

## CHEMISTRY OF THE ACTINIDE ELEMENTS

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## Summary

This chapter describes selected aspects of the physical, chemical, and biological properties of the actinide elements, their typical compounds, and their ions in aqueous solutions. These 5f-block elements have many unique features, and a comparison with similar species of the lanthanide (4f-block) and actinide transition series provides valuable insights into the properties of both. Some comparative data are presented on the electronic configurations, oxidation states, oxidation-reduction (redox) potentials, thermochemical data, crystal structures, and ionic radii of the actinide elements, as well as their environmental properties and toxicology.

## 1. Introduction

The actinide elements are the fifteen chemical elements with atomic numbers 89 through 103, the first member of which is actinium and the last of which is lawrencium. The actinides are a transition series of elements that fill the 5f subshell. The actinide series is unique in several respects:

- Most of the elements (those heavier than uranium) were first discovered by synthetic methods: bombardment of heavy atoms with neutrons in nuclear reactors, bombardment with other particles in accelerators, or as the result of nuclear detonations.
- All actinide isotopes are radioactive, with a wide range of nuclear decay properties, especially that of spontaneous and induced nuclear fission.
- They are all metals with very large radii, and exist in chemical compounds and in solution as cations with very large ionic radii.
- The metals exhibit an unusual range of physical properties. Plutonium, with six allotropes, is the most unusual of all metals.
- Many of the actinide elements have a large number of oxidation states. In this respect plutonium is unique, being able to exist in aqueous solution simultaneously in four oxidation states.
- In metallic materials and in some other compounds with elements lighter than plutonium, the 5f orbitals are sufficiently diffuse that the electrons in these

orbitals are “itinerant” (delocalized, chemically bonding, often with unique magnetic moments and electrical conductivity). In metallic materials and in most compounds with elements heavier than plutonium the 5f electrons are “localized” (not contributing significantly to electrical conductivity or to chemical bonds). Materials with plutonium and adjacent elements can exhibit both itinerant and localized behavior, depending on conditions such as temperature and applied pressure.

- Actinium (which has no 5f electrons in the metal, free atom, or in any of its ions) and the elements americium through lawrencium are similar in many respects to the lanthanide elements (the elements that fill the 4f electron subshell). The elements, thorium through neptunium, have some properties similar to those of the d transition elements.
- Relativistic contributions to electronic properties and spin-orbit effects are important in the chemical properties of actinides.

## 2. Sources of Actinide Elements

### 2.1. Natural Sources

The elements, actinium through plutonium, occur in nature. Only the elements thorium, protactinium, and uranium are present in amounts sufficient to warrant extraction from natural sources. Thorium and uranium are widely disseminated in the Earth's crust (~10000 and ~3000  $\mu\text{g}/\text{kg}$  respectively), and, in the case of uranium, in significant concentrations in the oceans (3.3  $\mu\text{g}/\text{L}$ ). More importantly, thorium and uranium are found highly enriched in certain mineral formations, and are obtained by conventional mining operations. Extraction of thorium and uranium from their ores had been practiced for many years prior to the discovery of the transuranium elements, and an extensive technology exists for the extraction of thorium and uranium from many different types of ores.

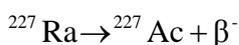
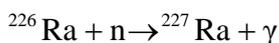
Hundreds of thousands of tons of uranium have been processed for isotopic separation. Five grades of isotopically separated uranium are commonly recognized: depleted uranium (less than 0.71%  $^{235}\text{U}$ ), natural uranium (0.71%  $^{235}\text{U}$ ), low-enriched uranium (0.71-20%  $^{235}\text{U}$ ), highly enriched uranium (HEU) (20-90%  $^{235}\text{U}$ ), and weapon-grade uranium (greater than 90%  $^{235}\text{U}$ ). Of these grades, HEU and weapon-grade uranium are the most important. Highly enriched uranium (HEU) is produced from natural uranium for use in nuclear weapons and for use in commercial nuclear reactors. Worldwide, at the end of 1994, 450 metric tons of HEU (calculated as weapon-grade equivalent) was inside nuclear weapons and 1300 tons had been removed from weapons. An additional 20 tons exist in civil inventories. This total (1770 tons) is slowly being decreased (20 tons per year in 1996) by blending down of Russian HEU.

Neptunium ( $^{237}\text{Np}$  and  $^{239}\text{Np}$ ) and plutonium ( $^{239}\text{Pu}$ ) are present in extremely minute amounts in nature as a result of natural nuclear reactions with neutrons in uranium ores. The longer-lived  $^{244}\text{Pu}$  has been found in the rare-earth mineral bastnasite to the extent of 1 part in  $10^{11}$ . This may possibly be a primordial endowment.

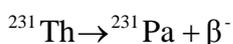
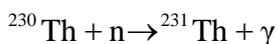
Actinium and protactinium are decay products of the naturally occurring uranium isotope  $^{235}\text{U}$  and are present in uranium minerals in such low concentration that recovery from natural sources is a very difficult task. For their artificial production, see the next section.

## 2.2. Artificial Production of Actinides

It is relatively straightforward to obtain actinium, protactinium, and most of the remaining transuranium elements by neutron irradiation of elements of lower atomic number in nuclear reactors. Thus, actinium has been produced in multigram quantities by the transmutation of radium with neutrons produced in a high-flux nuclear reactor:

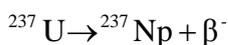
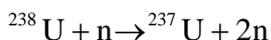


The product actinium can be separated from the precursor radium by solvent extraction or ion exchange, and gram amounts of actinium have been obtained by this procedure. This is not at all an easy task, considering the highly radioactive substances involved and the hazards of radon emission that accompanies these nuclear reactions, but it is preferable by far to extraction from natural sources. Protactinium can be produced by the nuclear reactions:

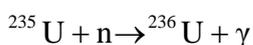


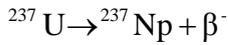
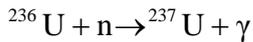
The amount of  $^{231}\text{Pa}$  that has been produced in this way, however, is much less than the amounts (more than 100 g) of protactinium that have been obtained from residues accumulated from the very large-scale extraction of uranium from ores. Because of the extreme tendency of protactinium(V) to form colloidal polymers that are easily adsorbed on solid surfaces, and cannot be removed from aqueous media by solvent extraction, the recovery of protactinium from uranium ore processing residues can only be described as a heroic enterprise.

Neptunium-237 is a long-lived isotope of element 93 that is produced in kilogram amounts. It is formed as a by-product in nuclear reactors when neutrons produced in the fission of uranium-235 react with uranium-238:



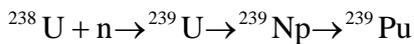
Neptunium-237 is also formed by neutron capture in uranium-235:



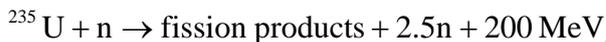


The waste solutions from the processing of irradiated uranium fuel usually contain the neptunium, which can be isolated and purified by a combination of solvent extraction, ion exchange, and precipitation techniques.

The strategically important isotope  $^{239}\text{Pu}$  is produced by the ton in nuclear reactors. Excess neutrons from the fission of uranium-235 are captured by uranium-238 to yield plutonium-239:



Neutrons also cause the uranium nuclei to fission:



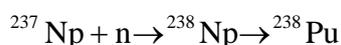
After removal from the reactor, the irradiated fuel can be chemically separated to extract the plutonium and also to separate the highly radioactive fission products.

Plutonium produced in nuclear reactors in which the fuel is irradiated for long periods of time contains plutonium isotopes with mass numbers up to 244, formed from  $^{239}\text{Pu}$  by successive neutron capture. Three grades of plutonium are commonly recognized: weapon-grade plutonium (less than 7%  $^{240}\text{Pu}$ ), fuel-grade plutonium (7-18%  $^{240}\text{Pu}$ ), and reactor-grade plutonium (more than 18%  $^{240}\text{Pu}$ ).

Plutonium is an element that was until recently produced in “production” reactors, i.e. reactors that were dedicated to produce nuclear materials for military purposes. More than 260 metric tons of military plutonium have been produced and separated worldwide. Plutonium is also formed as a by-product of electricity production in all commercial (civilian) nuclear power reactors. As of 2000 the total “discharge” of plutonium (plutonium in and separated from civilian spent fuel) from commercial nuclear power reactors was estimated to be 1,380 metric tons, and this amount is predicted to increase by 741 metric tons in the decade 2001-2010.

Although highly enriched uranium (HEU) and weapon-grade plutonium are primarily for military purposes and the lower grades are used in commercial nuclear reactors, enriched uranium and plutonium of all grades are commonly discussed together because  $^{235}\text{U}$  and  $^{239}\text{Pu}$  are the two primary fissile isotopes and because they have the greatest security risk of potential diversion from peaceful to military or terrorist uses. Therefore the total inventory must include both military and civilian stocks.

The nuclide  $^{238}\text{Pu}$  is an important heat source for terrestrial and extraterrestrial applications. The heat is generated by its nuclear alpha-particle decay. This nuclide is available in kilogram quantities from the neutron irradiation of neptunium-237:



followed by chemical separation. Its 87.7-year half-life makes it the best nuclide for this purpose.

The elements americium and curium are obtained as by-products of the large-scale production of plutonium-239, or by irradiation of plutonium-239 or isotopes of transplutonium elements in special high-neutron-flux reactors. The plutonium in a typical pressurized water reactor fuel is approximately 14%  $^{241}\text{Pu}$  after 33,000 Mwd/t burnup, producing  $^{241}\text{Am}$  by beta decay with a half-life of 14.4 years. Kilogram quantities of americium-241 can be separated from irradiated fuel by a combination of precipitation, ion exchange, and solvent extraction.

Isotopes of curium are also found in waste streams from plutonium-239 production, but in amounts smaller than those of americium. Curium (as  $^{242}\text{Cm}$  and  $^{244}\text{Cm}$ ) is produced in nuclear reactors primarily by the beta decay of  $^{242}\text{Am}$  and  $^{244}\text{Am}$ , isotopes that were formed by neutron capture in  $^{241}\text{Am}$  and  $^{243}\text{Am}$ , respectively. The amount of  $^{244}\text{Cm}$  accumulated in process wastes and in unprocessed irradiated fuel elements as of 2000 is estimated to be more than one ton. The high specific radioactivity (18.1-year half-life) of  $^{244}\text{Cm}$  means that it has to be considered in separation schemes for reprocessing and transmutation, but it decays to low enough levels in a few hundred years that long-term storage is not a concern. Separation and purification of curium and americium is best carried out by the ion-exchange procedures described below because both are present as +3 ions, which have similar chemical properties.

The sequence of neutron captures and beta decays that forms transuranium elements by slow neutron capture starting with plutonium-239 is shown in Figure 1. A high neutron flux is essential to expedite the production of transplutonium elements. Starting with 1 kg of  $^{239}\text{Pu}$ , about 1 mg of  $^{252}\text{Cf}$  would be present after 5-10 years of continuous irradiation at a neutron flux of  $3 \times 10^{14} \text{ cm}^{-2} \cdot \text{s}^{-1}$ . To increase the production rate, large quantities of  $^{239}\text{Pu}$  can first be irradiated in production reactors, followed by continued irradiation in higher-neutron-flux reactors. The High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory in Tennessee can provide neutron fluxes of about  $2 \times 10^{15} \text{ cm}^{-2} \cdot \text{s}^{-1}$ . This reactor has made major contributions to the production of transcurium elements. A special facility was established at Oak Ridge in 1966 to fabricate plutonium targets, to extract transplutonium elements from the highly irradiated targets, and to provide pure samples of transplutonium nuclides as heavy as  $^{257}\text{Fm}$  for research and industry. Neutron irradiation cannot be used to prepare the elements beyond fermium ( $^{257}\text{Fm}$ ) because some of the intermediate nuclides have such short half-lives that the low equilibrium concentrations present effectively prevent the formation of significant amounts of the desired elements. Thus, only milligram amounts of einsteinium and picogram amounts of fermium can be obtained by protracted neutron

irradiation even under the most favorable circumstances. Isotopes that are sufficiently long-lived for work in weighable amounts are obtainable at least in principle for all of the actinide elements through einsteinium. The elements above einsteinium appear likely to remain amenable to chemical study only by tracer techniques because (except for  $^{257}\text{Fm}$ ) only isotopes with short half-lives are known, and because it is unlikely that isotopes of the elements beyond einsteinium can be formed in weighable quantities by neutron irradiation or any other process.

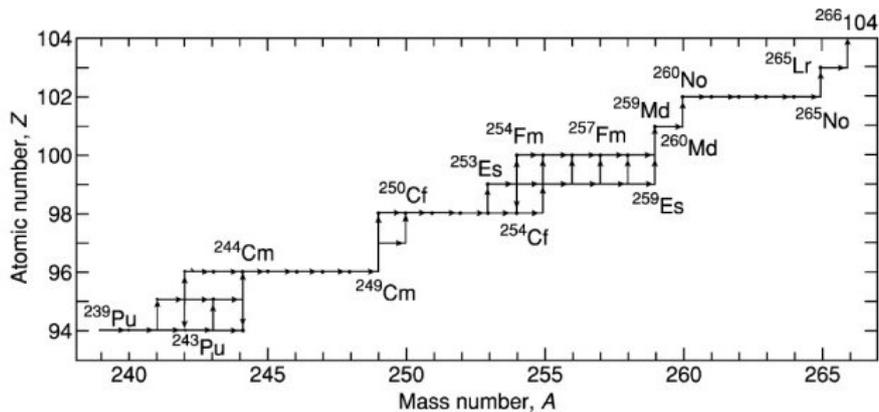


Figure 1. Nuclear reaction sequence for production of transplutonium elements by intensive slow-neutron irradiation. The principal path is shown by heavy arrows (horizontal, neutron capture; vertical, beta decay). The sequence above  $^{258}\text{Fm}$  is a prediction.

Although Figure 1 indicates that the heaviest actinides (Md-Lr) are produced in nuclear reactors, the half-lives of their isotopes are so short that the only practical way to study these elements is to synthesize them by bombardment of heavy-element targets such as  $^{249}\text{Cf}$  with ions such as  $\alpha$  particles or  $^{16}\text{O}$  in cyclotrons or accelerators, followed by very rapid separation. Table 1 lists the primary actinide nuclei used for laboratory studies.

Element, Z	Isotope	Half-life
actinium, 89	$^{227}\text{Ac}$	21.772 a
thorium, 90	$^{232}\text{Th}$	$1.405 \times 10^{10}$ a
protactinium, 91	$^{231}\text{Pa}$	$3.276 \times 10^4$ a
uranium, 92	$^{233}\text{U}$	$1.592 \times 10^5$ a
	$^{238}\text{U}_a$	$4.468 \times 10^9$ a
neptunium, 93	$^{236}\text{Np}$	$1.54 \times 10^5$ a
	$^{237}\text{Np}$	$2.144 \times 10^6$ a
plutonium, 94	$^{238}\text{Pu}$	87.7 a
	$^{239}\text{Pu}$	$2.411 \times 10^4$ a
	$^{240}\text{Pu}$	6564 a
	$^{242}\text{Pu}$	$3.733 \times 10^5$ a

	$^{244}\text{Pu}$	$8.08 \times 10^7$ a
americium, 95	$^{241}\text{Am}$	432.2 a
	$^{243}\text{Am}$	7370 a
curium, 96	$^{244}\text{Cm}$	18.10 a
	$^{245}\text{Cm}$	8500 a
	$^{246}\text{Cm}$	4760 a
	$^{247}\text{Cm}$	$1.56 \times 10^7$ a
	$^{248}\text{Cm}$	$3.48 \times 10^5$ a
	$^{250}\text{Cm}^b$	~8300 a
berkelium, 97	$^{247}\text{Bk}^c$	1380 a
	$^{249}\text{Bk}$	330 d
californium, 98	$^{249}\text{Cf}$	351 a
	$^{252}\text{Cf}$	2.645 a
einsteinium, 99	$^{253}\text{Es}$	20.47 d
	$^{254}\text{Es}$	275.7 d
	$^{255}\text{Es}$	39.8 d
fermium, 100	$^{257}\text{Fm}$	100.5 d
mendelevium, 101	none	
nobelium, 102	none	
lawrencium, 103	none	

<sup>a</sup> Natural isotopic composition is 99.275%  $^{238}\text{U}$ , 0.720%  $^{235}\text{U}$  and 0.005%  $^{234}\text{U}$ . Half-life given is for the major constituent  $^{238}\text{U}$ .

<sup>b</sup> Produced only in very small amounts from neutron irradiations in thermonuclear explosions.

<sup>c</sup> Produced so far only in tracer quantities from charged particle irradiations.

Table 1. Long-lived actinide nuclides suitable for physical and chemical investigation (a = year, d = day).

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### Biographical Sketches

**Norman M. Edelstein** was born in Chicago, Illinois and graduated from the University of Illinois with a B. S. degree in Chemistry in 1957. He did his graduate work with Professor R. J. Myers in the College of Chemistry at the University of California, Berkeley, California and received his Ph.D. in 1962. He spent two years in the Chemistry Department, Harvard University as a postdoctoral fellow with Professor August H. Maki. In 1964 he joined the University of California Radiation Laboratory (now the Lawrence Berkeley National Laboratory, LBNL) as a Research Chemist in the Nuclear Chemistry Division, working in the actinide chemistry group of Professor Burris Cunningham. He was promoted to the Senior Staff, Nuclear Chemistry Division in 1966. Upon the death of Professor Cunningham in 1971 he became the group leader of the actinide chemistry group. In 1972 Dr. Edelstein spent a sabbatical year at the University of Oxford, Physical Chemistry Laboratory as a NATO Senior Fellow and returned to the Lawrence Berkeley Laboratory in 1973. From 1975 – 1999 he was group leader of the actinide chemistry group and principal investigator in the Chemical Sciences Division, LBNL. From 1991 to 1999 he also was the Deputy Division Director of the Chemical Sciences Division except for the period 1998 – 1999 when he was Acting Division Director of the Chemical Sciences Division. In 2000 – 2001 Dr. Edelstein

was on assignment to the Department of Energy, Office of Science, Basic Energy Sciences, as acting program manager for the Heavy Element Chemistry program. He returned to LBNL in 2001 and became an emeritus Senior Scientist in 2002.

Dr. Edelstein's research interests include the optical, magnetic, and electronic structure of the actinides and lanthanides, the general inorganic and solution chemistry of the actinides, and synchrotron radiation studies of actinides and other environmentally relevant materials. Dr. Edelstein has published over 220 scientific papers on these and other topics and has edited or co-edited three earlier volumes on actinide subjects. He is one of the three co-editors of a more recent work "The Chemistry of the Actinide and Transactinide Elements," 3<sup>rd</sup> ed., 5 vol., Springer, 2006.

**Lester R. Morss** was born in Boston, Massachusetts. He received his BA degree from Harvard University in chemistry and physics. After four years of military service, he carried out graduate research in transuranium chemistry at the University of California Radiation Laboratory (now Lawrence Berkeley National Laboratory) under Professor Burriss B. Cunningham, and he obtained his Ph.D. at University of California, Berkeley in 1969. His postdoctoral research was at Purdue University with James W. Cobble, followed by a decade as an assistant and associate professor of chemistry at Rutgers University, New Jersey. Most of his research career was in transuranium research at Argonne National Laboratory. His primary research interests are in the inorganic chemistry, thermochemistry, and solid-state chemistry of lanthanides and actinides, in particular micro-scale syntheses of pure compounds as well as structure-bonding relationships among metals, ionic and organometallic compounds, and ions in solution (especially ions in unusual oxidation states).

He had two sabbatical research fellowships, in 1978-79 at the University of Liège, Belgium with Professor Jean Fuger, and in 1993 as an Alexander von Humboldt Senior Scientist fellow at University of Hannover, Germany with Professor Gerd Meyer.

Dr. Morss has been an author of 100 scientific papers as well as 17 book chapters and scientific articles. He co-edited "The Chemistry of the Actinide Elements," 2nd edition with Joseph J. Katz and Glenn T. Seaborg, 1986, is one of the three co-editors of "The Chemistry of the Actinide and Transactinide Elements," 3<sup>rd</sup> ed., 5 vol., Springer, 2006, and co-edited two other books.

He has been the program manager for basic research in heavy element chemistry at US Department of Energy since 2002.