

Special Topic Notes: Radioactive Waste

A “Radiation Primer”

Radioactivity is defined as the emission of particles or electromagnetic radiation from the unstable nuclei of an element (recall that each atom generally has a nucleus containing neutrons and protons, with electrons orbiting around). Radioactivity occurs naturally in the environment. For example, there are more than 60 naturally occurring radioactive elements, which may be found in both soil and water. These include carbon-14, potassium-40, calcium-45, oxygen-15, and tritium. Solar radiation is also a significant contribution of human exposure to radiation. Radioactivity is also released into environment from coal combustion. Finally, a variety of radioactive products form from nuclear fission that is used to produce energy. A table of some radioactive isotopes is shown below.

element	Atomic #	Common Isotopes				
		Atomic Mass half-life				
C carbon	6	10 19.3 s	14 5,570 y			
Tl Thallium	81	206	207 4.78 m	210		
Pb Lead	82	210 22 y	211 36.1 m	212 0.44 d	214 26.8 min	
Bi Bismuth	83	210 5.01 d	211 2.15 m	212 0.42 d		214 19.7 min
Po Polonium	84	210 138.4 d	214 1.6E-4 sec	215 1.78E-3 s	216 0.15 s	218 3.05 min
At Astatome	85	218				
Rn Radon	86	219 4 s	220 6.4E-4 d	222 3.825 d		
Fr Francium	87					
Ra Radium	88	223 11.7 d	224 3.64 d	226 1620 y	228 2.4E3 d	
Ac Actinium	89	227 8000 d	228 0.26 d			
Th Thorium	90	227 228 18.4d 700d	230 75,200 y	231 1.07 d	232 1.41E10 y	234 24.1 d
Pa Proactinium	91	231 1.3E7 d	233 27 d	234 1.18 min		
U Uranium	92	233 1.6E5 y	234 2.48E5 y	235 2.6E11 d	238 4.51E9 y	239 23 m
Np Neptunium	93	239 2.35 d				
Pu Plutonium	94	239 24,000 y				
Am Americium	95					

There are three types of nuclear decay that are common to unstable nuclei:

alpha = 2 protons + 2 neutrons,

spontaneously emitted from many heavy nuclei with mass #s >150
decrease 4 in mass number and two in atomic number

very short range in air, interact with matter, energetic

beta = fast electron

emitted in decay of many radioisotopes

no mass change in the element, increase 1 atomic number

medium range in air

gamma = high energy photon

no change in element or mass occurs, just change from “excited” to “less excited”

release of energy

more penetrating than alpha or beta

In addition, x-rays are fairly high energy photons, similar to gamma rays. However, they are non-nuclear in origin (electromagnetic), occurring when electrons are transferred from a higher to a lower atomic energy state. They are very penetrating, but not as penetrating as gamma rays.

When nuclear decay occurs, the “parent” nuclide is converted into a “daughter” nuclide:
Parent Nuclide - Decay Particle = Daughter Nuclide

An example of radioactive decay is that of uranium-238. U238 is naturally present in ore in the earth’s crust (the relative abundance of 238 to 235 is 99.3:0.7. The average concentration of uranium in rock in the earth’s crust is 1 pCi/g (or 3 ppm).

Element	Atomic #	Mass #	half-life	decay	energy of decay particle, MeV
U	92	238	4.51 x E9 yrs	alpha	4.15
Th	90	234	24.1 days	beta	0.15
Pa	91	234	1.18 min	beta	0.7
U	92	234	2.48 x E5 yrs	alpha	4.6
Th	90	230	75,000 yrs	alpha	4.5
Ra	88	226	1620 yrs	alpha	4.3
Rn	86	222	3.8 days	alpha	5.0
Po	84	218	3.05 min	alpha	6.0
Pb	82	214	26.8 min	beta	0.65
Bi	83	214	19.7 min	beta	1.5
Po	84	214	1.6 x E-4 sec	alpha	7

Note: continues decay until it reaches lead (Atomic #=82, Mass=206), a stable product. The energy of the decay particle relates to its ability to cause damage; higher energy cause higher damage.

The “rate of decay” of radioactive compounds is first order

Rate of Decay = λ * N where λ = rate constant, N = # of parent atoms

half - life = time until the number of parent atoms has been reduced by half

half - life = $0.693 / \lambda$

Curie = unit of activity = 3.7×10^{10} dps

$A = \lambda * N = \text{decay constant} * \# \text{ of atoms} = 0.693/\text{half life} * N$

example: 10 g Pu-239, half-life 24,000 years

Activity = ?

$10 \text{ g Pu} * 1 \text{ mol}/239 \text{ g} * 6.02 \times 10^{23} \text{ atoms/mol} * 0.693/24000 \text{ yrs} = \text{dpy}$

Harmful effects of ionizing radiation to living organisms are due to the energy absorbed by cells in the body. This energy can disrupt the molecules in living cells. Ionizing radiation interacts with body tissue, and the amount of tissue damage is due to the amount of energy this radiation deposits in the body. Long-term effects of radiation include cancer, such as lung cancers in miners and leukemia in survivors of the atomic bomb at Hiroshima. This cancer is caused by the genetic mutation resulting from radiation interaction with cells. (~ 0.1 mrem/yr dose = excess of 10^{-6})

lifetime cancer risk). Acute effects will occur if a large dose of radiation is delivered to a large portion of the body, including nausea, fever, cardiovascular collapse. A “no effect” threshold is around 100 rads, while the mean lethal dose is 450 rads (LD-90 is 800 rads).

DOSE = quantity of radiation, based on energy absorbed, and is measured in rad.

A rad is equal to the absorption of 100 ergs of energy / g mass of material.

REM = unit to measure the amount of damage to human tissue from a dose of ionizing radiation

= dose in rad * Quality Factor

Quality factors: 1 for x, gamma, and beta

3 for thermal neutron;

20 for alpha and fast neutron

Dose equivalent = rem * QualFactor * DistribFactor

= compare total effects of irradiation received

Humans are naturally exposed to ~ 360 mrem per year from:

natural radon in soil/rocks (180 - 250 mrem)

cosmic radiation (25-50 mrem)

medical, internal radioactive elements in body, etc.

Drinking Water standards are gross 15 pCi/L, gross + = 4 mrem/yr

Drinking water standards for specific radionuclides, in pCi/L, are: 5 radium; 300 radon; 6 Sr; 20,000 Tritium; also 20 µg/L U.

AVERAGE drinking water exposure results in 0.244 mrem/yr.

The suggested EPA limit for radon exposure in air is 4 pCi/L air.

Measuring Radioactivity

Geiger-Muller counter = radiation enters counter tube, interacts with argon in the tube and ionizes the atoms causing an electrical discharge which is detected and counted electronically

Scintillation counter = radiation strikes a specially doped crystal of alkali-metal halide, causing it to emit light which is detected by a photocell and counted.

Transuranics in Soils

Transuranics are elements with an atomic number from 92 to 103. These elements are commonly man-made, being present naturally in only very small quantities. The most important transuranic element is plutonium, with other important elements including neptunium, americium, curium, and californium (also termed members of “Actinide Series”). These transuranic elements are formed by neutron capture reactions, starting generally from U-238. There are 16 common isotopes of plutonium, and Pu oxidizes rapidly in air to form PuO₂.

Sources of Transuranics in the environment include:

Global Fallout from nuclear weapons explosions in atmosphere; fallout occurs over time from the stratosphere, and serves as a continuous source of contamination.

Local Fallout - low level chronic releases from nuclear fuel processing

Accidental releases - from waste disposal facilities, and destruction of nuclear weapons

element	half-life, yr	element	half-life, yr	element	half-life, yr
Np-237	2.1E6	Pu-242	3.79E5	Cm-243	32
Np-239	0.0064	Pu-243		Cm-244	17.6
Pu-236	2.85	Am-241	458	Cm-245	9300
Pu-238	86.4	Am-242m	152	Cm-246	5500
Pu-239	2.44E4	Am-242	0.00182	Cm-247	
Pu-240	6580	Am-243	7950		
Pu-241	13.2	Cm-242	0.445		

RADIOACTIVE WASTES

Radioactive wastes are not regulated by EPA under CERCLA or Superfund, but are rather regulated by the Nuclear Regulatory Commission (NRC) under the Atomic Energy Act. Other regulations pertinent to radioactive wastes include:

Marine Protection, Research, and Sanctuaries Act of 1972

Uranium Mill Tailings Radiation Control Act of 1978 (about 170,000 tons of tailing waste are generated per year)

Low Level Radioactive Waste Policy Act of 1980

Nuclear Waste Policy Act of 1982, Amend. 1987

Low Level Radioactive Waste Policy Amendments Act (LLRWPA) 1985

Energy Policy Act of 1992

There are six classes of Radioactive Waste:

1. Low Level Wastes (LLW):

Contributors to low level radioactive wastes include: 50% from nuclear power plants, 30% from hospitals and research labs, 20% industry/other commercial and defense sectors. Low level wastes are defined as those wastes to do NOT fit into one of the other 5 waste categories, and contain less than 100 nCi/g. These wastes may be in the form of solids, liquids, or gases. It is estimated that approximately 57 million metric tons of LLW is generated in the U.S. per year. This was about 180,000 cubic meters per year in the 1980s, but due to recent efforts to minimize waste volumes via compaction this value has decreased by more than half. In 1993, only 23,000 cubic meters of LLW was disposed.

There are three classes of LLW based on the concentrations of radioactive elements present in the waste: A = lowest concentrations; B; C = highest concentrations. Each element (for example, Co-60, H-3, Cs-137, Ni-63, C-14, etc) has a maximum concentration limit in Ci/m³ which determines the classification of the waste. Disposal of these wastes regulated under the Low Level Radioactive Waste Policy Amendments Act (LLRWPA) 1985. There are currently only 2 locations in U.S. where LLW can be disposed (Richland, WA and Barnwell, SC). In the past, LLW were disposed by so-called "random placement" in shallow trenches without engineering liners or caps. Past commercial LLW disposal sites included Beatty, CA; Sheffield, IL; Maxey Flats, KY; West Valley, NY. However, in all these cases the non-engineered disposal of the waste allowed water seeping through to carry away the radioactive materials leaking out of drums and from the waste. LLW was also disposed at US DOE sites in Idaho (INEL), Hanford, WA; Nevada (NTS), Argonne Natl Lab IL, Oak Ridge, TN; Savannah River, SC; Los Alamos, NM.

Due to the regulatory mandate that states have a plan for LLW disposal, many states have formed "compacts" to use a single common waste disposal site. Currently there are 10 compacts active (number of states listed in parenthesis): NW (6), Midwest (8), Northeast (11), SE (7), Central (5), Rocky Mt (5), Texas (3), CA (1), Dakota (2). Some of these compacts have a site under review for licensing: Boyd County, NE (central), Hudspeth County, TX; Wake County, NC (SE), Ward Valley, CA (NW and RM compacts sending waste to Hanford). There are only 3 current, active licensed disposal sites are Barnwell, SC; Hanford, WA; and Clive, UT (Class A and mixed LLW only).

2. Uranium mining and milling waste:

Uranium mining and milling waste contain low concentrations of naturally occurring radioactive materials. These wastes are primarily solids and gases. Although there are no currently operating uranium mines in the U.S., in 1977 there were approximately 35 active mines (4 located in Colorado). In addition, there are 9 major reactor fuel processing and fabrication facilities in the U.S.

3. High level waste (HLW):

High level waste is generated from the reprocessing spent fuel from defense or commercial nuclear reactors. HLW emits highly penetrating radiation and most of the radioactive isotopes it contains have relatively short decay times. Prior to treatment, most of this waste is in liquid form. The storage repository planned at Yucca Mountain is being designed to accept HLW.

4. Spent fuel from nuclear reactors:

Spent nuclear reactor fuel includes waste products from fission, among which are some long-lived radioactive elements such as plutonium. The waste is typically in the form of solids and gases. Currently, there are about 111 nuclear power plants in U.S. fueled by uranium. The uranium in these reactors is in the form of small ceramic pellets inside long metal tubes. These tubes are replaced every 12 to 18 months, and the tubes removed are then “waste”. Currently, these spent fuel rods are stored on-site in liquid pools or dry casks. As of 1993, approximately 28,000 metric tons of spent nuclear fuel was stored on-site at the commercial nuclear power reactors; this amount was predicted to increase to 48,000 metric tons by 2003. (www.nrc.gov; Jan 1997)

5. Transuranic Waste (TRU)

Transuranic waste emits moderately penetrating radiation (including alpha particles) and has long decay times. The waste is typically in either solid or liquid form and contains elements heavier than uranium, which result from processing spent fuel from nuclear reactors. This waste is largely generated by the defense sector. The Waste Isolation Pilot Plant (WIPP) which is located in New Mexico is being designed to accept TRU wastes.

6. Mixed waste is any waste that has “hazardous” characteristics as defined under RCRA in addition to being radioactive. Currently, there is only one incinerator in the U.S. which treats mixed waste. These wastes are dually regulated under US Atomic Energy Act and RCRA. Due to inability to treat much of this waste, 24 million kg is currently being stored by DOE at Oak Ridge, in violation of RCRA.

Concerns with radioactive waste are primarily associated with radionuclides from Energy and Weapons use and research, and in particular wastes at Department of Energy (DOE) and Department of Defense (DOD) sites.

Radioactive Wastes

Thermal Treatment of Mixed Wastes

At the present time there is only one industrial boiler facility licensed to treat mixed liquid wastes in the U.S. This facility is located in Kingston, TN, and is owned and operated by Diversified Scientific Services, Inc. (DSSI) The plant produces electrical power from the burning of the waste and propane. Waste accepted can contain limited quantities of radioisotopes with atomic numbers 1 through 83, and americium, plutonium, radium, thorium, and uranium. The wastes accepted may be co-contaminated with chlorinated solvents, fluorinated hydrocarbons, CFCs, freon, and heavy metals. Effluent gas from the boiler is treated by spray quenching, baghouse, scrubber, and HEPA filters, with continuous monitoring of effluent radioactivity.

Recycling, Treatment, and Facilities at the Savannah River Site

The U.S. Department of Energy Savannah River Site (SRS) has both a tritium mission and a High Level Waste Division. Tritium reservoirs are one essential component within a nuclear weapon, and since tritium has a relatively short half-life of about 12 years, the reservoirs must be periodically replenished with fresh tritium to keep the weapons operational. As a result of the end of the Cold War era, the tritium mission also supports the downsizing of the US nuclear weapons stockpile. The HLW Division manages the 34 million gallons of liquid HLW stored in the tank farms at the SRS. This waste has been accumulating since the 1950s from the processing of nuclear materials for national defense, space, medical, and research programs. SRS has a specially designed process of in-tank precipitation (ITP) which removes the radionuclides from the liquid salt solution. This results in a small amount of highly radioactive precipitate and a decontaminated liquid (more than 90% of the total volume). The precipitate is processed by a washing/filtering step and is then vitrified into glass (melted at 2100°F, glass solidifies in 10' tall stainless steel canisters). The low-level radioactive liquid is stabilized in cement that is pumped into above-ground engineered vaults.

The SRS also stores spent nuclear fuel in water-filled basins. They carefully monitor the liquid to ensure that proper water chemistry is maintained, which aids in preventing corrosion of the basin.

Ocean Disposal

U.S. started “ocean disposal” of radioactive wastes in late 1944 with discharge from reactors at Hanford, WA, to the Pacific Ocean via the Columbia River. From 1946 to 1970, the U.S. disposed of radioactive waste at 35 dump sites located in the Atlantic Ocean, Pacific Ocean, and Gulf of Mexico. A total of approximately 34,000 containers of radioactive waste, containing an estimated 79,000 Ci of radioactivity, were dumped in the Atlantic Ocean. An additional 52,000 containers containing approximately 15,000 Ci of radioactivity were dumped in the Pacific Ocean. The practice of ocean disposal has been prohibited since the U.S. 1972 Marine Protection Act. Current radiological surveys in the ocean areas where wastes were disposed have found low concentrations of Cs-137, Pu-239, and Pu-240 in the skin and gastro-intestinal tract of some of the fish collected at the sites. In addition, there are some contributions to the total “radioactivity” deposited in the ocean due to sunken nuclear submarines (3 subs total containing 10 million to 1000 million curies each). However, no radioactivity release from these subs has been detected more than 10 years later. Further, ocean testing of nuclear weapons has contributed tens of millions of curies worth of fission products to the levels of radioactive elements in the oceans. Of European nations, both the United Kingdom and France dump several hundred thousand curies of radioactive elements per year into their coastal waters in from their nuclear fuel reprocessing facilities.

SPENT FUEL from nuclear power plants

In the U.S., this spent fuel is currently stored on-site, with storage at some locations since the 1950s. Spent fuel is stored in steel-lined concrete pools filled with water or in concrete or steel containers above ground (so-called “dry casks”). However, these sites are running out of space. Generation of spent fuel is on the order of 30 tons of used fuel per year per plant (about 3000 tons per year total in U.S.). This waste would occupy a total area similar to that of a football field with waste 5 yards deep.

Monitored Retrievable Storage (MRS)

central storage location for used nuclear fuel from power plants
 “monitored” continually by US NRC and local regulatory agencies
 “retrievable” used fuel is stored in MRS site temporarily until it is sent to a permanent underground repository (such as Yucca Mountain)

Spent fuel is shipped to the site via trucks or trains in heavy metal casks weighing 20 to 100 tons with steel walls 9-12 inches thick; casks designed to withstand collisions, punctures, fire, underwater.

on site, stored in large dry casks which are housed in massive steel+concrete vaults stored such that the steel, lead, iron, concrete absorb the radiation and it does not escape to the environment

radiation monitors in the fuel storage area will be monitored round the clock
 site on several hundred acres of land, with large buffer area of open land between the facility where the fuel is housed and the property line

access to the site strictly controlled, with high-security fence and security force

Considered the first step towards safe, permanent disposal of used fuel. The U.S. government was required by law to begin accepting used fuel in 1998, but a permanent facility won't be ready until AT LEAST 2010 or later!

Sub Sea-Bed Disposal

Sub Sea-Bed Disposal (SSBD) is one potential option for radioactive waste disposal. In SSBD, the wastes would be placed below the sea floor, which would use the clays of the seabed to isolate the HLW from the “environment”. The wastes would first be placed in sealed metal canisters, and these canisters would subsequently be buried 20-50 meters below the sea-bed at locations where the ocean floor is seismically stable (far from the edges of the continental plates). The potential use

of this waste disposal method is being evaluated by multi-nation consortia including nations with greater than 75% of the current worldwide nuclear power capacity (U.S., Europe, etc.)
A related web-site is: www.tinet.ch/odm01/welcome.html

Long-Term Storage Facilities (LTSF)

LTSF are intended to allow radioactive decay to harmless stable products, therefore storage for 1000s of years is needed.

Yucca Mountain is being considered as the site for permanent storage for used nuclear fuel. Yucca Mountain is located about 100 miles from Las Vegas, and is in a arid climate. The site is currently being studied to fully characterize the hydrogeology, air quality, background radiation, etc. For example, the a probabalistic volcanic hazard analysis was conducted and determined that the predicted risk of a volcanic center disrupting a nuclear waste repository at Yucca Mountain in the next 10,000 years is about 1 chance in 70 million per year. In addition, 40 groundwater monitoring wells are in place and 8 dust-monitoring remote towers. Data is also being gathered to aid in repository design and construction. This includes the excavation of an exploratory studies facility in the mountain. Excavation of the main drift of the tunnel was completed in July 1996, using a tunnel boring machine which had completed 20,643 ft of the 25,800 ft loop planned. Studies at Yucca Mountain have been on-going for over 10 years, starting in 1987, to determine if the site is suitable for the high-level nuclear waste repository. If built, the repository would be 1000 ft below the ground surface and over 800 ft above the water table in volcanic tuff, a very hard rock. The site would contain multiple barriers (both engineered and natural) to isolate the waste from the environment. It is hoped that a license application for operations at the site can be submitted to the Nuclear Regulatory Commission by 2002.

Waste Isolation Pilot Plant (WIPP) is a research and development facility of the DOE in SE New Mexico (near Carlsbad). It was designed to demonstrate the safe transport, handling, and disposal of defense-generated transuranic radioactive waste underground in deep salt beds. Wastes to be disposed includes mostly plutonium and may be mixed with lead, cleaning solvents. Salt beds would permanently encapsulate the waste, isolating it from the environment. The cost of developing this site was about \$155 to \$200 million per year.

After many years of research and development, the EPA certified WIPP as the first underground repository for TRU waste. To date, waste from Rocky Flats and Los Alamos National Laboratory have been sent to WIPP, with Idaho National Laboratory also certified to ship waste.

International

Many countries are investigating the disposal of radioactive wastes, and a number of locations are already permitted and/or in use. A summary of activities is provided in the table below.

Country	Nuclear Waste Activities
Slovenia	Krsko on-site temporary rad waste storage at Krsko nuclear power plant
Canada	LLW stored at Chalk River Laboratories
Spain	HLW from Trillo NPP stored in metal cask used for both transport and long term storage
Belgium	7 operational NPPs SCK-CEN nuclear research center Mol-Dessel site for HLW disposal 225-m deep in clay layer being investigated
Germany	Morsleben repository Konrad repository – 2002, for MOX fuel waste Gorleben repository – 2012, salt dome, HLW disposal
France	3 potential sites for deep rad waste disposal: 2 in sedimentary clay and 1 in crystalline rock
Norway	Himladen site for LLW and ILW

(number of growing seasons required). The costs associated with soil washing included: digging up the contaminated soil; mechanical mixing of the soil washing basin, chemical addition, liquid waste stream treatment, and residual solids disposal. (*Note: both of these methods are considered "innovative treatment technologies" by EPA, so costs were estimated.*) This study found that phytoremediation was one-third the cost of soil washing, in addition to "economy of scale" benefits with phytoremediation. It was also considered to be a conservative cost estimate for phytoremediation since low accumulation factors were assumed.

Activity	Cost Estimate, \$ Phytoremediation	Cost Estimate, Soil Washing
Site preparations	63,000	61,000
Production/processing	395,000	1,336,000
Waste mgmt/disposal	35,000	175,000
Project closeout	18,000	62,000
EH&S, design, proj mgmt	153,000	490,000
Overall Estimated Cost	664,000	2,124,000
Cost per metric ton soil	200	600

* total soil treated = 2,500 m³ @ 1,420 kg/m³

Biomagnetic separation: microbes form extracellular metal precipitate, then high gradient magnetic separation is used to remove metal-laden microbes from the treated matrix. This can be used for heavy metals and radionuclides, with either aerobic or anaerobic living cells. Non-viable, microbially produced magnetic adsorbent (metal salt) can be used in microbially hostile environments. The microbes can accumulate metal coating up to 4 times its own weight. Magnetic-separation removed >98% of microbes, and can treat low concentrations of radionuclides, 3 ppb, down to sub-ppb levels.

SITE EXAMPLE: HANFORD, WA

Environmental restoration budget: 1995 = \$232M, 1996 \$160M, 1997 \$100-150M

Problems include: CT contaminated groundwater, currently hot-spot containment wells

Cs, Sr along Columbia River, containment

Groundwater velocity ~0.5 to 1 ft/d movement; low organic content in soil, gravels

Classes of Waste

1. Tank Waste - instability (seismic) and leaking tanks, gas production, etc.
2. Sr and Cs capsules
3. TRU retrievable solids
4. TRU-contaminated soils

liquid wastes were directly discharged into the ground via trenches

Goal: prevent further migration of contaminated groundwater into the Columbia River
stabilize the unstable wastes for on-site storage until WIPP open....

Use of new technology - a fiber-optic sensor immediately detects U-238 and Sr-90 in soils

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