NATIONAL LOW-LEVEL WASTE MANAGEMENT PROGRAM RADIONUCLIDE REPORT SERIES[a]

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ABSTRACT

The National Low Level Waste Management Program at the Idaho National Engineering and Environmental Laboratory has published a report containing key information about selected radionuclides that are most likely to contribute significantly to the radiation exposures estimated from a performance assessment of a low-level radioactive waste (LLW) disposal facility. The information includes physical and chemical characteristics, production means, waste forms, behavior of the radionuclide in soils, plants, groundwater, and air, and biological effects in animals and humans. The radionuclides included in this study comprise all of the nuclides specifically listed in 10CFR61.55, Tables 1 and 2, ^{3}H, ^{14}C, ^{59}Ni, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{137}Cs, ^{241}Pu, and ^{242}Cm. Other key radionuclides addressed in the report include ^{237}Np, ^{238}U, ^{239}Pu, and ^{241}Am. This paper summarizes key information contained within this report.

INTRODUCTION

The National Low-Level Waste Management Program (NLLWMP) at the Idaho National Engineering and Environmental Laboratory (INEEL) assists the U.S. Department of Energy (DOE) in fulfilling its responsibilities under the Low-Level Radioactive Waste Policy Amendments Act of 1985. The NLLWMP assists the DOE by providing technical assistance to states and compact regions as they develop new commercial low-level radioactive waste (LLW) management and disposal systems.

Construction and operation of a low-level radioactive waste (LLW) disposal facility requires a license under 10 CFR 61.(1) The objective of the NLLWMP is to provide technical expertise, information, and other resources to states and compact regions in support of this. The NLLWMP maintains contact with state and compact region officials to identify and provide general and specific assistance as needed. One avenue of assistance through the NLLWMP is developing technical documents. The purpose of one such document "Selected Radionuclides Important to Low-Level Radioactive Waste Management(2) is to provide information to state representatives and developers of LLW management and disposal facilities and other concerned entities about the radiological, chemical, and physical characteristics of selected radionuclides and their behavior in the environment. Although such information has been available, it was widely scattered throughout numerous references and often written in a highly technical manner, not easily understandable by nontechnical reviewers of LLW facility performance assessments.

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Performance assessment analyses predict radionuclide release, transport, and resulting dose that may be delivered to individuals from such a facility. Radiation exposure can result from exposure to direct gamma rays from the wastes, from direct and indirect uses of surface and ground water that has filtered through the waste, and from atmospheric transport of radionuclides via breathing and deposition to the ground surface and plant surfaces. Computer codes are utilized in a performance assessment to estimate the effects from the processes that contribute to the radiation exposures to humans resulting from LLW disposal operations. These processes include: direct gamma ray exposure from the wastes, surface water infiltration into the waste, flow of water in the waste environment, engineered barrier and waste container degradation, leaching of radionuclides from waste forms, and transport of radionuclides in the waste environment, including groundwater transport, surface water transport, and atmospheric transport of radionuclides from disposal units.

To provide background information in a readily understandable, concise format, a series of reports was published by the NLLWMP, documenting information about the radiological, chemical, and physical characteristics of each selected radionuclide and its expected behavior in the disposal facility environment. These reports have provided useful background information for people writing performance assessments and license applications for commercial LLW sites as well as for people who respond to comments received in public meetings on siting issues. After the first few individual reports were issued, various state personnel contacted the NLLWMP to request additional reports be written on other nuclides. Because of the high interest expressed in these reports, Reference 2 was written to combine these individual reports into a single, comprehensive, easy to read document. This document has been issued, by request, to several hundred people.

Reference (2) includes references to the individual reports. The radionuclides included in this study comprise all of the nuclides specifically listed in 10CFR61.55, Tables 1 and 2, ^{3}H, ^{14}C, ^{59}Ni, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{137}Cs, ^{241}Pu, and ^{242}Cm. The following additional radionuclides, not specifically listed in Tables 1 and 2, were also included in this series, ^{237}Np, ^{238}U, ^{239}Pu, and ^{241}Am. Together, these radionuclides are those that are judged by the NRC to most likely contribute significantly to the radiation exposures estimated from a performance assessment of a proposed commercial LLW disposal facility

The following paragraphs summarize key information for each radionuclide as addressed in the comprehensive report.(2) The interested reader is directed to this report for additional details and to reference the original nuclide reports. Table I contains the half-life and principal radiation(s) associated with each nuclide.

TECHNETIUM-99

^{99}Tc is produced by the fission of uranium and plutonium and is not a naturally occurring radionuclide. Reactor wastes such as evaporator bottoms, filters, and sludges may contain ^{99}Tc. Small amounts of ^{99}Tc can also be present in LLW generated at medical laboratories and academic institutions. The predominant form of ^{99}Tc at an LLW disposal facility is the

Table I: Radiation properties

Nuclide	**Half-life**	**Principal radiation(s)**
Tc-99	2.13E5 y	0.293 MeV (β)
C-14	5.73E3 y	0.156 MeV (β)
I-129	1.6E7 y	0.15MeV (β)
H-3	12.3 y	0.019 MeV (β)
Cs-137	30.17 y	0.662 MeV (γ)
Sr-90	29.1 y	0.546 MeV (β) + 2.28 MeV (β) [daughter decay]
Ni-59	7.6E3 y	1.06 MeV (x)
Ni-63	100 y	0.659 MeV (β)
Pu-239	2.44E4 y	5.2 MeV (α)
Pu-241	14.4 y	0.021 MeV (β) + 5.49 MeV (α) [daughter decay]
Nb-94	2E4 y	0.47 MeV (β) + 0.703, 0.871 MeV (γ)
Co-60	5.272 y	1.17, 1.33 MeV (γ)
Cm-242	163 d	6.11, 6.07 MeV (α) + 5.5 MeV (α) [daughter]
Am-241	432 y	MeV (γ)
U-238	4.47E9 y	4.15, 4.2 MeV (α)
Np-237	2.14E6 y	4.78 MeV (α)

pertechnetate ion (TcO_4^-). Because the pertechnetate ion is highly mobile in groundwater and because ^{99}Tc has a long half-life, its disposal presents a potential long-term hazard to the public. However, the pertechnetate ion can be reduced to less-soluble forms of technetium that are not mobile in environmental media. The NRC, in its draft Environmental Impact Statement (EIS) (3) on 10 CFR 61, identified ^{99}Tc as one of four radionuclides (tritium, ^{14}C, ^{99}Tc, and ^{129}I) that will require site-specific considerations to ensure that performance objectives for long-term environmental protection are met for disposal of commercial LLW. Because ^{99}Tc is a low energy beta emitter, it poses a greater internal than external hazard to humans. The main route ^{99}Tc can enter the human body is ingestion.

CARBON-14

The principal means of producing ^{14}C is by neutron activation of stable ^{14}N and stable ^{17}O, in a nuclear reactor. The fission process of the reactor supplies the large source of neutrons necessary to induce the neutron activation. Before the advent of manmade disruptions of the ^{14}C atmospheric concentration (burning of fossil fuels and atmospheric testing of nuclear weapons), this nuclide was produced, at a relatively constant rate, by cosmic rays impinging on the atmosphere. LLW that contains ^{14}C includes activated metals from reactors, sealed sources, and animal carcasses from research laboratories. It is usually present in chemical forms, such as carbonates, than can be quite mobile in groundwater systems. Another concern is that ^{14}C, in the form of CO, or CO_2, is very volatile. Because ^{14}C is a low-energy beta emitter, it poses a greater internal than external hazard to humans. The main route that ^{14}C can enter the human body is by either inhalation or ingestion.

IODINE-129

^{129}I is produced by uranium fission primarily in nuclear power reactors, and is not a naturally occurring radionuclide. ^{129}I is a long-lived radionuclide present in LLW generated at nuclear power plants. LLW that contains ^{129}I includes ion-exchange resins, concentrated liquids (e.g. from reactor coolant leaks), filter sludge, cartridge filters, and trash. Disposing of ^{129}I-contaminated waste at LLW disposal facilities is a potential concern because ^{129}I often exists in chemical forms that are highly mobile in groundwater. Because ^{129}I is primarily a low-energy beta emitter, it poses a greater internal than external hazard to humans. The main route that ^{129}I can enter the human body is ingestion. Whether inhaled or ingested, most of the radioactive iodine will dissolve in the body fluids. When dissolved, iodine travels and concentrates in the thyroid. If present in sufficient concentrations, the low-energy beta particle radiating from this nuclide may irradiate the thyroid to the point of inducing cancerous thyroid nodules.

TRITIUM

Tritium (^3H) is a radioactive isotope of hydrogen. It is naturally produced in very small quantities in the atmosphere. Because of its very small naturally occurring concentration, tritium exists, for all practical purposes, as a manmade isotope. Tritium is primarily used in industrial thickness gauges, luminous paints, nonpowered (self-luminous) light sources, fusion research, thermonuclear weapons, and as a radioactive tracer in chemical and biological experiments. Tritium decays to ^3He with the emission of a beta particle but no gamma ray. Tritium produces a very-low-energy beta particle and is usually considered one of the least hazardous radionuclides. However, since it can replace normal hydrogen in water and other chemical compounds essential for life, tritium poses a unique hazard that can be very mobile within the physical and biological environment, including the human body. Many waste types disposed of in LLW disposal

facilities contain tritium. These waste types include reactor evaporator bottoms and institutional and academic research laboratory trash. Tritium is very mobile in groundwater and surface water systems.

CESIUM-137

^{137}Cs is produced by the fission of uranium and is not a naturally occurring radionuclide. The largest source of ^{137}Cs and potential waste material is from nuclear reactor operations. The gamma-ray of ^{137}Cs has been used to sterilize medical supplies, milk cartons, and irradiate food. Industrial applications of ^{137}Cs include the production of plastic shrink tubing (irradiated plastic has the tendency to shrink after being heated), radiography to inspect metal castings and welds for flaws and material defects (e.g., cracks in steel pipes), radioactive measurement gauges for liquid or solid thicknesses (e.g., gauging of automobile sheet steel), treatment of sewage sludge to kill bacteria and viruses, and radiotherapy to kill cancerous tissue. The quantity of ^{137}Cs waste material generated in medical, academic, or commercial facilities is small compared with the quantity of ^{137}Cs waste produced as a result of nuclear reactor facilities. Ion-exchange resins used to purify coolant water in a nuclear power plant frequently contain large amounts of ^{137}Cs. The common chemical compounds of ^{137}Cs are water soluble and will readily move with groundwater unless preferentially retained in the soils, particularly by clay soils. Absorption of cesium by humans takes place primarily through the digestive tract. Cesium and potassium have similar chemical properties because both are alkali metals. Potassium serves a very important function in the body as an electrolyte, and one potential effect of ^{137}Cs is on the potassium levels in the body in addition to its potential radiation effects on the various body tissues.

STRONTIUM-90

^{90}Sr is not a naturally occurring radionuclide. The main source of ^{90}Sr is from fission product recovery. ^{90}Sr emits a high-energy beta particle following decay. Almost no gamma-ray particles are released from the ^{90}Sr decay sequence. Due to the high beta particle energies of ^{90}Sr and its daughter nuclide ^{90}Y, ^{90}Sr has been used in industrial applications to measure thicknesses of paper, plastic, rubber, and metal foils. It also has some medical applications such as treatment for some eye and skin diseases. The largest source of strontium, and the largest potential source of waste material, is the inventory of ^{90}Sr stored in commercial spent nuclear fuel. Most of the LLW generated at nuclear power plants will contain some ^{90}Sr; however, other radionuclides (e.g., ^{137}Cs) are usually more prevalent and generally determine the waste characterization. Wastes that contain ^{90}Sr include both wet and dry wastes such as spent ion-exchange resins, filter sludge, filter cartridges, evaporator bottoms, compactible and non-compactible trash, and irradiated components. A smaller fraction of wastes containing ^{90}Sr are generated from industrial, institutional, and medical applications. Strontium is very soluble and is transported readily with precipitation and groundwater deep into soils. As such, it can be taken up by plants through the roots, which is the principal means by which strontium gets into the food chain. Once ^{90}Sr is ingested, it concentrates in bone tissue. Since these areas are near blood-producing bone marrow, one of the principal health hazards associated with ^{90}Sr contamination of humans is reduction in the blood platelet production. Other hazards include the possibility of bone cancer.

NICKEL-59 AND NICKEL-63

Neither ^{59}Ni nor ^{63}Ni is a naturally occurring radionuclide. They are produced by neutron activation of ^{58}Ni and ^{62}Ni (both naturally occurring and stable isotopes of nickel) in the structural steels and internal components of nuclear reactor vessels. Nickel metal is resistant to attack by water or air and, therefore, is often used as a protective coating for other metals or as an alloy to create a corrosion-resistant metal such as stainless steel and Inconel. Limited amounts of these nickel isotopes can enter the environment through operational wastes from a nuclear reactor (i.e., corrosion of stainless steel surfaces and accompanying release of nickel into circulating coolant). ^{59}Ni decays by electron capture. ^{63}Ni decays by beta decay and is primarily a hazard for people who inhale it. The major radiological concern with these two nuclides is related to limiting the exposure to people who are decommissioning and dismantling reactors, primarily for reactors in service for more than 30 years. At that point, these two nuclides represent important activation products and will exceed the activity from the other major activation products: ^{59}Fe and ^{60}Co. Neither nickel isotope is commonly used for medical or industrial purposes. Nickel tends to be easily absorbed by soils. However, the ability of soils to absorb nickel out of groundwater varies widely, depending on soil pH and soil type.

PLUTONIUM-239 AND PLUTONIUM-241

Neither ^{241}Pu nor ^{239}Pu is a naturally occurring radionuclide. ^{241}Pu is produced within nuclear reactors via neutron absorption by ^{240}Pu or through multiple neutron absorptions and decays of other transuranic isotopes. ^{239}Pu is produced in nuclear reactors via neutron absorption by ^{238}U (the principal naturally-occurring isotope of uranium) and subsequent beta decay. ^{239}Pu is used extensively as nuclear weapons material as well as an alternate fuel for nuclear reactors. ^{241}Pu decays primarily by beta particle emission to ^{241}Am. However, a very small fraction decays to ^{237}U. The released radiation is less penetrating than that of most radionuclides, and in particular is comparable with that of tritium. Both plutonium isotopes concentrate in bone tissue once they enter the human body. Both plutonium isotopes are classified in the very high radiological hazard group, (^{241}Pu because of its principal daughter product, ^{241}Am, a high-energy alpha particle emitter, and ^{239}Pu because of the high-energy alpha particles emitted by its decay). Generally, plutonium waste ends up as transuranic or high-level waste; however, small amounts of plutonium can exist in LLW, due to various waste streams from nuclear reactor operations. Few medical or industrial uses of ^{241}Pu are known. Most medical and industrial uses of plutonium center on ^{238}Pu for cardiac pacemakers and radioisotopic thermoelectric generators. $^{238,239.240}$Pu isotopes are also used in biological studies. Plutonium tends to be relatively immobile in soil and its principal radiological hazard for humans occurs when plutonium-contaminated soil is resuspended in air and subsequently inhaled.

NIOBIUM-94

^{94}Nb, an actinide, is produced from the neutron activation of ^{93}Nb, the only stable isotope of niobium. ^{94}Nb is not a naturally occurring nuclide. ^{94}Nb, like ^{59}Ni and ^{63}Ni, is found in metallic reactor components. The major concern with ^{94}Nb is in limiting the exposure that people receive who decommission and dismantle reactors, primarily reactors that have been in service for more than 30 years. If decommissioning is delayed for a long time after reactor shutdown, this nuclide would provide the dominant, long-term source of gamma radiation from these components. This nuclide can contaminate LLW waste through corrosion of stainless steel and Inconel surfaces in nuclear power plants and subsequent discharge with the primary coolant. ^{94}Nb is commercially available and has been used as an external source of gamma rays for

laboratory use. Few medical uses for this nuclide have been identified. In general, niobium is easily adsorbed by soils and is not easily leached into the groundwater (it will not migrate appreciably from the original site). When mammals ingest this nuclide, most is excreted within a few days. The fraction that remains is generally uniformly distributed in the body.

COBALT-60

^{60}Co is produced in the structural steels and other alloys of nuclear reactor vessels and internal components from neutron activation of ^{59}Co, the only stable isotope of cobalt. ^{60}Co is not a naturally occurring radionuclide. ^{60}Co decays to minimal activity levels within 50 years of generation. However, ^{60}Co poses an external exposure hazard. Because of its high-energy gamma radiation, ^{60}Co has been used extensively as a radiation source for both commercial and medical uses. Users include the transportation industry, which uses this nuclide to measure the thickness of automobile sheet steel. The metals industry uses ^{60}Co as a radiography source to detect flaws in welded joints and casting. The chemical and medical industries use the nuclide as a catalyst in flow studies, drug-metabolism studies, and sterilization of medical supplies. This nuclide is used extensively as an irradiation source for treatment of cancer in humans. Waste reactor components are the primary LLW material containing ^{60}Co. In general, cobalt is easily adsorbed by soils and is not easily leached into the groundwater. The main concern surrounding ^{60}Co is in limiting the exposure received by persons handling sealed sources containing this isotope or associated with the decommissioning and dismantling of reactors.

CURIUM-242

Multiple neutron absorption of nuclear fuel constituents such as ^{238}U and ^{239}Pu produces ^{242}Cm. Small amounts of ^{242}Cm can enter the primary coolant of nuclear reactors and be removed by filters and/or cleanup resins. ^{242}Cm is important in LLW disposal primarily because of the radiological daughters produced through the ^{242}Cm decay scheme. Although ^{242}Cm decays with a relatively short half-life, its radiological daughters, such as ^{238}Pu with a half-life of 86 years, can persist for much longer in radioactive waste. There are some commercial applications of ^{242}Cm. For example, this nuclide has been used as the power source for an isotopic power reactor. There are few medical uses for this nuclide. In general, curium is easily adsorbed by soils and is not easily leached into the groundwater. The principal radiological hazard associated with this nuclide is due to its high-energy alpha particles. The principal risk is due to inhalation of this nuclide. ^{242}Cm can decay and concentrate in bone tissue once it enters the human body.

AMERICIUM-241

^{241}Pu, produced by neutron absorption of ^{238}U and ^{239}Pu, decays to ^{241}Am. The decay process also produces gamma-rays and x-rays. ^{241}Am enters the environment from the activities related with reactor operations and decommissioning, atomic weapons production, and when sealed sources containing ^{241}Am are manufactured, used, and disposed. One major application of ^{241}Am as a radioactive sealed source is in smoke detectors (e.g. the radiation from ^{241}Am is used to detect the presence of smoke or heat sources). The regulatory limits for ^{241}Am disposal and personnel exposure are very restrictive. Relatively small concentrations of ^{241}Am can become a major contributor to the calculated doses to individuals in performance assessment calculations of an accidental release involving this radionuclide. This is primarily due to the long half-life and high energies of the alpha particles emitted during the decay process. ^{241}Am is not very mobile in soils and only small quantities can be expected to migrate within the soil to the biosphere. As stated, ^{241}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.

URANIUM-238

^{238}U is a naturally occurring radioactive nuclide. ^{238}U is the most abundant isotope of uranium, comprising >90% of all naturally occurring uranium. Another isotope of this element, ^{235}U, is used widely in energy and weapons production, and ^{238}U is a byproduct of the ^{235}U enrichment process. Therefore, this nuclide is often equated with the term "depleted uranium." As such, this nuclide has widespread industrial and military usage, including shielding for radioactivity, counterweights, vehicle armor, and weapons. Moderately high-energy alpha particles, low-energy gamma rays, and low-energy beta particles are emitted when this nuclide decays. Because of the long half-life of ^{238}U, it is not, itself, a major factor in the ability of an LLW disposal facility to meet performance objectives. The presence of the nuclide, however, can be relatively significant in assessing the long-term performance of such a site due to the quantity, radiotoxicity, and mobility of its daughter products, which include isotopes of radium and radon. The principal radiological hazard associated with this nuclide is due to its relatively high energy alpha particles. Direct exposure from ^{238}U is negligible. The principal risk is due to either inhalation or ingestion.

NEPTUNIUM-237

^{237}Np is principally produced in nuclear reactor fuel rods from the bombardment of ^{238}U with neutrons. Some naturally occurring ^{237}Np is produced when natural uranium ore is bombarded with neutrons generated as a result of spontaneous fission. Low-energy gamma rays and low-energy electrons are also emitted when this nuclide decays. Because of the long half-life (and consequent low specific activity) of ^{237}Np, it is not itself a major factor in the ability of an LLW disposal facility to meet performance objectives. The presence of the nuclide, however, can be relatively significant in assessing the long-term performance of such a site due to the quantity, radiotoxicity, and mobility of its daughter products, which include isotopes of radium and radon. Because this nuclide is present in the spent fuel of nuclear power plants, some contamination of the reactor coolant system is possible. After irradiation, the fission and activation products can be transported with the coolant to other parts, filters, etc. of the reactor. Through operational wastes and decommissioning, small amounts of these radioactive nuclides, including ^{237}Np, will be shipped to LLW Sites for disposal. This nuclide has very few commercial applications. Neptunium is more soluble than other actinides and tends to remain in the groundwater rather than being adsorbed by the soil. The principal radiological hazard associated with this nuclide is internal due to its relatively high-energy alpha particles. The principal risk is due to either inhalation or ingestion.

SUMMARY

In summary, general information for sixteen key radionuclides in LLW has been presented, including the sources of these radionuclides, their interaction with the environment, and hazards to humans. These nuclides have been designated as being significant with respect to the licensing and operation of commercial LLW sites, including calculations of potential contamination and exposure of humans to radiation resulting from these sites. More details regarding these nuclides are available in Reference (2).

The behavior of radionuclides and their interaction within the disposal facility environment is important information used by operators of disposal facilities, individuals that site and license new facilities, and regulators of radioactive waste disposal facilities to protect the environment and public from contamination and exposure to radiation. Operators, siting contractors, regulators and the public have found Reference (2) useful in helping them to communicate the significant factors related to radionuclides in commercial LLW disposal facilities. This report is written in a simple to read, overview fashion that individuals with limited technical experience can understand. For detailed information, the reader is encouraged to refer to the listed references.

REFERENCES

1 Title 10 Code of Federal Regulations, Part 61, *Licensing Requirements for Land Disposal of Radioactive Waste*, last revision January 1, 1998.

2 *Selected Radionuclides Important to Low-Level Radioactive Waste Management*, DOE/LLW-238, November 1996.

3 U.S. Nuclear Regulatory Commission, *Licensing Requirements for land Disposal of Radioactive Waste*, NUREG-0782, Volume 1, September 1981.