uranium storage?

3.1 DECAY SEQUENCES FOR URANIUM ISOTOPES

Tables 2 (A through F) show the decay sequences of the uranium isotopes. The tables were truncated at the point where the terminal nuclide has a half-life that is long when compared with the amount of time that has lapsed since the uranium in storage has been processed (and thus purified of decay products).[6a] Thus the nuclear disintegrations listed in the tables are the primary processes that govern the net radiation field for materials that are in storage at Y-12 and that contain approximately equilibrium concentration levels of entrained decay products but have not yet accumulated a sufficiently high concentration of the more stable isotopes (e.g., half-lives greater than 10^4-10^5 years) to contribute significantly to the net disintegration rate.

Naturally occurring uranium contains only the three isotopes ^{238}U, ^{235}U, and ^{234}U. The other isotopes listed are formed in nuclear reactors due to neutron capture by thorium or ^{235}U. The uranium in the nuclear weapons complex was historically isolated from irradiated material. Thus data for the isotopes ^{236}U, ^{233}U, and ^{232}U are included in this document only for reference purposes, and not in anticipation that materials containing significant concentrations of these isotopes will ever be stored at Y-12. The current criteria for acceptance or rejection of incoming uranium by Y-12 are from the Egli Report.[7] A revised set of criteria for uranium acceptance at Y-12 is currently being prepared.[8] Data for the isotopes ^{237}U, ^{239}U, and ^{240}U were not included because their half-lives are a few days or less, thus significant concentrations do not exist for long enough time periods to be of interest in a storage situation. Reference 9 is an excellent source for the energies of the alpha and beta particles and gamma rays emitted by the nuclear disintegrations and for the energies of the secondary electrons that accompany alpha decay.

3.2 CALCULATION OF RADIOACTIVE EMISSIONS

The data from Tables 2 (A to F) can be used to calculate the net radiation fields generated by uranium metal samples of known mass and isotopic composition as will be described in Sections 3.2.1 through 3.2.3.

Table 2.
Decay Schemes of the Uranium Isotopes (a)

Table 2A. ^{232}U Decay Sequence

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

Alpha Decay Subsystem for ^{212}Bi

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

Beta Decay Subsystem for ^{212}Bi

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

Table 2B. ^{233}U Decay Sequence

Process	$\tau_{1/2}$
$^{233}U \rightarrow {}^{4}\alpha + {}^{229}Th$	$1.62(10^{5})$ y
$^{229}Th \rightarrow {}^{4}\alpha + {}^{225}Ra$	7340 y

Table 2C. ^{234}U Decay Sequence

Process	$\tau_{1/2}$
$^{234}U \rightarrow {}^{4}\alpha + {}^{230}Th$	2.47 (10^5) y
$^{230}Th \rightarrow {}^{4}\alpha + {}^{226}Ra$	8.0 (10^4) y

Table 2D. ^{235}U Decay Sequence

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

Table 2E. ^{236}U Decay Sequence

Process	$\tau_{1/2}$
$^{236}U \rightarrow {}^{4}\alpha + {}^{232}Th$	2.39(10^7) y
$^{232}Th \rightarrow {}^{4}\alpha + {}^{228}Ra$	1.41(10^{10}) y

Table 2F. ^{238}U Decay Sequence

Process	$\tau_{1/2}$
$^{238}U \rightarrow {}^{4}\alpha + {}^{234}Th$	4.51(10^9) y
$^{234}Th \rightarrow {}^{0}\beta + {}^{234}Pa$	24.1 d
$^{234}Pa \rightarrow {}^{0}\beta + {}^{234}U$	6.75 h
$^{234}U \rightarrow {}^{4}\alpha + {}^{230}Th$	2.47 (10^5) y

(a) References 9 and 10.

3.2.1 First-Order Kinetics:

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

$dN/dt = -kN$, or $dN/N = -kdt$. (3.1)

Integrating, $\text{Ln } N = -kt + C$. (3.2)

When $t = 0$, $N = N_0$ by definition of N_o, so $C = \text{Ln } N_o$.

Then $\text{Ln } N = \text{Ln } N_o - kt$, or $\text{Ln } N/N_o = -kt$ (3.3)

When $t = \tau_{½}$ (the half life), $N = N_o/2$, so

$\text{Ln } [N_o/(N_o/2)] = \text{Ln } 2 = k\tau_{½}$ (3.4)

or $k = \text{Ln}2 /\tau_{½}$ (3.5)

and $dN/dt = -(N \text{ Ln } 2)/\tau_{½}$, (3.6)

3.2.2 Radioactive Decay

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

Counts per second = number of Bq $= (\text{Ln } 2/\tau_{½})\cdot 6.023\ (10^{23})$ (3.7)

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

number of Bq $= (\text{Ln } 2/\tau_{½})\cdot 6.023\ (10^{23})\cdot(M/IW)$ (3.8)

For half-lives expressed in years,

number of Bq $= (\text{Ln } 2/\tau_{½})\cdot 6.023\ (10^{23})\cdot(M(g)/IW)/(365\cdot 24\cdot 60\cdot 60)$ (3.9)

Numerically,

number of Bq $= 1.322(10^{16})\cdot M(g)/(\tau_{½}\cdot IW)$, $\tau_{½}$ in years. (3.10)

Count rates are often expressed in units of the Curie (Ci):

1 Ci $= 3.7(10^{10})$ disint/sec $= 3.7(10^{10})$ Bq (3.11)

Count rate $= 3.5755(10^{5})\cdot M(g)/(\tau_{½}\cdot IW)$ Ci (3.12)

Table 3 gives the calculated count rates for pure isotopes based on Equation (3.10). The values correspond to the net number of disintegrations that occur. They are known as specific activities, and in the case of alpha decay from EU in high-density configurations, the vast majority of the alpha emissions are self-attenuated.

Table 3.
Count Rates for Pure Isotopes (Alpha)

Isotope	$\tau_{1/2}$ (years)	Count rate, Bq g^{-1} (a)
^{232}U	73.6	$7.742(10^{11})$
^{233}U	$1.62(10^{5})$	$3.502(10^{8})$
^{234}U	$2.45(10^{5})$	$2.306(10^{8})$
^{235}U	$7.037(10^{8})$	$7.994(10^{4})$
^{236}U	$2.39(10^{7})$	$2.344(10^{6})$
^{238}U	$4.468(10^{9})$	$1.243(10^{4})$

(a) Equation (3.10).

3.2.3 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 (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 2C, 2D, and 2F, it is seen that the appropriate multipliers are:

$I(\alpha) = 1$ for ^{234}U, (3.13)
$I(\alpha) = 1$, $I(\beta) = 1$ for ^{235}U, and (3.14)
$I(\alpha) = 1$, $I(\beta) = 2$ for ^{238}U. (3.15)

Then the equilibrium alpha and beta count rates are found by multiplying the value from Equation (3.10) by the appropriate multipliers shown above. Calculated values are shown in Table 4.

Table 4.
Count Rates Including Decay Products[a]

	Multipliers		Count Rates, Bq/g	
Uranium Isotope	Alpha	Beta	Alpha	Beta
^{234}U	1	0	2.305 (10^8)	0
^{235}U	1	1	7.994 (10^4)	7.994 (10^4)
^{238}U	1	2	1.243 (10^4)	2.486 (10^4)

From Equation (3.10), with modifications for decay products based on the multipliers listed in Equations (3.13), (3.14), and (3.15).

The net count rates for enriched, natural, and depleted uranium are given in Table 5, based on taking appropriate linear combinations from values in Table 4.

3.3 Attenuation of Radioactive Emissions

This section addresses the following issues: (1) how much of the radiation from radioactive decay will actually impinge on the plastic materials used in storage, (2) how much of this radiation will be absorbed by the plastic, (3) what will be the results of this absorption in terms of the mechanical integrity of the plastic, (4) what degradation products (particularly gases) will be produced due to irradiation of the plastic.

Table 5
Isotopic Blends: Alpha and Beta Activity

	Count Rates (Bg/g)			
	Pure Isotopes		Including Decay Products	
Material	Alpha	Beta	Alpha	Beta
Enriched U[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)

[a] 1% ^{234}U, 93%^{235}U, and 6% ^{238}U.
[b] 0.005% ^{234}U, 0.713% ^{235}U, and 99.275% ^{238}U.
[c] 0.001% ^{234}U, 0.2% ^{235}U, and 99.8% ^{238}U.

3.3.1 Attenuation of Alpha Particles

Alpha particles are energetic helium nuclei; that is, doubly charged ions having no electrons and a nucleus composed of two protons and two neutrons. As seen in Tables 2 (A to F), alpha emission is the initial decay mode for all the uranium isotopes and of several daughter products as well. The alpha particles emitted in the radioactive decay of the uranium isotopes are fairly energetic, with energies in the range 4.0 to 5.4 MeV. However, alpha particles are very readily attenuated in passing through matter, and have a range of only 0.2 cm in air and much shorter values in condensed phases, such as 5(10^{-4}) cm in water.[4a] 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.[4a] Typically, about 35 eV is expended per ion pair produced.[4a] 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 rare cases where they are scattered by collision with a nucleus).

3.3.2 Attenuation of Gamma Rays (and Xrays).

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.[4b] 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,

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

where I is the intensity of the beam transmitted through the absorbing medium, I_o 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 Xrays) 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 which 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.[4c, 11] Thus, plastics do not absorb gamma radiation effectively by photoelectric absorption.

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. Every 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.[4d, 11]

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 $2 \cdot 9.109(10^{-28})g \cdot [3(10^{10})cm\ s^{-1}]^2 = 1.640(10\text{-}6)erg = 1.023(10^6)eV$, 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.[4h, 11] 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.[9]

3.3.3 Attenuation of Beta Particles (Electrons)

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 3.3.2). 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.[4j] 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.[4j] The typical energy loss per ion-pair formation by beta particles is about the same as that for alpha particles, 35eV.[4e] 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. Thus, the use of polyvinylchloride in uranium storage may be beneficial in reducing the background level of electron radiation, but the use of polyvinylchloride as a structural material could be limited if the combination of electron flux and exposure time were sufficient to cause mechanical degradation.

3.3.4 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 so-called alpha-neutron (α-n) reactions, in which alpha particles interact with light isotopes such as ^{9}Be, ^{6}Li, ^{7}Li, ^{9}Be, ^{10}B, ^{11}B, ^{19}F, ^{27}Al, and ^{28}Si.[12,13] 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 polymers such as Polytetrafluoro-ethylene (commonly called Teflon) were used as a containment material.

Neutron attenuation is qualitatively different from α, β, or γ radiation because neutrons interact almost exclusively with nuclei rather than with electrons.[4f] 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).[4f] 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.[4f]

3.4 Effects of Ionizing Radiation on Polymers

A review of Section 3.3. with specific reference to storing the naturally occurring uranium isotopes in polyethylene and polyvinylchloride indicates the following:

1. Alpha particles cause significant damage to plastics, but only a small portion of the alpha activity generated within a sample of uranium reaches the uranium-plastic interface. Most of the alpha flux is attenuated by self-shielding. Because the distance travelled by alpha

radiation through hydrogenous media is short (see Section 3.3.1), the effect of the radiation damage on polymers is expected to be superficial, perhaps eventually resulting in the spalling-off of a thin inner-surface layer.

2. Sources of electrons associated with radioactive decay of the naturally occurring uranium isotopes includes: (1) beta decay of the short-lived daughter products ^{234}Th, ^{234}Pa, and ^{231}Pa; (2) electron emission that occurs in conjunction with alpha decay; and (3) electron release due to photoelectric absorption and Compton scattering of gamma radiation. As noted in Section 3.3.3, electrons are expected to interact significantly with a polymer medium; a significant portion of the electrons generated within the bulk material will be expected to reach the uranium-polymer interface. Depending on the thickness of the polymer layer, many or most of the electrons will be attenuated by the polymer. The immediate consequence of electron attenuation will typically involve ion-pair formation.

3. Most of the gamma radiation associated with radioactive decay will pass through a polymer layer without interacting significantly with the polymer medium.

4. Neutrons interact efficiently with plastics, causing significant structural damage at high exposure levels. However, neutrons are not liberated as a direct result of radioactive decay of the uranium isotopes or decay products. Significant neutron activity is to be expected only if the uranium medium contains a significant concentration of a nucleus that shows significant (α,n) activity (see Section 3.3.4). Therefore, neutron damage is not considered to be a major concern for the storage of uranium metal or uranium oxides.

Two specific types of reaction that occur in polymers as a result of irradiation-induced ionization are polymer scission and cross-linking.[4g] These are considered below.

3.4.1 Polymer Scission

Scission occurs when chemical bonds in the polymer backbone (C-C bonds) are broken by irradiation. When scission is the primary consequence of irradiation, the molecular weight decreases as dose increases.[4g] This reduces the polymer chain length and causes a general weakening of the bulk material.

3.4.2 Polymer Cross-Linking

The mechanical properties of polymers can be modified by cross-linking. This is the process of forming direct carbon-to-carbon bonds between polymer chains.[4g, 14] Cross-linking enhances the hardness and effective melting point of polymers. It can be achieved by several means including chemical agents such as sulfur, heat treatment, gamma irradiation, ultraviolet irradiation, exposure to high-energy electrons, and ion implantation. From the preceding discussion, the principal cause of ionization-induced cross-linking for polymers used in uranium storage will be electron irradiation. In the case of polyethylene, two C-H bonds are broken and a C-C bond is formed between polymer chains.

3.4.3 Net Effects of Irradiation on Polymer Properties

The overall effect of irradiation on the mechanical properties of polymers used in packaging depends on whether scission or cross-linking is the dominant process. For polyethylene, cross-linking predominates;[4g] either cross-linking or scission may dominate in polyvinyl chloride, depending upon specific conditions of irradiation.[4g] In terms of mechanical properties, cross-linking enhances the mechanical strength of polymers (except in the case of extremely high-flux irradiation), whereas scission reduces the mechanical strength.

3.4.4 Container Pressurization Due to Polymer Degradation

Both scission and cross-linking of polyolefins involve the evolution of gases. Hydrogen is the main gaseous product of the degradation of polyethylene while hydrogen chloride is the main product of polyvinylchloride degradation (although some chlorine and hydrogen is also formed). Gaseous evolution due to polymer irradiation poses a safety hazard if the accumulated pressure of gas evolved is sufficient to rupture the container. When the material being stored is uranium metal, both hydrogen and hydrogen chloride gases are expected to be gettered through UCl_x and UH_3 formation. For oxide storage, some gettering of HCl may occur, but hydrogen will not be gettered.

3.5 Neutron Moderation

As noted in Section 3.3.4, plastics used in the containment of EU metal or oxide will not be subjected to significant neutron exposure under approved storage conditions. However, the subject of neutron moderation is a serious consideration in terms of criticality safety because both polyethylene and polyvinylchloride have high net hydrogen concentrations. This is a serious consideration in planning a storage strategy for fissionable uranium because the hydrogen nucleus is a powerful neutron moderator and reflector. Thus, the presence of either polymer must be included in a determination of the criticality potential of a proposed storage configuration.

3.6 Sample Calculations: Ionizing Radiation Emitted from Enriched Uranium

In this section the flux of ionizing radiation (alpha, beta, gamma, electrons accompanying alpha decay, and neutrons) is estimated for a polyethylene container containing EU. From this, the number of ion pairs that would be formed per year is determined. The purpose is to decide which, if any, of the types of radiation emanating from EU should be considered to limit the time span over which the plastic container should be expected to maintain its physical integrity.

To simplify calculations, the distance travelled by alpha particles and electrons (including beta particles) in the sample medium were taken to be $5(10^{-4})$ cm and 0.5 cm, respectively. These are typical values for cases where water is the medium, as noted in Sections 3.3.1 and 3.3.3. To account for self-absorption of the particles by the emitting media, the net flux was taken to be one-fourth of the net activity of a volume element having a 1 × 1-cm surface and a thickness equal to the accepted travel distance. This assumption is justified as follows:

If the particles emitted from a point source all travel a distance r prior to attenuation and they are emitted in random directions, the envelope of their termination sites will form a sphere of radius r. If the point source is located a distance x (x < r) from a planar boundary, the fraction of particles from the point source passing through the planar boundary is the area of the sphere across the plane from the point source divided by the surface area of the entire sphere:

$$\text{Fraction F1} = (4\pi r^2)^{-1} \int_{\Theta=0}^{\alpha} 2\pi r^2 \sin\Theta \, d\Theta,$$

Where $\cos \alpha = x/r$. Evaluation gives the fraction $F1 = \frac{1}{2}(1-\cos\alpha)$.

For a continuum of point sources (such as a uranium body) having a planar surface, the fraction of particle emissions passing through the plane is given by:

$$\text{Fraction F2} = \frac{\int_{x=0}^{r} \frac{1}{2}(1-\cos\Theta)\, dx}{\int_{x=0}^{r} dx}, \text{ or } 1/r \int_{x=0}^{r} (1 - x/r)dx.$$

An evaluation yields one-fourth. Thus one-fourth of the particles emitted within a volume element having a thickness equal to the travel distance are expected to cross the planar boundary.

Alpha Activity

The data in Tables 4 and 5 show that the primary source of ionizing radiation from EU is the result of ^{234}U alpha decay. Table 5 gives the specific activity of EU as $2.384(10^6)$ counts per g second. Thus, the net alpha flux actually reaching the surface of a polyethylene container is estimated by assuming that one-fourth the alpha particles in a volume element having a thickness of $5(10^{-4})$ cm will impinge on the surface. For a 1 × 1-cm surface element, the corresponding volume element is $5(10^{-4})$ cm^3; because the density of uranium metal is about 19 g/cm^3, the volume element has a mass of $5(10^{-4})cm^3$, $19g/cm^3 = 9.5(10^{-3})$ g. Then the alpha activity impinging on the plastic surface is:

$$\text{Activity} = \tfrac{1}{4} \cdot 9.5(10^{-3}) \cdot 2.384(10^6) = 5.662(10^3)\ \alpha\ cm^{-2}s^{-1}.$$

From Reference 9, the alpha emission from ^{234}U is about 4.7 MeV = $4.7(10^6)$eV. Then the net energy associated with the alpha decay in the volume element is:

$$\text{Energy} = 5.662(10^3)\ \alpha\ cm^{-2}s^{-1} \cdot 4.7(10^6)eV/\alpha = 2.661(10^{10})eV\ cm^{-2}\ s^{-1}.$$

As noted in Section 3.3.1, the attenuation of alpha particles causes numerous ion pairs to be formed with 35 eV typically being expended per ion pair produced. Then the number of ion pairs formed by alpha activity is:

Number of ion pairs = $2.661(10^{10})$eV cm^{-2} s^{-1} /(35 eV/ion pair)
= $7.60(10^{8})$ ion pair/cm^2 second.

This corresponds to $2.40(10^{16})$ ion pairs per year per cm^2.

Electron Emissions Accompanying Alpha Decay

Values for the energies and probability per alpha decay of electrons emitted during alpha decay were taken from Reference 9. The values and summations are as follows:

Probability	Energy (MeV)	Prob. × E (MeV)
0.0969	0.00948	0.000917
0.2006	0.03273	0.006565
0.0546	0.04802	0.002622
0.02018	0.05187	0.001047
0.00139	0.1004	0.000140
Total		0.01129 MeV

Therefore, the energy associated with the secondary electrons emitted during alpha decay is:

$2.384(10^{6})$ decay $g^{-1}s^{-1}$ ·0.01129 MeV/decay = $2.692(10^{4})$ MeV g^{-1} s^{-1}.

The volume element considered as a source of electron irradiation impinging on the wall of a plastic container is 0.5 × 1 × 1 cm = 0.5 cm^3. Again using the density of uranium as 19 g/cm^3, the mass of the volume element is 0.5·¼·19 = 2.375 g. Multiplying this by the energy of secondary elections calculated above gives:

2.375 g · $2.692(10^{4})$MeV g^{-1} s^{-1} = $6.39(10^{4})$MeV cm^{-2} s^{-1}

or $6.39(10^{10})$ eV cm^{-2} s^{-1}; the potential rate at which ion pairs are formed is:

$6.39(10^{10})$ eV cm^{-2} s^{-1} /(35 eV/ion pair) = $1.83(10^{9})$ ion pair per cm^{-2} per s^{-1}.

This is the equivalent of $5.76(10^{16})$ ion pairs per cm^{-2} per $year^{-1}$. However, a significant portion of the electrons are expected to pass through a thin plastic wall without being attenuated. If the plastic wall thickness is taken to be 0.1 cm and the typical range of electrons in polyethylene is estimated to be 0.5 cm (the value for water), only about 0.1/0.5 or 20 percent of the electrons would initiate ion pair formation. This corresponds to $1.152(10^{16})$ ion pairs per cm^{-2} per $year^{-1}$, which is slightly less than the value calculated for alpha particles.

Beta Activity

From Tables 4 and 5, the primary source of beta activity arising from EU is the ^{231}Th decay process, which yields ^{231}Pa and a beta particle. As in the prior section, the electron flux impinging on a 1-cm^2 plastic surface is estimated as the emission from 2.375 g of EU.

Nine betas of defined energy profiles are emitted, as follows[9]:

Probability	Average energy (MeV)	Prob. × E(MeV)
0.0270	0.0374	0.00101
0.0032	0.0455	0.00014
0.1500	0.0554	0.00831
0.0125	0.0581	0.00073
0.0900	0.0793	0.00713
0.4100	0.0796	0.03264
0.3500	0.0848	0.02968
0.0041	0.0868	0.00036
0.0012	0.0334	0.00003
Total		0.08003

Taking the ^{235}U concentration in EU as 93 percent and the specific activity of ^{235}U to be $7.994(10^4)\beta\ g^{-1}s^{-1}$ (from Table 4), the beta energy impinging on a cm^2 of plastic is:

$7.994(10^4)\beta\ g^{-1}s^{-1} \cdot 2.375\ g(U) \cdot .93\ g^{235}U/gU \cdot .08003\ MeV/\beta = 1.413(10^4)\ Mev\ s^{-1}\ cm^{-2}$.

This corresponds to a potential for forming $4.037(10^8)$ ion pairs $cm^{-2}\ s^{-1}$, or $1.273(10^{16})$ ion pairs per cm^{-2} per $year^{-1}$. Again, assuming that 80 percent of the beta particles are not attenuated by a 0.1-cm-thick layer (as in the previous section), ion pairs are formed in the polyethylene layer at the rate of $2.546(10^{15})$ ion pairs per cm^{-2} per $year^{-1}$.

Gamma Activity

The alpha decays of ^{234}U are accompanied by the release of gamma activity at three energy levels[9]:

Energy (MeV)	Probability per Decay
0.0130	0.105
0.0532	0.0012
0.1214	0.0004

While these radiation levels have some significance in terms of health physics considerations, gamma emissions contribute only slightly to degradation of hydrocarbon polymers because very little gamma attenuation occurs in media containing only atoms of low atomic number, as discussed in Sections 3.3.1 and 3.3.2.

Neutron Activity

Neutron emissions are not a major concern for the storage of EU in plastics because the natural isotopic decay schemes do not involve neutron release. The only time the neutron flux might become significant would be in cases where a significant concentration of alpha-neutron (α,n) active nuclei are in contact with the EU, as noted in Section 3.3.3.

Summary

The following values were calculated for the number of ion pairs formed in one year in a 1 cm^2 section of polyethylene having a thickness of 0.1-cm:

Alpha particles: $2.40(10^{16})$ ion pairs per year

Electrons from alpha decay: $1.152(10^{16})$ ion pairs per year

Beta particles: $2.546(10^{15})$ ion pairs per year

Gamma rays: much lower because hydrocarbons are relatively transparent to gamma radiation.

Neutrons: not a consideration unless the stored material contains a high concentration of a nucleus that undergoes an α,n interaction.

The most significant point to be noted from the calculations is that the numerical estimates of the ion pair formation concentration from all types of ionizing radiation emanating from HEU is about six orders of magnitude lower than the number of photon activations of sufficient energy to initiate photochemical degradation when the plastic is in direct sunlight.

The comparison of ion pair formation due to ionizing radiation with photochemical degradation through free radical initiation is certainly not a one-to-one correlation, but the vast disparity between the estimated values argues convincingly that radiation damage from EU is of much less consequence in causing deterioration of polyethylene than photochemical degradation.

4. Y-12 Policy on Storage of Uranium in Plastics

Recommendations:

Plastic containers should not be used as the primary container for the long-term (PLMS) storage of EU. The term *primary container* is taken to mean the vessel that is intended to maintain containment of the material under both normal storage conditions and under possible upset conditions.

Polyethylene bottles are acceptable primary containers for interim storage of HEU. Two conditions must be met: (1) the residence time in interim storage must be limited to ten years, and (2) the materials must be stored indoors, protected from high-intensity ultraviolet light sources such as sunlight.

The use of sealed polyvinylchloride bags as a containment material for compounds of HEU in interim storage is an acceptable practice. Use of sealed polyvinylchloride bags inside stainless steel storage cans for the long-term storage of HEU oxide (U_3O_8) as a means of reducing the likelihood of supplementary containment and prevention of contamination at the time the cans are opened is acceptable but not mandated. Use of sealed polyvinylchloride bags inside stainless steel cans for long-term storage of HEU metal would not cause a significant problem, but it appears superfluous and thus is not recommended. Any plastics used in the storage, transfer, or processing of EU must be approved explicitly by a Criticality Safety Approval.

The use of polyethylene containment for the long-term storage of depleted uranium metal billets is encouraged but not mandatory. Use of polyethylene as an alternative to aluminum or steel will reduce corrosion rates of the material because the potential for galvanic corrosion is eliminated. If the material is stored outdoors, the plastic should be shielded from direct sunlight.

Any uranium material that contains significant concentrations of the isotopes ^{232}U, ^{233}U, ^{236}U or of transuranics, decay products, or fission products must be evaluated as a potential cause of radiation damage prior to being approved for storage in any plastic. The definition of significant concentration will be consistent with the Y-12 uranium acceptance criteria.[8] Until the latter has been issued, the evaluation will be based on the recommendations in the Egli Report.[7]

Although polyethylene is fairly robust in terms of its resistance to chemical degradation, it is expected that long-term contact with uranyl nitrate crystals, uranyl nitrate solutions, or other oxidizing media will increase the rate at which the material undergoes mechanical degradation. Therefore, uranyl nitrate crystals, uranyl nitrate solutions, and other uranium-containing oxidizing media should be converted to an alternate chemical form such as metal or oxide rather than being held in interim storage in polyethylene containers for extended time periods (i.e., more than one year).

The use of polyethylene safe bottles for the interim storage of uranium solutions in both aqueous and organic media is an approved practice. The potential for pressurization within the safe bottles must be avoided by either using vented caps or conducting inspections on a regular basis to ensure the caps are not tightly sealed to the safe bottles. The safe bottles should be inspected routinely for mechanical deterioration to avoid the possibility of spills.

The use of polyethylene bottles for collection of samples is an approved practice. The sampling procedure, including sample size and bottle capacity, must be covered by a Criticality Safety Approval.

Plastics other than polyethylene or polyvinylchloride may be used in the storage of uranium if they can be shown to be equivalent or superior in terms of all relevant properties.

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