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Perspective on Plutonium†

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I. Introduction

Plutonium (Pu) was discovered in 1941 by G.T. Seaborg in the Radiation Laboratory of the University of California. An unknown alpha-emitter was noticed as an unexpected product of a reaction that was intended to produced neptunium-238. Following the planetary sequence, Seaborg dubbed it plutonium. The isotopes that Seaborg discovered (Pu-238 and Pu-239) were long-lived and underwent fission when exposed to thermal neutrons. These properties made them easier to study and better suited for atomic weapons than other materials. As a result, plutonium became one of the best-studied elements of the periodic table, but most of the studies remained classified for quite some time.

Plutonium is the first primarily man-made element to play a significant role not only in technological development (as subject of scientific investigation), but also in the economic growth of many countries. The importance of plutonium centers around its enormous energy: Pu-239 has a specific energy of 86.5×10^{12} J kg^{-1}, making it ideal for wide-scale use in reactors. While the nuclear industry continues to work toward improving safety and efficiency of plutonium as a reactor fuel, politicians and the public still debate over the safety and benefits of nuclear power. Many people perceive plutonium as sinister and dangerous; this is a result of its association with nuclear weapons. This paper is intended as a brief overview on the element plutonium, its uses as a power source, and concerns over its disposition.

II. Physical and Chemical Properties

Plutonium, atomic number 94, is a brittle, silvery-white metal with a density near 20 g cm^{-3}. It has six different allotropic modifications, each of which has its own mechanical properties, and a number of oxidation states with their own chemical properties. Information on nuclear and thermodynamic properties is readily available (Taube, 1974). Data on plutonium in aqueous solution and chemical separation have been comprehensively documented (*e.g.*, Cleveland, 1979).

In aqueous solution, plutonium exists in five states. The oxidation states (III, IV, V, and VI) can coexist simultaneously in water as a thermodynamically stable system. The most stable oxidation state is the tetravalent, which is readily hydrolized. In general, dissolved plutonium samples usually contain a host of undesirable components (especially other actinides like uranium, neptunium, and americium) and its isolation can be a difficult process.

Plutonium isotopes are generally products of beta-minus decay of neptunium or electron capture in americium. Plutonium emits X-rays of about 17 keV, regardless of isotope; the ratio of x-ray emission to total emission varies, *e.g.*, 4.6% of the emissions from Pu-239 are X-rays. Alpha particles of average energy 5.2 MeV are also emitted. Plutonium isotopes are also a source of beta-rays, gamma-rays, and neutrons. In practice, the intensity of radiation from Pu-239 and heavier isotopes is about 20-30 μGy h^{-1} at a distance of 1 m. The dose rate at the surface of 1 kg of metallic Pu-238 is about 10 mGy h^{-1} g^{-1}. Sixteen isotopes of plutonium have been observed, ranging from Pu-233, with a half-life of 21 min, to Pu-244, with a half-life of 83 million years. The most commonly recognized isotopes are Pu-239 and Pu-240, which are fission products of a nuclear spallation (or light-emitting) reactions. Pu-239 has a half-life of 24,390 yr, a specific activity of 2.3 GBq g^{-1}, and a thermal neutron cross section of 741 barns (10^{-24} cm^2) (Radiological Health Handbook, 1970).

III. Biological Effects

The alpha particles and x-rays emitted from plutonium isotopes are too weak to penetrate the

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skin; the element is of most concern when it is present internally. If inhaled, ingested, or absorbed through an open wound, even small amounts of plutonium can produce adverse effects. The maximum permissible body burden of Pu-239 recommended by the International Commission on Radiological Protection (ICRP) is only 0.64 µg, or 149 Bq (ICRP 2, 1959). The radiotoxicity index for Pu-239 was extrapolated originally from the total risk to bone in dogs from Ra-226.

Animal studies indicate that plutonium behaves metabolically like iron when it enters the blood. It binds easily with transferrin in the plasma, and is carried throughout the body. The major organs for deposition and accumulation of plutonium are the bones and liver. Once accumulated, plutonium is retained in the bone with a biological half-life of 100 y, i.e., longer than one's lifetime. Observations of the effects of plutonium in rats, monkeys, pigs, beagles, and other animals reveal that plutonium can induce osteosarcoma and related bone cancers (ICRP-19, 1972, Priest and Tasker, 1990). Since only 37 kBq of retained Pu-239 will result in a committed effective dose equivalent (CEDE) of 356 Sv over 50 y, even small amounts may present a very high risk for bone cancer.

IV. Detecting Plutonium in the Body

The average range of plutonium's alpha particles is extremely short: 40-80 µm. The x-rays have energies of about 17 keV and a half-thickness of absorption in human tissue of 6 mm. Due to this, neither the x-rays nor alpha particles may be detected *in vivo* (*ie.*, whole-body counting techniques are ineffective). In fact, it is impossible to detect activity below the dose limits recommended in ICRP Publication 60 in this manner. *In vitro* techniques, such as bioassay (*e.g.*, urinalysis) or air sampling, must be used to estimate the amount of plutonium in the body.

The sensitivity of detection depends greatly on the method used to assess plutonium. Electrodeposition, for example, is an alpha counting method that has a detection limit of about 400 µBq. A recent scintillation method, Photon Electron Rejection Alpha Liquid Scintillation (PERALS), has a detection level near 40 µBq. To quantify even smaller amounts of Pu-239 in human urine, fission track analysis (FTA) was developed (Moorthy, *et al.*, 1988). FTA takes advantage of the large cross-section of Pu-239, and is able to detect activity of about 4 µBq. This is a promising method for all low-level Pu-239 measurements for radiation and environmental protection programs.

V. Dosimetry and Bioassay Data

Biomedical research on plutonium in humans has been limited (MRC, 1975). Langham (1958) conducted the only study where soluble plutonium was intentionally injected into terminally ill human volunteers. By observing the patients over the next five years, Langham established two simple power functions to estimate the fraction of uptake excreted in the urine, U(t), and in the feces, F(t):

$$U(t) = 0.002t^{0.74} \quad \text{and} \quad F(t) = 0.0036t^{1.09}$$

where t is the time in days after uptake. Compared to the retention time of plutonium, Langham's observation period of five years is relatively short, so that these functions are reliable only for the first few hundred days after an acute intravenous injection.

Because of its uniqueness, Langham's model has been studied many times. It is believed that the Langham urinary function may underestimate long-term excretion and thus, overestimate original uptake. Because of the long-term behavior of plutonium, the health of the subjects, and additional data (most notably from surveys of Manhattan Project workers), modifications to Langham's functions have been suggested by several researchers. Most studies suggest a multiple-term exponential function including those by Beach and Dolphin (1964); Durbin(1972); Moss, *et al.*, 1983); Rundo, *et al.*, (1978); Jones (1985); Leggett (1984); and Sun (1987).

The ICRP-26 (1976) recommended that the ALI (Annual Limit on Intake for CEDE of 50 mSv) of all long-lived Pu compounds is 200 Bq for Class W (soluble) and 600 Bq for Class Y (insoluble) materials (ICRP-30, 1978). The new ICRP-60 Report recommended a 100 mSv occupational dose limit effective dose over five years (an average annual value of 20 mSv) with a limit of 50 mSv in any single year. The ALI for Pu-239 derived under the 1990 recommendations is 300 Bq (ICRP-61, 1991). An average adult may be expected to have at least 0.1 Bq of activity in his/her body due to worldwide fallout (Sing and Wrenn, 1983).

VI. Fallout Levels in the Environment

There is evidence that Pu-244 exists in trace amounts in meteorites and that it existed in the early solar system (Kuroda and Ganapathy, 1968), but all of the plutonium isotopes currently in the environment are products of weapon tests and nuclear power. Plutonium exists in the environment, usually entering the biosphere through plant life. It can also be ingested or inhaled. Many countries routinely measure plutonium levels in the air. A global survey (Holleman, *et al.*, 1987) indicated a level of 12,000 TBq for Pu-239 and 740 TBq for Pu-238, corresponding to an average of about 3.7×10^7 Bq km^{-2} for both isotopes. Holleman's report also notes a significant drop in the air activity, *e.g.* from 63 to 1.5 μBq m^{-3} in New York City between 1963 and 1972. This decrease corresponds to the cessation of above-ground testing by the nuclear powers. In general, such exposure is minor compared with the occupational exposure of those who work in facilities where plutonium is present.

Two recent studies dealt with Pu-239 contamination in soil near plutonium production facilities. In Rocky Flats, U.S., it was estimated that there has been a fivefold reduction in plutonium concentration in the soil in the past 15 years due to the natural erosion of 3 cm of soil (Webb, *et al.*, 1992). The current total estimate for the study area near the contamination source in Rocky Flats is 463 kBq m^{-2}. A later study conducted nearer to the Chernobyl site found 17 to 20 mBq g^{-1} of Pu-239+240 in soil, which was attributed to released fuel particles from that accident (Hölgye and Burčík, 1992).

VII. Concerns over Safety and Control

A preliminary draft of Basic Safety Standards (IAEA, 1992) was circulated in May 1992. The philosophy of these standards is consistent with the recent ICRP Publication 60 (1991): to provide a uniform system of radiological protection and dose limits concerning the use of radiation and radioactive materials in the workplace. Any quantities of plutonium isotopes 238, 239, and 240 with less than 10 Bq g^{-1} of activity are exempt form the regulations. This is the same exemption concentration listed for K-40 and Cs-137 and it is ten times larger than those for Co-60, Rn-222, Ra-226, and natural U-238 isotopes. These exempt concentrations would yield only an effective dose limit of 10 μSv y^{-1} for any member of the public.

Over the past 30 years, around 120 tons of weapon-grade plutonium fuel have been obtained from spent civil fuels worldwide. Two new processing plants, one in Britain and one in France, have been designed to extract usable plutonium from spent fuel and reduce the volume of high-level radioactive waste. Other countries are looking into this concept as well. As this implies, the greatest concern with plutonium has always been disposal of the waste rather than its use in general. It has an incredible potential as an energy source that does not adversely affect air quality. However, concerns over its toxic effects, including bone cancers and the proper handling of wastes, has kept many countries from fully embracing nuclear energy.

Japan's Atomic Energy Commission is openly committed to the use of nuclear power to free the country from dependence on foreign energy sources (*Nature*, 359/766, 1992). Consequently, Japan is preparing to receive about 30 tons of weapon-grade plutonium from France and Britain (*Nature* 359/663, 1992) despite the protests and concerns of its own citizens and international concern about shipping such large amounts of plutonium across the ocean.

At present, plutonium is not an object of wide international commerce, but, as the case of Japan shows, eventually it will be governed by free market conditions. The price also will be strongly influenced by the political development of our world, since, unfortunately, usable reactor technology goes hand in hand with the ability to produce nuclear weaponry. In spite of this, the future of plutonium should be discussed in a peaceful context, where it may serve as a basic fuel for energy production. However, the potential for weapons cannot be overlooked, and basic safeguards for health and methods of inventory, containment, and surveillance must be implemented in any nation using nuclear power.

The consequence of such development is that much of the plutonium becoming available will have to be dealt with as waste. The political decision to use nuclear energy depends heavily on the ability to obtain and apply objectively the necessary information for safe operation and control. The nuclear industry is a potentially dirty one and could have a serious environmental impact if not

managed conscientiously and intelligently. Thus far, the safety record for the industry has been good. It must stressed that environmental protection and restoration of the environment should be a significant consideration in the nuclear energy picture. As for plutonium toxicity, the policy has always been one of overestimation. Safe and secure methods have been developed for controlling plutonium (Faust, *et al.*, 1988). As a result, the question of whether or not a nation uses nuclear energy is more likely to be decided by politics and society than by scientists and engineers.

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