

RECEIVED

NOV 21 1995

OSTI

**National Low-Level Waste  
Management Program  
Radionuclide Report Series**

**Volume 14: Americium-241**

***National Low-Level Waste  
Management Program***

***September 1995***

**National Low-Level Waste Management  
Program Radionuclide Report Series**

**Volume 14: Americium-241**

**M. R. Winberg  
R. S. Garcia**

**Published September 1995**

**Idaho National Engineering Laboratory  
Lockheed Idaho Technologies Company  
Idaho Falls, Idaho 83415**

**Prepared for the  
U.S. Department of Energy  
Assistant Secretary for Environmental Management  
Under DOE Idaho Operations Office  
Contract DE-AC07-94ID13223**

**DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED**

**MASTER**

## **DISCLAIMER**

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

## **ABSTRACT**

This report, Volume 14 of the National Low-Level Waste Management Program Radionuclide Report Series, discusses the radiological and chemical characteristics of americium-241 ( $^{241}\text{Am}$ ). This report also includes discussions about waste types and forms in which  $^{241}\text{Am}$  can be found and  $^{241}\text{Am}$  behavior in the environment and in the human body.

## **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.

---



## FOREWORD

The purpose of the National Low-Level Waste Management Program Radionuclide Report Series is to provide information to state representatives and developers of low-level radioactive waste disposal facilities about the radiological, chemical, and physical characteristics of selected radionuclides and their behavior in the low-level radioactive waste disposal facility environment. Extensive surveys of available literature provided information used to produce this series of reports and an introductory report.

The National Low-Level Waste Management Program Radionuclide Report Series previously addressed the radionuclides technetium-99, carbon-14, iodine-129, tritium, cesium-137, strontium-90, nickel-59, plutonium-241, nickel-63, niobium-94, cobalt-60, and curium-242. These radionuclides contribute significantly to the dose estimated during a performance assessment analysis.

This report is Volume 14 of the series. It outlines the basic radiological, chemical, and physical characteristics of  $^{241}\text{Am}$ , waste types and forms that contain this radionuclide, and its behavior in environmental media such as soils, plants, water, air, animals, and the human body. Additional reports will be generated for other radionuclides.



## CONTENTS

ABSTRACT .....	iii
FOREWORD .....	v
INTRODUCTION .....	14-1
RADIOLOGICAL CHARACTERISTICS .....	14-2
CHEMICAL AND PHYSICAL CHARACTERISTICS .....	14-5
AMERICIUM-241 PRODUCTION .....	14-6
WASTE TYPES AND FORMS THAT CONTAIN AMERICIUM-241 .....	14-7
Nuclear Reactors .....	14-7
Government Waste Types and Forms .....	14-7
Medical, Academic Institutions, and Industrial Waste Types and Forms .....	14-7
BEHAVIOR OF AMERICIUM-241 IN THE ENVIRONMENT .....	14-9
Americium in Soils .....	14-9
Americium-241 in Water .....	14-10
Americium in Plants .....	14-10
Americium in Air .....	14-12
BEHAVIOR OF AMERICIUM-241 IN ANIMALS AND HUMANS .....	14-13
CONCLUSIONS .....	14-16
REFERENCES .....	14-18
BIBLIOGRAPHY .....	14-20

## FIGURES

1. Alpha particle ranges in air as a function of energy .....	14-2
2. Diagram for the primary decay sequence for $^{241}\text{Am}$ .....	14-4
3. In-growth of $^{241}\text{Am}$ activity from 1 g of $^{241}\text{Pu}$ over a 50-year period .....	14-6
4. $^{241}\text{Am}$ activity disposed of at commercial low-level waste disposal sites 1986-1994 .....	14-8



## TABLES

1. Thickness of lead required to reduce useful beam to 5% .....	14-3
2. Comparison of the radiotoxicity of several important radionuclides. <sup>4</sup> .....	14-4
3. Physical properties of americium and its compounds .....	14-5
4. <sup>241</sup> Am in bush beans for 96 hours to three different concentrations of <sup>241</sup> Am in solution culture .....	14-11
5. <sup>241</sup> Am in roots, stems, and leaves of plants grown in contaminated soil in a glasshouse .....	14-12
6. Committed dose equivalents per unit uptake via ingestion and inhalation .....	14-13
7. Annual limits on intake and derived air concentrations for <sup>241</sup> Am .....	14-15

# National Low-Level Waste Management Program Radionuclide Report Series

## Volume 14: Americium-241

### INTRODUCTION

This report outlines the basic radiological and chemical characteristics of americium-241 ( $^{241}\text{Am}$ ) and examines how these characteristics affect the behavior of  $^{241}\text{Am}$  in various environmental media, such as soils, water, plants, air, animals, and the human body. Discussions also include methods of  $^{241}\text{Am}$  production, waste types, and waste forms that contain  $^{241}\text{Am}$ .

All americium atoms contain 95 protons ( $Z = 95$ ) and various numbers of neutrons (typically 142 to 151 neutrons) within the nucleus. Americium is classified as one of the transuranic elements. Americium is not a normal constituent of the natural environment and is generated as a result of human activities. There are no stable isotopes of americium. The radioactive isotopes of americium have half-lives ranging from 0.9 minutes ( $^{232}\text{Am}$ ) to 7,370 years ( $^{243}\text{Am}$ ). The most common isotope and subject of this report is  $^{241}\text{Am}$  with a half-life of 432 years.<sup>1</sup>

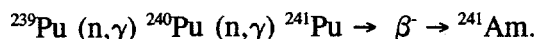
The regulatory limits for  $^{241}\text{Am}$  disposal and personnel exposure are very restrictive. Relatively small concentrations of  $^{241}\text{Am}$  can become a major contributor to the calculated doses to individuals in performance assessment calculations or an accidental release involving this radionuclide. Therefore, knowledge of the basic radiological and chemical characteristics, waste forms, and environmental behavior of  $^{241}\text{Am}$  is important.

The primary source of  $^{241}\text{Am}$  in the environment has been related to the manufacturing of atomic weapons. There is also commercial use of  $^{241}\text{Am}$  in radioactive sealed sources (e.g., smoke detectors). This nuclide is produced in nuclear reactors.  $^{241}\text{Am}$  enters the environment from the activities related with reactor operations and decommissioning, atomic weapons production, and when sealed sources containing  $^{241}\text{Am}$  are manufactured, used, and disposed.

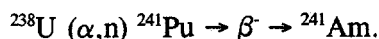
Information for this report was taken from standard reference materials and specific studies related to  $^{241}\text{Am}$ .

## RADIOLOGICAL CHARACTERISTICS

$^{241}\text{Am}$  was discovered in 1945 by American scientists G. T. Seaborg, R. A. James, and L.O. Morgan, who named it after the Americas.<sup>2</sup> The most recent information available concerning  $^{241}\text{Am}$  reports a half-life of 432.2 years. Reference 2 also states that the principle means of production of  $^{241}\text{Am}$  is by the bombardment of plutonium atoms in a nuclear reactor in the following reaction:



$^{241}\text{Am}$  can also be produced as a result of the bombardment of  $^{238}\text{U}$  with 40-MeV (million electron volt) helium ions in the following reaction:



Reference 1 lists the various energies and types of decay emissions from  $^{241}\text{Am}$ .  $^{241}\text{Am}$  undergoes radioactive decay via alpha particle emission from the nucleus to the radionuclide neptunium-237 ( $^{237}\text{Np}$ ). This decay results in a maximum alpha particle energy of 5.54 MeV (0.3% probability). The greatest intensity alpha energy in the decay process is 5.48 MeV (85.2% probability) followed by 5.43 MeV (12.8% probability). Since these alpha particles are easily shielded and have a relatively short range in air, they are of little concern for external radiation exposure. Figure 1 shows the alpha particle range in air as a function of energy (note that alpha particles with energies above 4 MeV tend to travel farther in air).

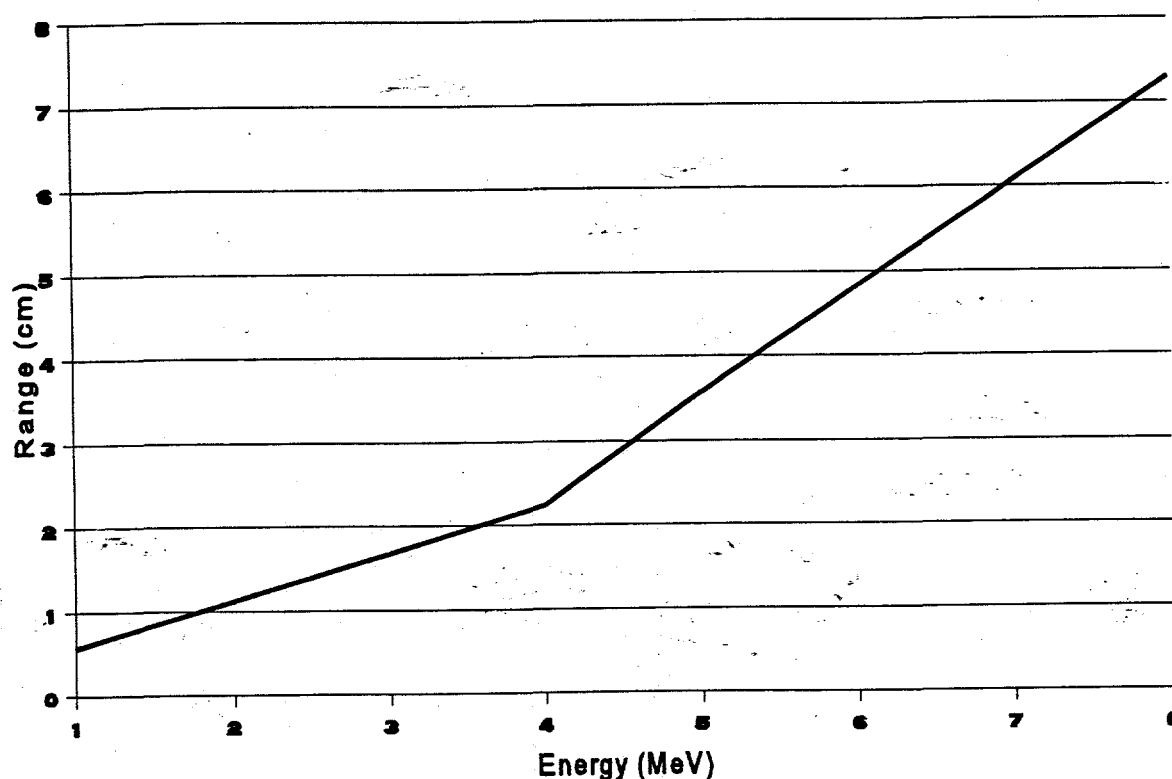


Figure 1. Alpha particle ranges in air as a function of energy.

Since the half-life of the parent nuclide plutonium-241 ( $^{241}\text{Pu}$ ) (14.4 years) is considerably less than the half-life of  $^{241}\text{Am}$ , the activity of  $^{241}\text{Am}$  will continue to grow over time unless the americium is chemically separated from the plutonium.

The decay process of  $^{241}\text{Am}$  also produces gamma-rays and x-rays. The gamma-ray and x-ray energies range from 11.9 to 955.7 keV. The most intense gamma-ray energy is 59.5 keV (35.9% probability) followed by a 17.61 keV (20.2% probability) x-ray. Nearly 77% of all of the photons emitted during the decay of  $^{241}\text{Am}$  occur in the range of 11.9 to 69.8 keV. The specific activity of  $^{241}\text{Am}$  is  $1.27 \times 10^{11}$  Bq/g (3.44 Ci/g).<sup>1</sup> Table 1 shows the thickness of lead shielding requirements as a function of energy to reduce a useful beam of gamma-rays or x-rays to 5%.

The primary decay sequence for  $^{241}\text{Am}$  showing the emissions of the alpha particle, x-rays, gamma-rays, and their respective decay probabilities is shown in Figure 2.

$^{241}\text{Am}$  also undergoes a spontaneous fission with a half-life of  $2 \times 10^{14}$  years. The calculated neutron (n) emission rate is 0.6 n/sec/g.<sup>3</sup> This makes  $^{241}\text{Am}$  a suitable source for use in neutron sources in combination with a multiplication material such as beryllium (Be). A typical AmBe source yields  $2.2 \times 10^6$  n/s per Ci of radioactive material.

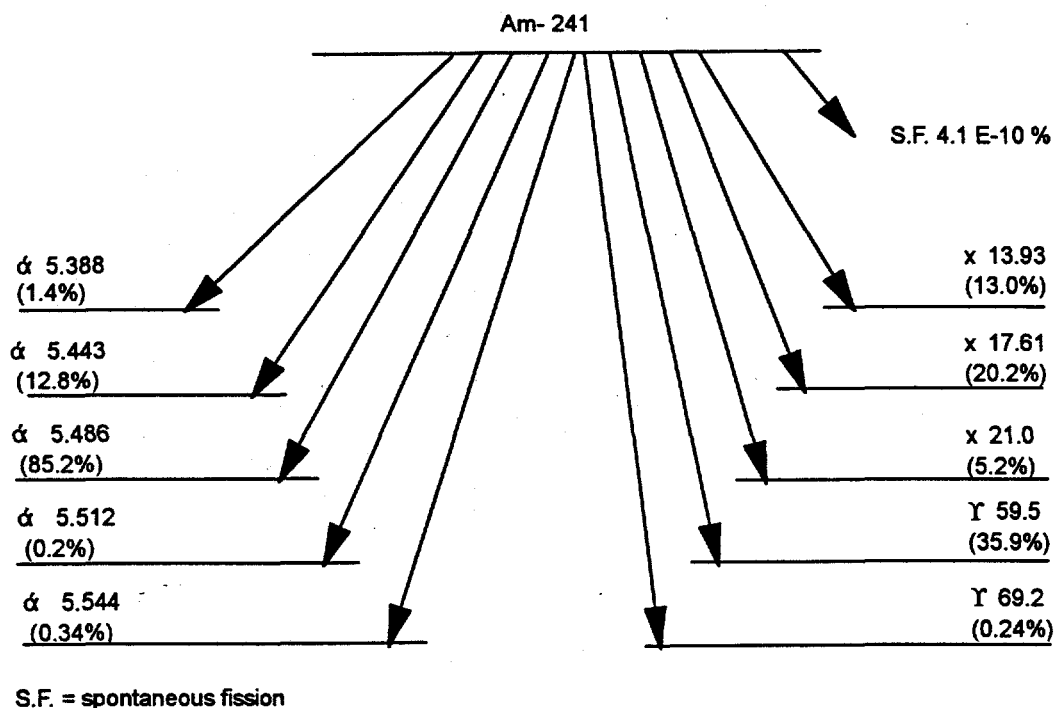
Table 2 lists the radiotoxicity of several important radionuclides. Notice that  $^{241}\text{Am}$  and its decay product  $^{237}\text{Np}$  are listed with the very high radiotoxicity radionuclides.

The very high radiotoxicity of  $^{241}\text{Am}$  is due to the long half-life and high energies of the alpha particles emitted during the decay process. Because of the limited range of alpha particles, they can be easily shielded from external exposure. A sheet of paper or dead layer of skin will attenuate most alpha particles. The danger from alpha particles comes from possible internal exposure due to inhalation, ingestion, or injection where the alpha particles can come in contact with living tissue. Alpha particles can cause extreme damage when they interact with living tissue. The biological effects of  $^{241}\text{Am}$  will be discussed in more detail in a later section of this report.

**Table 1.** Thickness of lead required to reduce useful beam to 5%.<sup>3</sup>

	Energy of source	Half-value layer <sup>a</sup> (cm)	Required lead (cm)
X-rays/gamma-rays	50 keV	0.005	0.02
	70 keV	0.010	0.04
	100 keV	0.025	0.11

a. The half-value layer is defined as the thickness of shielding material (in this case lead) required to reduce the beam of x-rays or gamma-rays by one-half.



Np-237

**Figure 2.** Diagram for the primary decay sequence for  $^{241}\text{Am}$  (alpha particle energies are in million electron volts, and x-ray, and gamma-ray energies are in kiloelectron volts).

**Table 2.** Comparison of the radiotoxicity of several important radionuclides.<sup>4</sup>

Radiotoxicity		Species
Very high	Group 1	$^{241}\text{Pu}$ , $^{242}\text{Cm}$ , $^{241}\text{Am}$ , $^{237}\text{Np}$
High	Group 2	$^{60}\text{Co}$ , $^{90}\text{Sr}$ , $^{94}\text{Nb}$
Moderate	Group 3	$^{14}\text{C}$ , $^{63}\text{Ni}$ , $^{137}\text{Cs}$
Low	Group 4	$^3\text{H}$ , $^{59}\text{Ni}$ , $^{99\text{m}}\text{Tc}$ $^{99}\text{Tc}$ , $^{129}\text{I}$

## CHEMICAL AND PHYSICAL CHARACTERISTICS

Americium is a member of the seventh series known as the actinide series of elements. In addition, americium is part of the transuranium group of the periodic table (all elements after uranium in the periodic table with a  $Z > 92$ ). This group includes plutonium, curium, neptunium, berkelium, californium, einsteinium, fermium, mendelevium, nobelium, and lawrencium. Like all transuranium elements, americium does not naturally occur in nature and all isotopes are radioactive. Americium can only be produced by the nuclear reactions described in the previous section of this report.<sup>5</sup>

Americium in metal form is more silvery in appearance than neptunium or plutonium metal and is malleable and ductile as compared to uranium or neptunium metal. Americium is thought to exist in two forms: an alpha form that has a double hexagonal close-packed structure and a loose-packed cubic beta form. The melting point of americium is  $994^{\circ}\text{C}$  and the boiling point is estimated at  $2,607^{\circ}\text{C}$ . The average density of americium is  $13.7\text{ g/cm}^3$  at  $20^{\circ}\text{C}$ .

Americium can be combined with several different elements to make compounds such as americium bromide ( $\text{AmBr}_3$ ), americium chloride ( $\text{AmCl}_3$ ), americium fluoride ( $\text{AmF}_3$ ), americium iodide ( $\text{AmI}_3$ ) and americium oxide ( $\text{Am}_2\text{O}_3$ ). Table 3 shows the physical properties of the different forms of americium.

**Table 3.** Physical properties of americium and its compounds.<sup>5</sup>

Name	Molecular weight	Crystal form	Density ( $\text{g/cm}^3$ )	Melting point ( $^{\circ}\text{C}$ )	Boiling point ( $^{\circ}\text{C}$ )
Am	243	Silvery, hex	13.7	$994 \pm 4$	2,607
$\text{AmBr}_3$	482.7	White, orthorhomb	—	Sublimes	—
$\text{AmCl}_3$	349.5	Pink, hex	5.8	Sublimes 850	—
$\text{AmF}_3$	300	Pink, hex	9.5	—	—
$\text{AmI}_3$	623.7	Yellow, orthorhomb	6.9	—	—
$\text{Am}_2\text{O}_3$	534	Reddish-brown, cube or tan, or hex	—	—	—

## AMERICIUM-241 PRODUCTION

As previously mentioned,  $^{241}\text{Am}$  is produced by the beta decay of  $^{241}\text{Pu}$ . Figure 3 shows the in-growth of the activity of  $^{241}\text{Am}$  from the decay of 1 g of  $^{241}\text{Pu}$  for 50 years.

Pure  $^{241}\text{Am}$  is produced by chemically separating the americium from its  $^{241}\text{Pu}$  parent.  $^{241}\text{Pu}$  is dissolved totally into a solution. After standing for a period of time, perhaps on the order of several years, the americium is then separated from the plutonium solution. The operation of recovering americium has been termed "milking" and the plutonium then is naturally designated as a "cow." As in the case of its bovine counterpart, it takes a big cow to provide even a little milk.<sup>6</sup>

The separation of americium is facilitated by the fact that  $\text{Am}^{3+}$  is more stable than the trivalent ions of plutonium, which pass into higher oxidation states in conditions under which americium remains trivalent. This allows americium to be separated from other transuranics by means of ion-exchange and extraction with organic solvents.

The only other method known to produce  $^{241}\text{Am}$  is as a result of the bombardment of  $^{238}\text{U}$  with 40-MeV helium ions. This method produces  $^{241}\text{Am}$ , but it still has to be separated chemically from its uranium and plutonium parents using the same type chemical extraction methods described above.

The major source of  $^{241}\text{Am}$  production is the manufacturing process for atomic weapons. National production reactors are designed to generate fissile material in the form of plutonium. As part of this process, some residual  $^{241}\text{Pu}$  is produced along with the preferred isotope of  $^{239}\text{Pu}$ . The target plutonium is processed to remove the unwanted plutonium isotopes until it is approximately 95%  $^{239}\text{Pu}$ . This material is commonly referred to as weapons-grade plutonium. The unwanted portion containing the excess  $^{241}\text{Pu}$  is then sold for producing  $^{241}\text{Am}$  for commercial use. The remaining 5% of the weapons-grade plutonium does contain some trace amounts of  $^{241}\text{Pu}$  plus other plutonium isotopes. Therefore, atomic weapons will contain some trace amounts of  $^{241}\text{Am}$ .

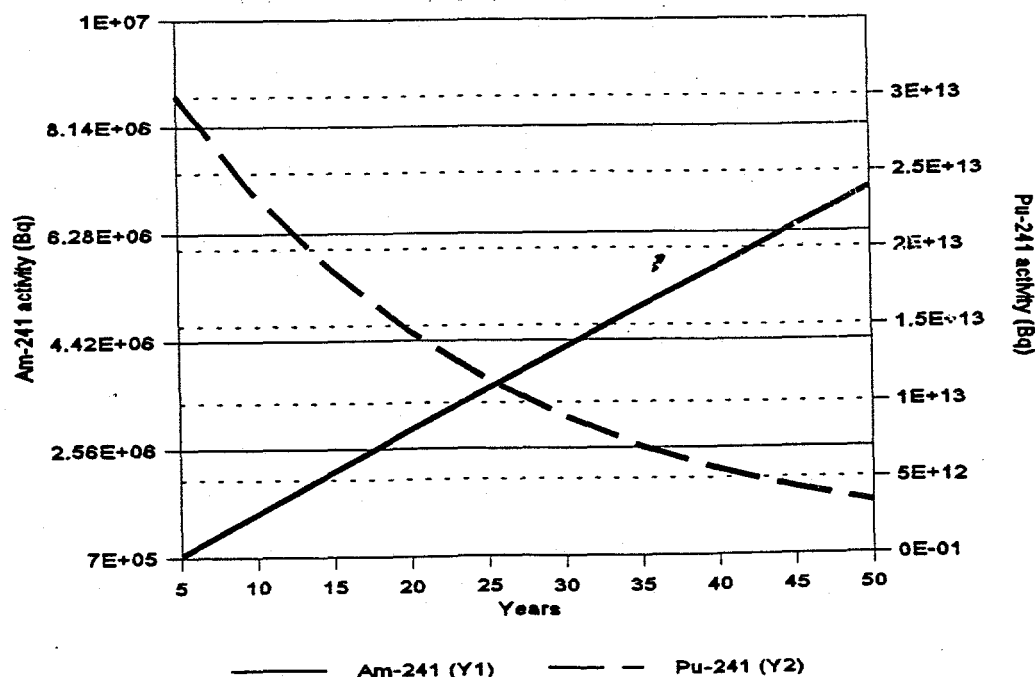


Figure 3. In-growth of  $^{241}\text{Am}$  activity from 1 g of  $^{241}\text{Pu}$  over a 50-year period.

# WASTE TYPES AND FORMS THAT CONTAIN AMERICIUM-241

## Nuclear Reactors

$^{241}\text{Am}$  is produced within the fuel and operational wastes from cleanup systems of nuclear reactors. The quantity of  $^{241}\text{Am}$  in spent fuel that is produced each year is proportional to the amount of  $^{241}\text{Pu}$  produced and continues to increase as the fuel is stored. Each year the operations of domestic light water reactors generate approximately 2,302 kg of  $^{241}\text{Pu}$  contained in the spent fuel. The cumulative quantity of  $^{241}\text{Pu}$  that existed in spent fuel as of 1991 was 19,200 kg.<sup>7</sup> All of the  $^{241}\text{Am}$  associated with spent commercial fuel is expected to stay with the fuel and be disposed of in the planned national high-level waste repository.

$^{241}\text{Am}$  associated with operational wastes from commercial reactors appears in spent radioactive resins, filter sludges, and evaporator bottom wastes.

The Code of Federal Regulations, 10 CFR 61.55, regulates the concentrations of radionuclides that are suitable for near-surface disposal. This regulation requires that wastes be classified for disposal as Class A, B, or C with Class C waste having the most rigorous disposal requirements. Wastes that exceed the Class C limits are not suitable for near-surface disposal. The Class C limit for  $^{241}\text{Am}$  is included in the category of "alpha-emitting transuranic radionuclides with half-lives greater than 5 years." This limit is 100 nCi/g of waste.<sup>8</sup>

## Government Waste Types and Forms

Government wastestreams containing  $^{241}\text{Am}$  are associated primarily with the manufacturing of atomic weapons and the decommissioning of the associated Government facilities. The majority of the waste forms containing  $^{241}\text{Am}$  are located at the U.S. Department of Energy (DOE) facilities located in the United States. These waste forms consist of drums and boxes containing small amounts of weapons-grade plutonium and associated decay products. The waste consists of contaminated personal protective equipment, tools, plastic, plant equipment, salts, solvents, and sludges. These wastes are referred to in the DOE system as transuranic waste. The total volume of transuranic waste currently stored at DOE facilities is 104,116 m<sup>3</sup> (Reference 7). These wastes are designated to be disposed of at the DOE Waste Isolation Pilot Plant (WIPP) in New Mexico when that facility is allowed to open.

## Medical, Academic Institutions, and Industrial Waste Types and Forms

There are no known medical uses and resultant wastestreams for  $^{241}\text{Am}$ . Academic wastestreams for  $^{241}\text{Am}$  consist primarily of sealed sources, liquids and sludges, and contaminated materials from research projects and operation of university reactors.

Industrial wastestreams containing  $^{241}\text{Am}$  include sealed sources, liquids, sludges, and contaminated materials from the manufacture of sealed sources, including smoke detectors. Fuel burn up laboratories and commercial research facilities with small reactors can also generate liquids, sludges, and contaminated materials that may contain  $^{241}\text{Am}$ .

Volumes and activities from these wastestreams are relatively small and are disposed of at commercial low-level waste disposal facilities.



Only a small amount of  $^{241}\text{Am}$  activity from nuclear reactors and other commercial generators is disposed of as low-level waste at commercial disposal sites each year as illustrated in Figure 4.<sup>a</sup>

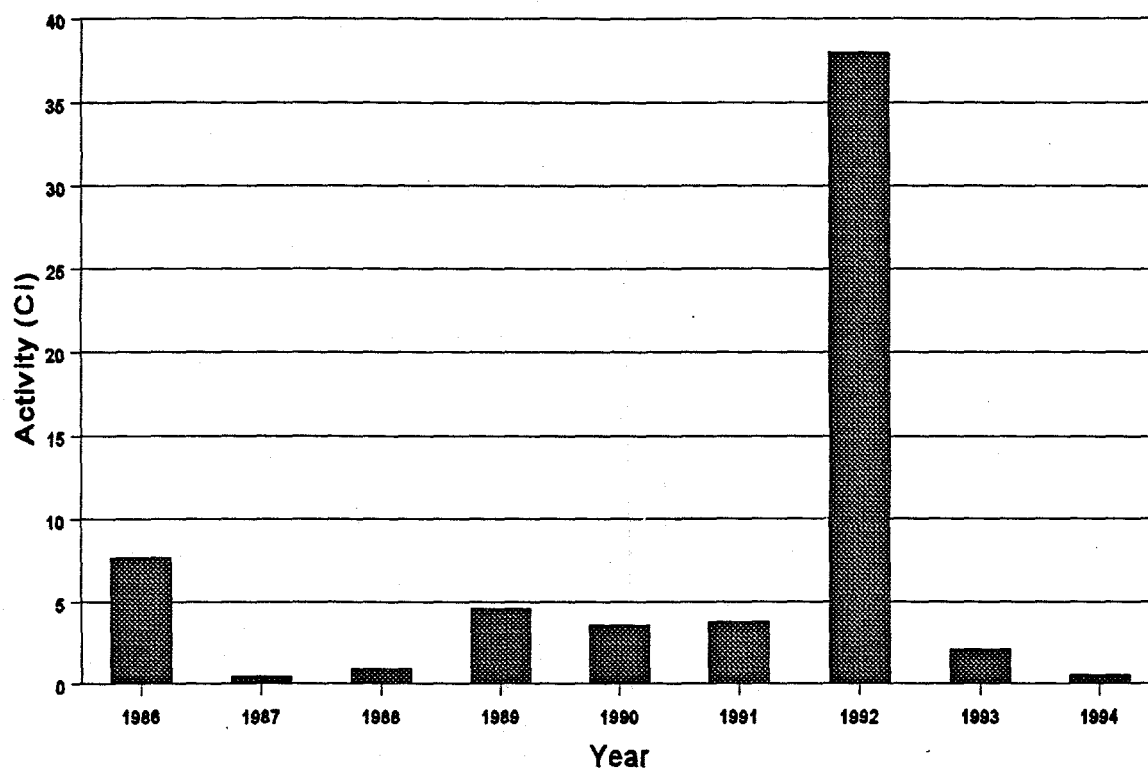


Figure 4.  $^{241}\text{Am}$  activity disposed of at commercial low-level waste disposal sites 1986–1994.

a. Data taken from the Manifest Information Management System data base maintained by the National Low-Level Waste Management Program, December 1994.

# BEHAVIOR OF AMERICIUM-241 IN THE ENVIRONMENT

## Americium in Soils

Understanding the behavior of radioactive materials such as  $^{241}\text{Am}$  is important in assessing the possibility of transport of this radionuclide to the biosphere. It is also important to understand the interactions between radionuclides and various media along the path to the biosphere, whether disposal is in deep or shallow geologic formations or in near-surface disposal facilities.

Four parameters were reported to be essential in accurately predicting soil concentrations from either contaminated groundwater or irrigation water. The four parameters, in order of importance, are

1. Soil retention
2. Annual precipitation
3. Soil texture
4. Depth to the water table.

The soil retention parameter represents the solid/liquid partition coefficient and is denoted by the symbol  $K_d$ .  $K_d$  is defined by  $K_d = C_s/C_l$ , where  $C_s$  is the americium concentration in the soil ( $\mu\text{g Am/g}$  of soil), and  $C_l$  is the americium concentration in the groundwater ( $\mu\text{g Am/mL}$  of liquid). Therefore, the units of  $K_d$  are  $\text{mL/g}$  (i.e.,  $\text{mL}$  of water per gram of soil). This empirical model combines all soil retention mechanisms into a simple linear partition relationship between the soil and the surrounding groundwater. This model assumes the americium concentrations in the groundwater and soil are in equilibrium. Using this definition for  $K_d$ , it follows that the larger the value, the slower the migration of the radionuclide relative to the groundwater flow. Therefore,  $K_d$  can be thought of as a measure of the amount of "fixing" or holdup in the soil.

Typically,  $K_d$  is measured under laboratory conditions with samples that are relatively homogenous and where equilibrium conditions can be met. Applying these values to specific soils in the field can be difficult because the actual soils are generally nonhomogeneous and there are uncertainties as to how long it takes americium concentrations in the soil and groundwater ( $C_s$  and  $C_l$ ) to reach equilibrium with each other. Therefore, one must be careful to ensure that soil samples used in laboratory studies are as closely representative of the field as possible. Even though the soil samples do not precisely match conditions in the field,  $K_d$  values from laboratory studies can be used in computer models to extrapolate data from laboratory experiments and field studies. In addition, some experiments are conducted using intact field studies to validate laboratory  $K_d$  values and to study the effects associated with soil nonhomogeneity.

Since americium is not naturally found in soils, studies of values for  $K_d$  are focused around soil types at sites where americium contamination of soils has occurred. These sites are typically associated with Government weapons production and testing activities, such as the Hanford Site in Washington and the Savannah River Site in South Carolina.

An experimental study was performed looking at the sorption of  $^{241}\text{Am}$  on subsoils from both South Carolina and Washington. This study examined the differences in  $K_d$  values as a function of calcium and sodium concentrations in the soil. For the South Carolina site,  $K_d$  values ranged from

1.0 to 67 for calcium concentrations and 1.6 to 280 for sodium concentrations.<sup>9</sup> This study indicated that for the South Carolina soils, the  $K_d$  values tended to increase as the calcium and sodium concentrations in the soil increased. The  $K_d$  values for Washington soils were  $> 1,200$  and were independent of the calcium or sodium concentrations.

Another study looked at  $K_d$  values from seven different soil types and pH.<sup>10</sup> This study determined the effect of soil type and pH on the extractability of  $^{241}\text{Am}$ . It was determined that the extractability of  $^{241}\text{Am}$  was high between pH 1.2 and 2 and then decreased to a slower rate to lower levels with increasing pH.  $K_d$  values for  $^{241}\text{Am}$  ranged from 182 to 47,230 depending on the soil type at pH ranges from 4.4 to 8.0. This study suggested that soils with strong humic or fulvic acid concentrations would tend to allow  $^{241}\text{Am}$  to migrate more freely than soils with higher pH contents.

Rockwell International conducted a study to evaluate radionuclide transport in relationship to the Columbia River Basalt Geohydrologic Basalt System.<sup>11</sup> This study was trying to determine if the basalt formations would be suitable for a geologic repository. The study looked at the Umtanum basalt and Mabton interbed formations under oxidizing and reducing conditions.  $K_d$  values for  $^{241}\text{Am}$  ranged from 340 for the basalt formations to  $> 10,000$  for interbed formations. These  $K_d$  values are similar to those found in other studies.

In summary, studies have shown that the behavior of  $^{241}\text{Am}$  in soils is dependent on soil type and pH. The studies agree that for most soil types (except those with low calcium or sodium concentrations)  $^{241}\text{Am}$  is not mobile in soils and that only small quantities can be expected to migrate within the soil to the biosphere.

## Americium-241 in Water

Americium does not naturally occur in the environment, including ground or surface waters. However, as a result of past atmospheric nuclear weapons testing, some nuclides of americium do exist in small concentrations on land, and in the oceans, and may be detectable in some groundwater systems due to radioactive fallout.

Americium, as is true for all actinides, typically does not move more than a few meters in the subsurface.<sup>12</sup> The high molecular weight fraction of americium (about half) attaches itself to colloids and is moved with groundwater. Mobile americium becomes tightly and irreversibly associated with colloidal material between 25-450 nanometer (nm) in size. Once the americium is bound to these colloids, it is removed only gradually by the groundwater. These colloids can, however, move slowly through a groundwater system. When attached to these colloids, americium typically does not migrate more than a few tens of meters.  $K_d$ s for americium in groundwater can be as high as 10,000, meaning the americium will move about one ten-thousandth as fast as the groundwater.

Based on the above information,  $^{241}\text{Am}$  generally migrates only short distances (meters) within groundwater and tends to bind with colloidal mixtures and clays found within the ground.

## Americium in Plants

The main route of uptake for plants is through the roots. The area of collection of the radionuclide in the plant may vary with each specie, though most seem to collect americium in the root with only a small amount going into the shoot (see Table 4). This table also illustrates that diethylenetriaminepentaacetate (DTPA) enhances the  $^{241}\text{Am}$  uptake to areas of the plant included in the study.

**Table 4.**  $^{241}\text{Am}$  in bush beans for 96 hours to three different concentrations of  $^{241}\text{Am}$  in solution culture.<sup>13</sup>

Treatment		Concentration ratios			
$^{241}\text{Am}$ , ( $\mu\text{Ci/mL}$ )	DTPA ( $10\text{E}-4 \text{ M}^a$ )	Leaves	Stems	Roots	Whole plant
1.0	-	0.005	0.011	24.8	2.63
1.0	+	3.39	0.9	11.2	3.36
0.5	-	0.008	0.007	16.2	2.17
0.5	+	2.55	0.37	5.0	2.04
0.25	-	0.006	0.007	15.5	2.83
0.25	+	0.81	0.13	6.7	1.35

a. + or - indicates that the DTPA treatment solution was either above or below a  $10^{-4}$  concentration.

Concentration ratios (CR) are a common method of measuring the uptake of radionuclides by plants. The measured ratio is the activity in the plant per weight of the plant to the activity in soil or other growth medium. Concentration ratios were calculated for  $^{241}\text{Am}$ ,  $^{237}\text{Np}$ ,  $^{244}\text{Cm}$ , and  $^{238}\text{Pu}$ . The CR for americium is 0.002, while the CR for neptunium is 0.04. The CR for plutonium is 0.0001.

Studies performed on the effect of chelating agents on the transport of americium within plants has shown that chelating agents increase the transport of americium through a plant as opposed to leaving it in the root as shown in Table 5. Americium is especially available to the plant at pH 7.7 and americium, being a metal, is transported through the plant as a metal chelate.

Tests by Schreckhise and Cline showed that americium and other transuranics behave differently in the biosphere.<sup>14</sup> The phytoavailability of four transuranics (americium, plutonium, curium, and neptunium) was tested in field conditions to measure their uptake into four plant species including alfalfa, barley, peas, and cheatgrass (tumbleweeds). The soil concentration did not appear to affect the plant uptake of  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{244}\text{Cm}$ , or  $^{237}\text{Np}$ . The relative plant uptake of the radionuclides was

$$^{237}\text{Np} > ^{244}\text{Cm} \geq ^{241}\text{Am} > ^{239}\text{Pu} \geq ^{238}\text{Pu}$$

Relative uptake values for americium were 10 to 20 times greater than plutonium.

Based on the above information,  $^{241}\text{Am}$  tends to deposit within the roots of plants. The total uptake of  $^{241}\text{Am}$  within plants can be increased by using a chelating agent such as DTPA. The uptake in plants seems to be most accomplished within a soil pH range of 7 to 8.

**Table 5.**  $^{241}\text{Am}$  in roots, stems, and leaves of plants grown in contaminated soil in a glasshouse. <sup>15</sup>

$\mu\text{Ci } ^{241}\text{Am}$ per gram (dry weight)						
Soil pH	Roots	Lower stem	Primary leaf	Upper stem	Trifoliolate leaf	Ratio upper stem
Without DTPA						
5.3	6.87	0.40	0.76	1.56	0.17	0.11
6.4	5.03	0.47	0.21	0.64	0.18	0.28
7.7	4.14	0.44	0.24	0.68	0.24	0.35
8.7	2.87	0.07	0.11	0.43	0.65	1.51
With DTPA						
5.3	10.38	0.51	7.69	0.70	0.74	1.06
6.4	13.25	0.88	12.88	1.41	1.27	0.90
7.7	35.64	1.49	48.63	2.33	16.75	7.19
8.7	28.65	1.53	32.13	5.65	10.34	1.83

### Americium in Air

$^{241}\text{Am}$  is not normally found in air, except for those amounts that result from fallout due to an atmospheric test of a thermonuclear weapon or from a nuclear accident (reactor accident such as the Chernobyl disaster or weapons facility accident such as the fire at Rocky Flats).  $^{241}\text{Am}$  that has been released will eventually deposit in the land or ocean as fallout. As a result, there are detectable amounts of americium in soils, water, and, in some cases, air.

The major concern related to  $^{241}\text{Am}$  is the resuspension in air associated with dusts and other particulates. Because of the highly charged valence states of  $\text{Am}^{+3,+4,+5}$ , this radionuclide tends to bind easily with dust particles that can become suspended in air. Researchers at DOE's Idaho National Engineering Laboratory study the behavior of transuranic radionuclides in air.<sup>16</sup> One study used contaminated soil from the DOE's Rocky Flats facility and suspended dust particles into the air containing transuranic radionuclides in a wind tunnel having laminar air flows. The respirable fraction (<5 microns in particle size) of  $^{241}\text{Am}$  suspended in the air averaged 38% of the total activity of  $^{241}\text{Am}$  in the source soil. The  $^{241}\text{Am}$  was found to be extremely mobile downstream of the generation point. While  $^{241}\text{Am}$  is not normally found in air, extreme care must be used when working in areas that may contain transuranic radionuclide contamination to limit the possibility of the spread of contamination and possible inhalation from the resuspension of contaminated dust particles.

## BEHAVIOR OF AMERICIUM-241 IN ANIMALS AND HUMANS

Radionuclides have been classified as to their relative toxicity in humans. In this report, radionuclides are grouped into four classifications: very high toxicity, high toxicity, medium toxicity, and low toxicity.  $^{241}\text{Am}$  is included in the very high toxicity group.

$^{241}\text{Am}$  is primarily an alpha emitter. In addition,  $^{241}\text{Am}$  emits x-rays and gamma-rays with energies up to 955.7 keV. As previously discussed, alpha particles have a short range in air and are easily shielded with paper or plastic and do not pose an external radiation hazard. However, alpha particles pose a extreme hazard if they are internally ingested or inhaled. Alpha particles tend to deposit all of their energy in a small space or area when they interact with other matter. Because of this, alpha particles tend to cause damage when they interact with living tissue. For this reason,  $^{241}\text{Am}$  is included in the very high toxicity group.

Several animal studies have been performed to determine the behavior of  $^{241}\text{Am}$  in animals. Studies have been performed on ducks, baboons, and monkeys.<sup>17,18,19</sup> In all of these studies, the dose from internal exposure to  $^{241}\text{Am}$  was primarily deposited, in order of greatest dose to least, to the bone surface, liver, and lung.

The biological hazard associated with a radionuclide can be characterized by the equivalent dose per unit of exposure. The exposure can be either by external radiation (primarily from gamma-rays, neutron flux, or both, or to a lesser extent, from beta-rays if they are sufficiently energetic). In addition, internal exposure can result from ingestion or inhalation. Table 6 lists the  $^{241}\text{Am}$  equivalent dose [sievert (Sv) or rem dose per internal unit of exposure becquerel (Bq) or curie (Ci)] for the principle human organs of concern, gonad, breast, lung, bone, thyroid, as well as overall dose equivalent.

**Table 6.** Committed dose equivalents per unit uptake via ingestion and inhalation.<sup>3</sup>

Organ	Ingestion effective dose Sv/Bq <sup>a</sup>	Inhalation effective dose Sv/Bq <sup>a</sup>
Gonad	2.70 E-07	3.25 E-05
Breast	2.62 E-11	2.67 E-09
Lung	3.36 E-11	1.80 E-05
Red marrow	1.45 E-06	1.74 E-04
Bone	1.81 E-05	2.17 E-03
Thyroid	1.32 E-11	5.64 E-10
Other (e.g., liver, kidney)	6.66 E-07	7.48 E-05
Effective dose	1.58 E-08	1.20 E-04

a. To convert to conventional units (mrem/Ci), multiply table entry by  $3.7 \text{ E}+10$ .

Two independent means can remove any radionuclide from the human body. The first is radioactive decay, characterized by the radiological half-life, where the nuclide is transmuted into another nuclide. For  $^{241}\text{Am}$ , the daughter decay product is  $^{237}\text{Np}$ , which itself is primarily an alpha emitter. This decay product can continue to pose a biological hazard. The second process is quantified by the "biological half-life," which is a measure of time from removal of one-half of the nuclide from the affected organ(s) and can differ for different body parts. The total removal rate is characterized by the "effective half-life," which is defined as

$$T_{\text{eff}} = \frac{T_r * T_b}{T_r + T_b}$$

where:

$T_{\text{eff}}$  = Effective half-life

$T_r$  = Radiological half-life

$T_b$  = Biological half-life.

The biological half-life of  $^{241}\text{Am}$  to the critical organs is 70,000 days for the bone, 30,000 days to the liver, 30,000 days to the kidneys, and 70,000 days to the total body.<sup>20</sup> Thus, the radiological half-life is longer than the biological half-life and the only removal mechanism is biological. However, studies have been conducted showing that zinc-diethylenetriaminepentaacetate (Zn-DTPA) is an effective chelating agent to enhance the removal of  $^{241}\text{Am}$  from the body and to limit the uptake of  $^{241}\text{Am}$  to the critical organs. In addition, Zn-DTPA has been found to be effective in limiting the maternal transfer of  $^{241}\text{Am}$  to unborn mice.<sup>21</sup>

Several studies have been performed on humans who have been subjected to accidental exposures to  $^{241}\text{Am}$ . On August 30, 1976, a worker at DOE's Hanford facility received an accidental exposure to  $^{241}\text{Am}$ .<sup>22</sup> The estimated total dose received by the worker was 130 rad to the lung, 160 rad to the liver, and 105 rad/year to the bone. (The actual energy absorption is measured in rads, with 1 rad as 0.01 watt-second energy absorbed per kilogram of tissue weight.) This worker was subjected to Zn-DTPA therapy some 2.5 hours after the accident. It was reported that if the chelating therapy had been administered immediately after the accident that the majority of the dose to the lungs, liver, and bone might have been avoided.

The Los Alamos National Laboratory (LANL) published a report analyzing bone and tissues from individuals who had occupational exposures to  $^{241}\text{Am}$ .<sup>23</sup> Samples were taken after the natural death of the individuals, and the  $^{241}\text{Am}$  activity of tissues and bones were calculated years after exposure. The results confirmed that the bones had the greatest  $^{241}\text{Am}$  activity followed by the liver and lung. The study concluded that 83% of the total  $^{241}\text{Am}$  was located in the bones and teeth. The remaining 18% was located within the soft tissues.

The annual limits on intake (ALI) and derived air concentrations (DAC) for  $^{241}\text{Am}$  are shown in Table 7. The ALI is defined as that annual intake of a radionuclide that would result in a radiation dose to a human (for calculational purposes, the characteristics of the "reference or standard" man are used) equal to the allowable limit. The DAC is defined as that concentration of a radionuclide in air which, if breathed for a work-year, would result in an intake corresponding to the ALI (or, in case of

submersion, to an external exposure corresponding to the primary guide for limiting the annual dose). DACs are used for limiting radionuclides through breathing, or submersion in, contaminated air. ALIs are used primarily for assessing doses that result from accidental ingestion of radionuclides.

Historically, maximum permissible concentrations (MPC) in air and water have been used to determine safety guidelines for released radionuclide concentrations. Currently, the derived guidelines are presented in terms of ALIs and DACs. For a radionuclide whose derived value does not change from the old definition, the DAC is numerically equal to the MPC value in air. The information in Table 7 applies only to  $^{241}\text{Am}$ . In the case of multiple radionuclides released in a mixture, additional guidelines outlined in the 10 CFR 20 must be followed.<sup>24</sup>

In summary,  $^{241}\text{Am}$  is primarily an alpha emitter. It is most dangerous in animals and humans when it is taken into the body via inhalation or ingestion where it can interact with living tissue. The critical organs of concern in internal exposures to  $^{241}\text{Am}$  are the bone, liver, and lung. Chelating therapy using Zn-DTPA has been found to be effective in limiting the internal exposure and dose to the critical organs when administered as quickly as possible after exposure occurs. Because of the internal hazard posed by  $^{241}\text{Am}$ , the regulatory guidance gives very low ALIs and DACs to protect individuals from hazardous exposures.

**Table 7.** Annual limits on intake and derived air concentrations for  $^{241}\text{Am}$ .<sup>a</sup>

Radionuclide	Component	Ingestion ( $\mu\text{Ci}$ )	Inhalation ( $\mu\text{Ci}/\text{mL}$ )
$^{241}\text{Am}$ (W) <sup>b</sup>	ALI	8 E-01	6 E-03
	ALI bone surface	1	1 E-02
	DAC	NA <sup>c</sup>	3 E-12
	$f_1$ <sup>d</sup>	3 E-3	3 E-3

a. Data taken from 10 CFR 20 [20.1001-20.2402], Nuclear Regulatory Commission Regulations Appendix B.

b. W = ALIs and DACs for this nuclide are based on a weekly exposure.

c. Not applicable.

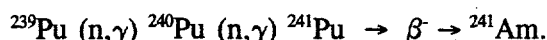
d. The  $f_1$  is the fractional uptake to the blood from the gastrointestinal tract from the common chemical forms of the radionuclide.



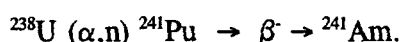
## CONCLUSIONS

$^{241}\text{Am}$  is an alpha-emitting transuranic radionuclide with a half-life of 432.2 years. The regulatory limits for  $^{241}\text{Am}$  disposal and personnel exposure are very restrictive. Relatively small concentrations of  $^{241}\text{Am}$  can become a major contributor to the calculated doses to individuals in performance assessment calculations or an accidental release involving this radionuclide.

The principle means of production of  $^{241}\text{Am}$  is by the bombardment of plutonium atoms in a nuclear reactor in the following reaction:



$^{241}\text{Am}$  can also be produced as a result of the bombardment of  $^{238}\text{U}$  with 40-Mev helium ions in the following reaction:



$^{241}\text{Am}$  is primarily an alpha emitter. The alpha particle energies range from 4.75 to 5.54 MeV with the most intense alpha particle energy at 5.48 MeV (85.2%). The radionuclide also emits gamma-rays and x-rays ranging from 11.87 to 955.7 keV with the most intense gamma energy emitted at 59.5 keV (35.7%).

Pure  $^{241}\text{Am}$  is produced by chemically separating the Am from its  $^{241}\text{Pu}$  parent.  $^{241}\text{Pu}$  is dissolved totally into a solution. After standing for a period of time, perhaps on the order of several years, the americium is then chemically separated from the plutonium solution.

$^{241}\text{Am}$  is produced within the fuel and operational wastes from cleanup systems of nuclear reactors. The quantity of  $^{241}\text{Am}$  in spent fuel that is produced each year is proportional to the amount of  $^{241}\text{Pu}$  produced and continues to increase as the fuel is stored. All of the  $^{241}\text{Am}$  associated with spent commercial fuel is expected to stay with the fuel and be disposed of in the national high-level waste repository.

The largest producer of  $^{241}\text{Am}$  is the U. S. Government. Government wastestreams containing  $^{241}\text{Am}$  are associated primarily with the manufacturing of atomic weapons and decommissioning of the associated Government facilities. The majority of the waste forms containing  $^{241}\text{Am}$  are located at the DOE facilities located around the country.

Studies have shown that the behavior of  $^{241}\text{Am}$  in soils depends on soil type and pH. Most studies agree that  $^{241}\text{Am}$  is not very mobile in soils and that only small quantities can be expected to migrate within the soil to the biosphere.

$^{241}\text{Am}$  generally does not migrate within groundwater and tends to bind with colloidal mixtures and clays found within the ground.

$^{241}\text{Am}$  tends to deposit within the roots of plants. The total uptake of  $^{241}\text{Am}$  within plants can be increased by using a chelating agent such as DTPA. The uptake in plants seems to be most accomplished within a soil pH range of 7 to 8.

$^{241}\text{Am}$  is not normally found in air. However, extreme care must be used when working in areas that may contain transuranic radionuclide contamination to limit the possibility of contamination

spread and possible inhalation from the resuspension of contaminated dust particles.

As stated,  $^{241}\text{Am}$  is primarily an alpha emitter. It is most dangerous to animals and humans when it is taken into the body via inhalation or ingestion where it can interact with living tissue. The critical organs of concern in internal exposures to  $^{241}\text{Am}$  are the bone, liver, and lung. Chelating therapy using Zn-DTPA has been found to be effective in limiting the internal exposure and dose to the critical organs when administered as quickly as possible after exposure occurs. Because of the internal hazard posed by  $^{241}\text{Am}$ , the regulatory guidance gives low ALIs and DACs to protect individuals from hazardous exposures.

## REFERENCES

1. E. Brown and R. B. Firestone, *Table of Radioactive Isotopes*, John Wiley and Sons, 1986, p. 241-2.
2. G. Seaborg and J. Katz, *The Actinide Elements*, 1st edition, McGraw-Hill, 1954, pp. 492-494.
3. B. Shleien, *The Health Physics and Radiological Health Handbook*, revised edition, Scinta, Inc., 1992, pp. 173, 238.
4. R. Ranier and D. Gambini, *Applied Radiobiology and Radiation Protection*, Ellis Horwood, 1990, pp. 346-347.
5. D. Lide, *Handbook of Chemistry and Physics*, 71st edition, Chemical Rubber Co., 1990-1991, pp. 4-42.
6. J. Katz and G. Seaborg, *The Chemistry of the Actinide Elements*, John Wiley and Sons, 1957, pp. 333-334.
7. Oak Ridge National Laboratory, *Integrated Database Report-1993: U.S. Spent Nuclear Fuel and Radioactive Waste Inventories, Projections, and Characteristics*, DOE/RW-006, Revision 10, December 1994, pp. 19-37, 111-120.
8. Code of Federal Regulations, 10 CFR 61.55, "Licensing Requirements for Land Disposal of Radioactive Wastes," Federal Register, Volume 47, No. 248, December 1982.
9. H. Nishita et al., "Effect of Soil Type on the Extractability of  $^{237}\text{Np}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$ , and  $^{244}\text{Cm}$  as a Function of pH," *Soil Science Magazine*, 132, No. 1, July 1981, pp. 25-34.
10. B. Hajek, *Plutonium and Americium Mobility in Soils*, Pacific Northwest Laboratories, BNLW-CC-925, November 1966.
11. P. Salter and G. Jacobs, *Evaluation of Radionuclide Transport: Effect of Radionuclide Sorption and Solubility*, Rockwell International, Hanford, RHO-BW-SA-192 P, June 1982.
12. W. Penrose et al., "Mobility of Plutonium and Americium Through a Shallow Aquifer in a Semi-Arid Region," *Environmental Science Technology*, 24, 1990, pp. 228-234.
13. A. Wallace, E. Romney, R. Mueller, S. Soufi, "Effect of Concentration on  $^{241}\text{Am}$  Uptake by Plants With and Without DTPA Treatment," *Soil Science*, 132, No. 1, July 1981, pp. 108-113.
14. R. G. Schreckhise and J. Cline, "Comparative Uptake and Distribution of Plutonium, Americium, Curium, and Neptunium in Four Plant Species," *Health Physics*, 38, May 1980, pp. 817-824.
15. A. Wallace, "Effect of Soil pH and Chelating Agent (DTPA) on Uptake by and Distribution of  $^{241}\text{Am}$  in Plant Parts of Bush Beans," *Radiation Botany*, 12, 1972, pp. 422-435.
16. D. Scott and M. Winberg, *Background Ventilation Studies for TRU-Waste Retrieval*, EG&G Idaho, EGG-WM-8802, October 1989.

17. O. D. Markham et al., "Plutonium, Am, Cm, and Sr in Ducks Maintained on Radioactive Leaching Ponds in Southeastern Idaho," *Health Physics*, 55, No. 3, September 1988, pp. 517-524.
18. J. Rosen, N. Cohen, M. Wrenn, "Short-Term Metabolism of  $^{241}\text{Am}$  in the Adult Baboon," *Health Physics*, 22, June 1972, pp. 621-626.
19. T. Lynch et al., "Comparative Skeletal Distribution of Am and Pu in Man, Monkey, and Baboon," *Health Physics*, 5, Supplement 1, pp. 81-89.
20. Y. Wang, M. D. (ed.), *Handbook of Radioactive Nuclides*, Chemical Rubber Company, 1969, p. 33.
21. R. Lloyd et al., "Effect of ZN-DTPA Therapy on the Maternal Transfer of  $^{241}\text{Am}$  or  $^{237}\text{Pu}$  to Young Mice," *Health Physics*, 49, No. 3, September 1985, pp. 405-410.
22. R. Thompson, 1976, "Hanford Americium Exposure Incident: Overview and Perspective," *Health Physics*, 45, No. 4, October 1983, pp. 837-845.
23. J. McInroy et al., "Part IV: Preparation and Analysis of the Tissues and Bones," *Health Physics*, 49, No. 4, October 1983, pp. 587-621.
24. Code of Federal Regulations, 10 CFR 20, "Standards for Protection Against Radiation," Nuclear Regulatory Commission Regulations.

## BIBLIOGRAPHY

Browne, E. and R. B. Firestone, *Table of Radioactive Isotopes*, John Wiley and Sons, 1986.

Linde, D. R., *Handbook of Chemistry and Physics*, Chemical Rubber Company, 71st Edition, 1990-1991.

National Low-Level Waste Management Program, *Characterization of Greater-Than-Class C Sealed Sources, Volumes 1-3*, DOE/LLW-163, 1994.

Oak Ridge National Laboratory, *Integrated Database Report - 1993: U.S. Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics*, DOE/RW-0006, Revision 10, December 1994.

Shleien, B., *The Health Physics and Radiological Handbook*, revised edition, Scinta, Inc., 1986.

Shapiro, J., *Radiation Protection*, second edition, Harvard Press, 1972.