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Volume 10: Nickel-63

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DISCLAIMER

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Volume 10: Nickel-63

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ABSTRACT

This report, Volume 10 of the National Low-Level Waste Management Program Radionuclide Report Series, discusses the radiological, chemical, and physical characteristics of nickel-63 (^{63}Ni). This report also discusses waste types and forms in which ^{63}Ni can be found and ^{63}Ni behavior in the environment and in the human body.

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 and chemical 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, and plutonium-241. These radionuclides contribute significantly to the dose estimated during a performance assessment analysis.

This report is Volume 10 of the series. It outlines the basic radiological, chemical, and physical characteristics of nickel-63, waste types and forms that contain it, and its behavior in environmental media such as soils, plants, groundwater, air, animals, and in the human body. Additional reports will be generated for other radionuclides.

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National Low-Level Waste Management Program Radionuclide Report Series

Volume 10: Nickel-63

INTRODUCTION

This report outlines the basic radiological, chemical, and physical characteristics of nickel-63 (^{63}Ni) and examines how these characteristics affect the behavior of ^{63}Ni in various environmental media, such as soils, groundwater, plants, animals, the atmosphere, and the human body. Discussions also include methods of ^{63}Ni production, waste types, and waste forms that contain ^{63}Ni.

All nickel atoms contain 28 protons (Z=28) and various numbers of neutrons (typically N=28 to 38 neutrons) within the atom's nucleus. Five stable isotopes of nickel exist, namely ^{58}Ni, ^{60}Ni, ^{61}Ni, ^{62}Ni, and ^{64}Ni. Their average naturally occurring abundances are 68.3% (^{58}Ni), 26.1% (^{60}Ni), 1.1% (^{61}Ni), 3.6% (^{62}Ni), and 0.9% (^{64}Ni). All other nickel isotopes, including ^{63}Ni, are radioactive, with ^{63}Ni having a 100 year half-life. The radioactive nuclides of nickel are not a normal constituent of the natural environment and are generated as a result of human activities.

The primary source of ^{63}Ni in the environment has been low-level radioactive waste material generated as a result of neutron activation of stable ^{62}Ni that is present in the structural components of nuclear reactor vessels. ^{63}Ni enters the environment from the dismantling activities associated with nuclear reactor decommissioning. However, small amounts of ^{63}Ni have been detected in the environment following the testing of thermonuclear weapons in the South Pacific.[1] Concentrations as high as 2.7 Bq[a] per gram of sample (or equivalently 0.0022 parts per billion) were observed on Bikini Atoll (May 1954). ^{63}Ni was not created as a fission product species (e.g., from ^{235}U or ^{239}Pu fissions), but instead was produced as a result of neutron capture in ^{62}Ni, a common nickel isotope present in the stainless steel components of nuclear weapons (e.g., stainless-304 contains ~9% total Ni or ~0.3% ^{62}Ni).

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a. One becquerel or 1 Bq is defined as 1 disintegration/second, and 1 curie (1 Ci) is defined as 3.7×10^{10} Bq.

RADIOLOGICAL CHARACTERISTICS

The half-life ($T_{1/2}$) of ^{63}Ni has been difficult to estimate accurately. Historically, it has been reported anywhere from 61 to 300 years.[2,3,4] The most recent and best available information concerning ^{63}Ni reports a half-life of 100 years.[5] ^{63}Ni is formed via neutron capture (n,γ) of ^{62}Ni (e.g., $^{62}Ni + n \rightarrow {}^{63}Ni + \gamma$-ray). ^{63}Ni undergoes radioactive decay via negative beta particle (β^-) emission (i.e., 100% of all decays result in an electron being emitted from the nucleus) leading to the creation of a stable copper-63 (^{63}Cu) nucleus.

The maximum beta particle energy is 67 keV (thousand-electron volts).[1,2] However, the average beta particle energy is only 17 keV per ^{63}Ni disintegration.[6] Since the released beta particle is usually emitted with an energy less than its maximum theoretical (e.g., 67 keV for ^{63}Ni decay), an antineutrino particle is simultaneously emitted and carries off the energy difference between 67 keV and that of the released beta particle. Consequently, beta particles are emitted with a continuous energy spectrum ranging from 0 to 67 keV. Since neutrinos (or antineutrinos) very rarely interact with matter, they are not considered radiologically important. Therefore, the antineutrino particle is usually not shown in the overall decay equation.

The decay sequence for ^{63}Ni showing the emission of a negative beta particle is illustrated in the following nuclear transformation:

$$^{63}Ni \xrightarrow[100\ y]{100\%} {}^{63}Cu + \beta^- \text{ (67 keV max energy)} \qquad (1)$$

The probability that a neutron passing through nickel will be absorbed by a ^{62}Ni nucleus is very small. However, due to the large amount of ^{62}Ni in a nuclear reactor vessel and the relatively long half-life of ^{63}Ni, the total amount of this radioactive nuclide present in the vessel will continue to increase during the reactor lifetime. By the time the reactor is decommissioned, a significant amount of ^{63}Ni will be generated and will pose special problems in the decommissioning of the reactor. Although the particle emissions from ^{63}Ni decay are <u>not</u> very energetic, large quantities of ^{63}Ni can pose significant radiological hazards. For example, under some conditions, ^{63}Ni can contribute a large fraction of the total radioactive dose received by persons involved in the dismantling of nuclear reactors. This situation can occur if the reactor has been operated for a long time (e.g., >30 years). In other words, ^{63}Ni does not normally represent a major concern in shielding or waste classification, but can be an important contributor to the overall radioactivity from activation nuclides and can affect the classification of certain reactor hardware as low-level radioactive waste material.

The 67 keV maximum energy beta particle emitted by the ^{63}Ni nucleus is weak. Table 1 lists the estimated maximum ranges for a 17 keV (average energy) beta particle emitted from ^{63}Ni decay. Note that the maximum particle ranges are small, and because it requires at least 70 keV for an electron to penetrate the protective layer of human skin, the ^{63}Ni beta particles lack sufficient energy to penetrate the 0.07 mm (dead layer) of human skin. ^{63}Ni is primarily considered a hazard internally and to the unshielded eye.

Table 2 lists the radiotoxicity of several important radionuclides, and Table 3 compares the average and maximum electron energies associated with ^{63}Ni decay with other well-known electron (beta particle) emitters. Notice that ^{63}Ni is shown in the same radiological group as carbon-14 (^{14}C) and cesium-137 (^{137}Cs); however, based on the information presented in Table 3, ^{63}Ni emits a beta-particle of smaller energy than either ^{14}C or ^{137}Cs.

Table 1. Comparison of the estimated maximum ranges for a 17.0 keV average energy beta particle emitted from ^{63}Ni decay for various materials.

Material (description)	Estimated maximum range for a 17.0 keV β^- (mm)[a]
Air (1 atmosphere pressure)	15.0
Water	0.020[b]
Plastic (Lucite)	0.014
Concrete	0.09
Aluminum or glass	0.008
Iron	0.0023
Lead	0.0015

a. Estimates are based on extrapolations of the maximum beta particle range data shown in Reference 7 or Reference 8 as a function of energy and materials.

b. An alternate estimate for the maximum range of a 17 keV beta particle in water is 0.04 mm, about twice the value listed in this table. This estimate is based on the observation that beta particles emitted by ^{241}Pu of energy 21 keV cannot penetrate more than 0.002 inches (0.051 mm) of water.[9]

Table 2. Comparison of the radiotoxicity of several important radionuclides (obtained from Appendix 2 of Reference 10).

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

Table 3. Average and maximum kinetic energies of beta particles and Auger electrons released during the decay of several important radionuclides. (Information compiled from data presented in References 1, 2, 5, 11, 12, 13.)

Radionuclide	Released electron energy	
	Average Energy (keV)[a]	Maximum Energy (keV)
Nickel-59 (^{59}Ni)	4.1[b]	~7.7[b]
Tritium (^{3}H)	5.7	19.0
Nickel-63 (^{63}Ni)	17.1	67.0
Iodine-129 (^{129}I)	40.0	150.0
Carbon-14 (^{14}C)	49.0	156.0
Technetium-99 (^{99}Tc)	85.0	293.0
Iodine-131 (^{131}I)	180.0	806.0[c]
Cesium-137 (^{137}Cs)	195.0	1176.0
Potassium-40 (^{40}K)	541.0	1330.0
Phosphorous-32 (^{32}P)	694.0	1710.0

a. 1000 keV = 1 MeV (million-electron volt). Beta particle energy unless noted.

b. The data for ^{59}Ni represent Auger electrons and not electrons emitted from the nucleus (i.e., beta particles). The maximum electron energy was estimated based on the assumption that an electron from the cobalt-59 (^{59}Co) atom (e.g., the daughter product from ^{59}Ni decay) absorbs a maximum energy x-ray.

c. 90.4% of the beta particle intensity for ^{131}I occurs at 606 keV, and only 0.6% occurs at 806 keV.

CHEMICAL AND PHYSICAL CHARACTERISTICS

Nickel is a silver-white metal with metallic properties. It has high electrical and thermal conductivities and can be drawn, rolled, forged, and polished. It is ferromagnetic, though not as strongly as iron. The finely divided metal is reactive to air and may be pyrophoric under some conditions.[14]

Nickel occurs in nature primarily in combination with arsenic, antimony, and sulfur. The largest nickel deposits are garnierite, a magnesium-nickel silicate, and certain varieties of pyrrhotite, an iron-sulfur compound that can contain 3 to 5% nickel.[14] The center of the earth is believed to contain considerable quantities of iron and nickel.

Nickel shares some chemical characteristics with its two neighboring elements in the periodic table, iron and cobalt. Palladium and platinum are in the same chemical family as nickel, although they are not as chemically active as iron and cobalt. The most abundant oxidation state for nickel is Ni^{2+}. Nickel may be oxidized to the +3 and +4 states, or reduced to the +1 state, in the presence of a suitable complexing agent (nickel can even form compounds as Ni^0), but such reactions are seldom carried out. The melting and boiling points for nickel are 1453 and 2732 degrees Celsius (°C), respectively.[15] The oxidation potential of nickel (to remove two electrons from an atom) is 0.25 eV. The metal dissolves readily in dilute mineral acids, but does not dissolve in concentrated nitric acid (similar to iron) because it is rendered passive by this reagent. Nickel is quite resistant to attack by air or water at ordinary temperatures (when compact) and is often electroplated on other metals as a protective coating. Table 4 summarizes some of the more important chemical and physical properties of nickel, extracted from various references.[16,17,18]

Nickel readily reacts to form compounds with oxygen (e.g., NiO), sulfur (e.g., NiS, $NiSO_4$), and the halogens (e.g., NiF_2, $NiCl_2$, and NiI_2). In addition, at high temperatures, nickel forms compounds with hydrogen, boron, carbon, nitrogen, silicon, and phosphorus. Nickel is important as a constituent in a wide variety of corrosion-resistant metal alloys including stainless steels, Inconels, and Monel.

The majority of anhydrous nickel salts are yellow (e.g., $NiBr_2$ is yellow), although several compounds exhibit other colors: $Ni(SCN)_2$ is brown, and NiI_2 is black. The colors of the familiar Ni^{2+} solutions used as laboratory reagents are not representative of the colors of the salts from which they are derived. Nickel forms an interesting variety of chemical complexes. Some of the most familiar of these are $Ni(H_2O)_6^{2+}$, and $Ni(NH_3)_6^{2+}$, which are both octahedral in structure.

Table 4. Chemical and physical properties of nickel. Obtained from References 16, 17, and 18.

Physical property (units)	Nickel data
Melting point (degrees Kelvin)[a]	1,726.0
Boiling point (degrees Kelvin)[a]	3,005.0
Heat of fusion (cal/g)	4,323.0
Heat of vaporization (cal/g)	90,800.0
Solid density (g/cm^3)	8.9
Oxidation potential (V)[b]	0.25
Ionization potential (eV)	7.635
Crystal form	face-centered-cubic

a. Note that zero degrees Kelvin equals -273.15°C and -459.67°F.

b. For $Ni = Ni^{2+} + 2e^-$.

NICKEL-63 PRODUCTION

The structural materials inside the reactor vessel of a nuclear power plant are exposed to neutron radiation. This radiation can cause many components of these materials to become radioactive with time via neutron activation. Those elements most susceptible to neutron activation are iron, nickel, and cobalt, found primarily in stainless steel and other important alloys (e.g., Inconel). Depending upon the amount of time these materials are irradiated and the integrated neutron flux, any of these nuclides may become the critical activation product affecting the dismantling activities of the reactor.

The principal means of producing ^{63}Ni is through neutron capture of ^{62}Ni, a stable isotope of nickel that exists in stainless steels and other nickel alloys. Nuclear fission does not play a significant role in producing ^{63}Ni. This process can be seen in Figure 1 where the ^{63}Ni position is shown with respect to the fission product yield distribution for thermal neutron fission of ^{235}U. The cumulative fission yield of ^{63}Ni from fission of ^{235}U atoms is essentially zero. This information was obtained from the fission yield data from the ORIGEN2[19] computer code cross-section data.

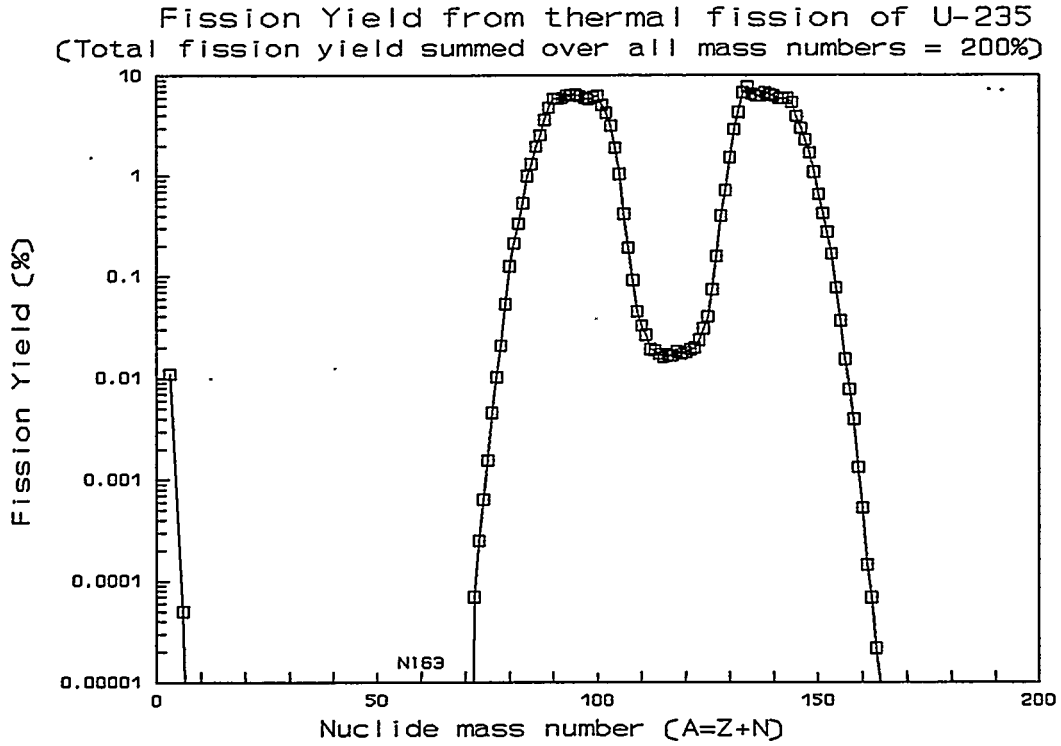

Figure 1. Fission product yield curve for thermal fission of ^{235}U. Note that the fission yield for ^{63}Ni should appear at the mass number of 63 (indicated by the "Ni63" position in the figure); however, the yield curve falls below the smallest displayed value (e.g., 0.00001%) shown for mass number 63.

Because ^{62}Ni atoms do not readily absorb thermal neutrons, and because ^{62}Ni comprises only a small fraction of the stainless steel in a nuclear reactor (~3.6% of the total nickel mass), it generally takes many years of neutron irradiation to produce a significant quantity of ^{63}Ni. However, because ^{63}Ni has a half-life that is longer than the normal life of a nuclear reactor (e.g., 100 years versus ~30 years for most reactors), nearly all of the ^{63}Ni that is produced within the reactor structure remains with the reactor until it is dismantled. A small fraction of ^{63}Ni can be released through corrosion of structural components and transport to other sites via the reactor coolant system. In other words, the ^{63}Ni inventory increases linearly with reactor operation and does not significantly decrease with time. Figure 2 shows the buildup of several different activation products in a pressurized water reactor vessel, including ^{63}Ni, ^{59}Ni, ^{94}Nb, and ^{60}Co, as a function of reactor operation.

^{63}Ni has not been considered a controlling radionuclide in nuclear reactor decommissioning until recently. This change has occurred because the periods of reactor operation have not been long enough to generate significant quantities of this nuclide. Figure 2 shows the relative importance of several radioactive activation products as a function of reactor operation. Reference 20 reported that if the internal components of a nuclear reactor are removed and shipped to licensed disposal facilities, then ^{63}Ni can become the controlling activated radionuclide.

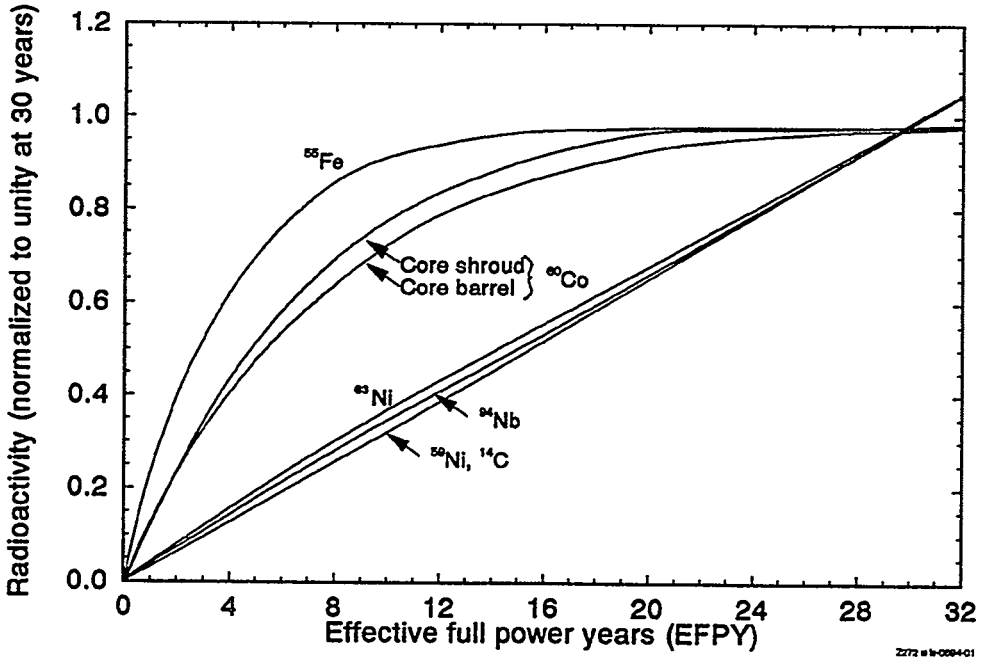

Figure 2. Buildup of some important activation products in a pressurized water reactor as a function of reactor operation. Note that the lowest curve from 0 to ~30 years represents ^{59}Ni, and the third lowest curve from 0 to ~30 years is ^{63}Ni. (Obtained from References 21 and 22).

WASTE TYPES AND FORMS THAT CONTAIN NICKEL-63

Nuclear Reactors

The quantity of ^{63}Ni generated within the structural components of a nuclear reactor increases linearly with burnup; that is, double the reactor operation and the ^{63}Ni inventory doubles. Because the half-life of ^{63}Ni is much longer than the life of the reactor, no process effectively decreases the ^{63}Ni inventory over the reactor's useful life time. The inventory of ^{63}Ni in a reactor that operated at 80% power (880 MWe) for 40 years would be approximately 1×10^6 Ci.[23] Consequently, ^{63}Ni is important in the classification of radioactive wastes from nuclear power plants.[24] Because nickel and cobalt are chemically similar, much of what is known about the presence of radioactive nickel in radioactive waste has been inferred from similar studies involving cobalt, especially ^{60}Co, a species that is easier to detect and measure.

The primary low-level radioactive wastes containing ^{63}Ni are those solid waste forms produced by activation of structural materials, mainly stainless steel components. During decommissioning and dismantling of these components, low-level radioactive wastes are produced that contain ^{63}Ni. For example, it is possible that structural components from decommissioned nuclear power plants will be melted down for recycling. For this reason, a laboratory study was conducted by the German company, Siemens, to investigate the mobility of various radioactive species in melted steel.[25] The results from this study indicate that nuclides of iron, cobalt, and nickel remain in the melt and that release to the slag and off-gas systems was negligible.

Low-level radioactive wastes from nuclear reactors always contain small quantities of fission products or activation products, including ^{63}Ni. During the operation of a light water reactor, oxide corrosion films form on the surfaces of piping, pumps, valves, steam generator tubing, and many other components in contact with the primary coolant. Some of these corrosion products dissolve or erode in the circulating coolant, are deposited on the fuel cladding, and become activated by the neutron flux. The principal radionuclides formed by this activation process are ^{58}Co, ^{60}Co, ^{55}Fe, ^{59}Fe, ^{54}Mn, ^{51}Cr, ^{65}Zn, and ^{63}Ni. These radionuclides are eventually released from the core region of the reactor and are carried by the coolant and incorporated into oxide films on the surfaces of the primary coolant system (PCS) components.[26] The principal waste types that can contain radionuclides that are produced as a result of nuclear reactor operations are contaminated scrub water and decontamination solutions; contaminated clothing; contaminated tools and equipment used in radioactive areas; pumps, valves, seals, bearings, scrap materials, and ion-exchange resins and filters. However, none of the principal low-level radioactive waste types contain radiologically significant quantities of ^{63}Ni. In the case of ^{63}Ni, only very small quantities of this radionuclide are found in the primary coolant system of operating reactors as a result of activation of suspended metals or crud. However, the equipment and systems used to clean this water will contain much higher concentrations of ^{63}Ni, as well as other radionuclides. In some cases, ^{63}Ni may be the limiting radionuclide that determines handling, storage, and disposal requirements.[27] The types of safeguards to be used in the disposal of decommissioned reactor parts and the long-term safety of the disposal of these parts are dictated by the amount of radionuclides, including ^{63}Ni, contained in the parts.

In a European laboratory study, representative batches of primary water, evaporator concentrates, ion exchange resins, and filter cartridges were collected at different pressurized

water reactors and destructively analyzed to determine the content of, among other species, ^{63}Ni.[24] On the basis of this study, the researchers concluded that the isotopes of nickel, including ^{63}Ni, were principally suspended in the water samples, rather than dissolved in the water itself.

The presence of ^{63}Ni in reactor waste streams is routinely measured at the Palisades Nuclear Power Plant.[28] Of the waste streams listed in this reference, ^{63}Ni was listed as showing up in only one specific location, the laundry system drain effluent, comprising a few percent of the total radioactivity in this stream.

Medical, Academic Institutions, and Commercial Wastes

Because ^{63}Ni has a relatively long half-life and because it emits only weak beta radiation, medical applications of the radionuclide do not exist. In addition, since ^{63}Ni decays directly to the ground state of ^{63}Cu, there would be neither alpha nor gamma radiations. Therefore, the use of ^{63}Ni as an external radiation source would be severely limited.[29] No significant commercial or medical wastes containing or using ^{63}Ni have been identified; however, ^{63}Ni has been used in the form of a nickel sulfate solution in some small scale laboratory experiments.[30] ^{63}Ni has also been used in some experiments involving radionuclide imaging (e.g., autoradiography) of cells.[31] There are also no significant waste streams of ^{63}Ni from academic institutions; however, some small quantities of ^{63}Ni have been used in a limited number of experiments. Most ^{63}Ni wastes from small laboratory or commercial experiments enter the same low-level radioactive waste streams as other commonly used radioactive materials. Personnel at the American College of Radiologists reported that ^{63}Ni is not used for the diagnosis of disease or for medical treatments.[b]

Nevertheless, ^{63}Ni has been used in the laboratory during tests on experimental medications. For example, this nuclide was used as part of the measurement system during an experimental study of Nitrazepam (a drug used in the treatment of insomnia).[32] As such, it will eventually enter the environment in some waste streams.

Use was made of ^{63}Ni in an epidemiology study conducted by the Lovelace Biomedical and Environmental Research Institute.[30] This study investigated the exposure of refinery workers to airborne nickel. The radioactive nuclide was used to trace the nickel contamination in laboratory animals and was not itself the subject of the investigation.

This radionuclide is also used in laboratory studies at the Chalk River Nuclear Laboratories.[33] In a study investigating the release of several laboratory nuclides to the environment, researchers assumed that the radionuclides would be incinerated and that the ash would be transported to a municipal landfill. From the landfill, the radionuclides could be leached into the ground water. Even with worst case assumptions, the study concluded that only very low exposure to the public would occur from landfill disposal of all the studied nuclides.

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b. Private communication between C. Sperry (ACR) and J. Adams (INEL) on November 7, 1994.

Historically, ^{63}Ni compounds have also been available in the following chemical forms: $NiCl_2$ in solution (e.g., 1 N HCl) and $Ni(NO_3)_3$ in solution (e.g., in 1 N HNO_3).[34] These materials have been used in some unspecified industrial applications, possibly small research experiments.

In summary, ^{63}Ni, though not used specifically to diagnose or treat diseases, has been used in a small number of laboratory studies. Although it can enter the environment via several methods, the small amount would pose very little risk to the public.

BEHAVIOR OF NICKEL-63 IN THE ENVIRONMENT

Nickel in Soils

Understanding the behavior of radioactive materials such as ^{63}Ni in the soil is important in assessing the possibility that these radionuclides will be transported through the biosphere.[35] It is important to understand the interactions between radionuclides and various media along the path to the biosphere, whether disposal is in deep or shallow rock caverns or in shallow overburden facilities.

Nickel occurs naturally in many soils. In fact, on an elemental basis, the naturally occurring forms of nickel can be much more prevalent than the trace amounts of ^{63}Ni found in typical waste streams.[36] The typical range for naturally occurring nickel concentrations in soils is 5 to 1000 parts per million (ppm), with some soils containing up to 6200 ppm nickel. Concentrations, either expected or actual, for radionuclides, including ^{63}Ni, are much lower. Nickel also occurs naturally in groundwater, with typical concentrations up to 50 ppm, much higher than radionuclide concentrations. For water with a pH less than 8, the dominant ionic form for nickel is Ni^{2+}.

Four parameters were reported to be essential in accurately predicting soil concentrations from either contaminated ground water or irrigation water.[35] The four parameters, in order of decreasing importance, are (a) soil retention, (b) annual precipitation, (c) soil texture, and (d) 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 nickel concentration in the soil (i.e., µg/gram of soil) and C_l is the nickel concentration in the groundwater (i.e., µg/ml of liquid). Therefore, the units of K_d are ml/g (i.e., mL of water per gram of soil). This empirical model combines all soil retention mechanisms into a simple linear partition relation between the soil and the surrounding groundwater. This model assumes that the nickel concentrations in groundwater and soil are in equilibrium with each other. Using this definition for K_d, it follows that the larger the value (that is, the higher the radionuclide concentration in the soil relative to that in the groundwater), the slower the migration of the radionuclide relative to 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, using samples that are relatively homogenous, and where it can be ensured that equilibrium conditions are met. Applying these values to specific soils in the field can be difficult because actual soils are generally nonhomogeneous and there are uncertainties as to how long it takes for the nickel 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 that extrapolate data from laboratory experiments and field studies. In addition, some experiments are conducted using intact field samples to validate the laboratory K_d values and to study the effects associated with soil nonhomogeneity.

The Environmental Protection Agency (EPA) performed a study of K_d values for a number of radionuclides, including ^{63}Ni, as part of the low-level radioactive waste program.[37] In this

study, the EPA characterized the soils covering most of the United States into three major deposits: the glaciated central region, atlantic and gulf coastal plains, and the alluvial basin. They then researched K_d values for each radionuclide and soil type. The values of K_d for ^{63}Ni ranged from 2,000 to 5,000 mL/g for the alluvial basin (soil retention high) to between 100 and 500 mL/g for the coastal plains (soil retention moderate) to less than 100 mL/g for the central region (soil retention low). The differences between these values are judged to result from soil pH differences — higher pH values result in higher retention.

To verify this conclusion, the EPA quotes a separate effects study[38] performed on soil samples taken from Hanford, Washington. First, the K_d for the as-collected soil with a pH = 9 was measured to be 1,300 mL/g. Then, the soil pH was adjusted to a value of 6 and the K_d again measured. The K_d value for the high pH soil sample was about an order of magnitude higher than in the pH-adjusted sample.

A summary of the environmental interactions of various radioactive species, including ^{63}Ni, has been published in Reference 39. In this summary, it is concluded that when ^{63}Ni enters a soil system, most of it (90%) is rapidly adsorbed in the soil and therefore removed from ground-water migration.

In *The Soil Chemistry of Hazardous Materials*,[36] the adsorption of nickel is presented as a function of pH in two different complexing environments, an iron-gel and an aluminum-gel. For the aluminum-gel environment, there is almost a step change in percent adsorption of nickel from less than 10% to nearly 100% as the pH is increased above 6. A similar step change occurs for the iron-gel environment, but at the lower pH value of 5.

Soil composition may also play a significant role. The central plains soil has a clay-silt composition, resulting in the least nickel soil retention. The alluvial basin soil has a sandy composition and the highest retention. The coastal plain soils lie between these two (e.g., a clay-sandy composition), and has an intermediate retention.[37]

As a final conclusion of the EPA study, nickel adsorption in soil is also known to be significantly affected by precipitation and coprecipitation reaction with hydrous oxides of iron and manganese.

Reference 38 reports on an investigation by Pacific Northwest Laboratory into the effect of organic complexing agents on the adsorption of nickel in soils. Several organic complexants were used, including ethylenediaminetetraacetic acid (EDTA), diethylenetriaminepentaacetic acid (DTPA), oxalate, and citrate. Each of these complexants is widely used in the commercial nuclear industry as a decontaminating agent and will, therefore, be present in untreated nuclear wastes. The presence of these complexants tends to make the nickel more soluble in water and less likely to be adsorbed in soils. However, it should be noted that these are mixed wastes (both hazardous (as defined by the Resource Conservation and Recovery Act) and radioactive; they will not be co-disposed with commercial LLW.

Of the complexants studied, EDTA seemed to have the largest effect. In a nickel solution that was pretreated with this complexant, nickel did not appreciably adsorb onto soil from the Hanford, Washington area even though it was in contact with the soil for more than one year.

These tests were repeated one year later—while the adsorption rate was still very slow, there was some adsorption of nickel-EDTA solution in the Hanford soil.[40] This experience illustrates the difficulties involved in generating reproducible results using field samples of soils. There was a significant difference between nickel that was treated with EDTA before coming in contact with soil and nickel that was adsorbed in the soil before being treated with EDTA. Nickel that had been adsorbed in the Hanford soil (pH = 9) before being treated with EDTA was only slowly dissolved by the complexant. After 6 months, only about one-half of the sorbed nickel was dissolved, whereas pretreated nickel remained completely dissolved. Very little additional nickel redissolved, even though the test ran for an additional 9 months, indicating that an equilibrium between dissolved and sorbed nickel may have been reached. The study was repeated using soil from the Savannah River Site (near Aiken, South Carolina). The pH of the Savannah River soils was lower, ~6, and the nickel behavior was significantly different. First, in the pretreated case, about one-half of the nickel in solution was adsorbed in the soil, which was the same amount of nickel that was redissolved in the soil that was not pretreated. Second, the equilibrium was reached much sooner, within a few weeks.

Similar studies were conducted using DTPA as the complexing agent. In the Hanford soil, pretreated nickel slowly adsorbed into the soil and untreated nickel slowly redissolved. Equilibrium was not reached, even after one year. As was the case with EDTA, equilibrium was reached much more rapidly with the Savannah River soil, after a couple of days. The adsorbed fraction at equilibrium was approximately 20%. A separate test was conducted with Savannah River soil, but at a lower pH (~4). Equilibrium was reached at an adsorbed fraction of 70%, also within a couple of days.

Oxalate is known to form much weaker complexes with metal ions, such as nickel, than either the EDTA or DTPA. In tests involving this complexant and Hanford soil, the equilibrium was reached much more rapidly, within a few days, than with the other two complexants. Possibly because of the shorter equilibration times, parametric studies were conducted, varying concentrations of nickel and oxalate, solution to soil ratio, soil particle size, and direction of approach to equilibrium. Of these, only the complexant concentration seemed to affect the results. The equilibrium adsorbed fraction decreased nearly 100-fold with a 10-fold increase in complexant concentration. Decreasing the soil pH from 9 to 5 (using oxalic acid) resulted in an adsorbed fraction decrease by one-third. Repeating these tests using Savannah River soil indicated less of a dependence on oxalate concentration than with the Hanford soil.

Finally, tests were conducted using citrate as the complexing agent. Citrate, like oxalate, forms much weaker complexes with nickel than EDTA or DTPA. In the Hanford soil tests, the equilibrium was reached within a few days and ~90% of the nickel was adsorbed into the soil. The Savannah River soil tests did not reach an equilibrium, even after 25 days, though the adsorbed fraction was increasing throughout the test time.

The study reported in Reference 38 was expanded to include two additional complexants, HEDTA and NTA in 1983.[40] These two complexants are of intermediate strength, between the strong EDTA and DTPA and the weak citrate and oxalate. In addition, one more soil type was included, Oak Ridge soil, which was taken from an Oak Ridge waste burial ground and was significantly finer than either the Hanford or Savannah River soil samples. The Oak Ridge soil pH was close to that of Savannah River soil, pH = 5.5.

There were, however, significant differences between the adsorption characteristics of nickel in the Oak Ridge and Savannah River soil samples. For example, in the Savannah River soil tests using preadsorbed nickel (that is, nickel that was adsorbed into the soil prior to addition of the complexant), a significant amount of the nickel was redissolved when an EDTA-laden solution was passed through the soil. However, no redissolution of nickel occurred with the Oak Ridge soil sample.

In Hanford soil, equilibrium conditions were not met with the HEDTA-complexed nickel, even after 2 months of testing. Equilibrium was met with the NTA-complexed nickel after approximately 1 month. The equilibrium adsorption fraction with NTA-complexed nickel ranged between 70 and 90%, depending on the molarity of the NTA. Similar tests conducted with Oak Ridge soil samples were more complex. The dependence of the adsorbed fraction on NTA concentration was greater than with the EDTA-complexed nickel.

An analytical study of the leakage of ^{59}Ni from an underground vault is presented in Reference 41. Although this study is specific to ^{59}Ni migration from an underground storage facility, it also has implications for any isotope of nickel, including ^{63}Ni. In this study, radioactive structural components of a reactor were placed in a 500-meter deep tunnel, lined with concrete and covered with a clay/quartz mixture. Given best estimates on soil and groundwater conditions, it was calculated that the nickel would escape over a period of 20,000,000 years. Even if the concrete and clay/quartz were to become completely permeable to the groundwater (an extremely unlikely event), the maximum leach rate for the nickel was calculated to be 2×10^{-6} parts per year.

Independent studies have been made of the behavior of ^{63}Ni in soil by European scientists.[42] In this laboratory study, the diffusion of ^{63}Ni through various sedimentary rock samples in Germany was measured and compared with retention coefficients, K_d. The scientists concluded that diffusion coefficients could be measured and used to model transport of ^{63}Ni through various soil types. The diffusion coefficient for ^{63}Ni was comparable to that for other radioactive species included in the study (e.g., 2.63×10^{-9} cm^2/s for ^{63}Ni compared with a range of 3.29×10^{-8} cm^2/s to 5.64×10^{-9} cm^2/s for uranium and radium nuclides for the same soil type).

In summary, the behavior of nickel in soils depends on a variety of parameters. These include soil/groundwater pH, soil type and texture, and the presence or absence of complexing agents.

Nickel in Groundwater

Nickel tends to be easily adsorbed by soils, removing it from groundwater. However, this condition is dependent on a number of parameters, especially groundwater pH and whether or not complexing agents are present. For example, in a Hanford soil sample (with a pH = 9 and complexed with EDTA) nickel was not appreciably removed from the solution even after contact with the soil for over one year. Thus, the presence of complexing agents can significantly slow removal of nickel from groundwater. The equilibrium water concentration of nickel depends on pH and the particular complexant used, as discussed in the previous section.

Personnel at the Pacific Northwest Laboratories studied the transport and chemical behavior of a number of radionuclides in a slightly contaminated groundwater plume.[43] In this study, they observed that several radioactive species, including ^{63}Ni, were transported, principally in a nonionic form. In this form, these species are fairly mobile. At least for the case of ^{63}Ni, this behavior (nonionic chemical form) was not predicted using chemical thermodynamics. They assumed that complexing nickel with organic contaminants may explain the anomalous chemical form.

Reference 39 includes estimates on the uptake of ^{63}Ni from dilute concentrations in an aquatic environment into the underlying soil. The transfer factors (ratio of nickel concentration in the water to that in the soil) were in the range of 10^{-4}, indicating that very little of the original nickel would remain in solution and was therefore not available for transportation with the water.

While not specifically a ground water issue, the contamination of sediment and deep sea water were studied near two nuclear-powered submarines that were lost at sea in the 1960s.[44] In this study, there were measurable, though small, concentrations of ^{63}Ni in the sediment surrounding the submarines (probably resulting from corrosion of activated reactor structural parts or release of corrosion products from the reactor plants). However, no measurable ^{63}Ni was found in the water, marine animals, or in fauna.

Nickel in Plants

Nearly all information that was located on the uptake of nickel by plants is based on naturally occurring or stable isotopes of nickel. Little information specifically discussed ^{63}Ni; however, ^{63}Ni should follow the behavior of its stable isotopes. One summary reference, *Nickel*,[1] contains a discussion of naturally occurring nickel uptake into plants. Naturally occurring nickel concentrations in the soil range from a few parts per million (ppm) to several thousand ppm. Typical plant nickel concentrations in grains, fruits, and vegetables range from a few tenths of a part per million to a few part per million. Thus, the uptake of nickel from soil to these plants appears to be a very small fraction of the soil concentration. Some grasses, including field-grown oats, may contain more than 100 ppm nickel, and some flowers (alyssum, forget-me-nots, rice flowers), up to several thousand parts per million. In alyssum, the highest concentrations of nickel were in the leaves, with less than one-tenth the concentration in the seeds. Nickel can enter plants via the roots, via adsorption through the leaves or a combination.

Laboratory tests summarized in Reference 39 include studies of the uptake of ^{63}Ni by various plant species. Based on these tests (which were conducted on sunflowers), it was concluded that ^{63}Ni is very mobile within the plant; that is, it is distributed throughout the plant structure, although more than half of it migrates to newly developing organs within the plant, such as new leaves.

The transfer ratio between soil and plants (ratio of nickel concentration in the soil to that in the plant) is approximately 10% for most plants, although it can be as high as 50% for legumes. There was no evidence of accumulation of this nuclide in the seeds, fruits, or tubers of these plants.

Reference 39 also includes a study of the uptake of nickel by marine plants. In this study, the marine plants tended to concentrate the nickel by a factor of 1,000 over that in the water. Thus, any nickel in solution would tend to be taken up by marine plants where it would be available for ingestion by animals.

Nickel in Air

Nickel isotopes (radioactive or nonradioactive) are not normally found in air, except possibly for small amounts generated and released following an atmospheric test of a thermonuclear weapon. Measurements were made of ^{63}Ni in marine animals near the Pacific Ocean nuclear test sites, Bikini and Eniwetok Atolls.[45] Measurable amounts of ^{63}Ni were discovered in clams as a direct result of the atmospheric tests conducted at these sites.

Radioactive ^{63}Ni is produced entirely as an activation product in the reactor structural components and does not appear as a fission product radionuclide. Therefore, even if a severe core damage accident in a nuclear reactor released significant amounts of fission products, this would not necessarily mean that significant amounts of ^{63}Ni would be released into the air.

There is some indirect evidence of airborne radioactive nickel isotopes having resulted from nuclear explosions.[1] Very small amounts of ^{63}Ni have been measured in soil and marine life around the Pacific Proving Ground, the site of several atmospheric nuclear tests. The maximum concentrations of this nuclide are in the parts per trillion range.

BEHAVIOR OF NICKEL-63 IN THE HUMAN BODY AND IN ANIMALS

The presence and behavior of nickel has been studied both in humans and in a number of animal species. Annual limits for ^{63}Ni are based on this research, which is summarized in the following paragraphs.

Human Body

Nickel is present in all human tissue in trace amounts. Even newborn infants have measurable amounts (of the order of 10^{-5} g/liter of the umbilical cord serum). There is evidence that nickel may be an "essential micronutrient" for health and growth.[1]

Distribution of nickel is generally uniform throughout the human body. Data from the "Reference Man," used in studies of the International Commission of Radiation Protection, indicate that there are approximately 10 milligrams of nickel in a typical adult, with ~5 milligrams in the soft tissues and ~5 milligrams in the skeleton tissues.[46] Studies have investigated the effects of health problems associated with either an overabundance or deprivation of nickel. Many of the studies reviewed for this report, however, deal with naturally occurring, stable isotopes of nickel and not with radioactive nuclides such as ^{63}Ni. *Safe Handling of Radioisotopes*[47] states that the liver is the critical organ with respect to ingestion of ^{63}Ni.

Reference 39 concludes that nickel distributes uniformly throughout the human body. Ingested nickel is only poorly adsorbed by the intestinal tract (e.g., the fraction adsorbed is of the order of 1%). In animal studies, between 60 and 90% of intravenously injected nickel was excreted within 24 hours. The biological half-life of the remaining nickel in the human body (after the initial excretion) is estimated to be approximately 500 days.

Radionuclides have been classified as to their relative toxicity in man.[48] In this study, radionuclides are grouped into three classifications: high toxicity, medium toxicity, and low toxicity. Because of the large number of nuclides in the middle group, it was further subdivided into an upper (Group A) and lower (Group B) group. ^{63}Ni is included in the Medium Toxicity Group B.

The dose to the various body organs from ^{63}Ni inhalation has been reported in the *Health Physics and Radiological Health Handbook.*[49] In Table 11.3.1 of this reference, the dose (per exposure) for the internal organs (including lungs, gonads, bone marrow, and thyroid) is fairly uniform at $\sim 8 \times 10^{-10}$ Sv/Bq for a one day exposure. The dose from ingestion is also uniform at $\sim 8 \times 10^{-11}$ Sv/Bq.

Animal Studies

Reference 1 contains some information regarding the metabolism of nickel in laboratory animals. After nickel-chloride was injected into rats (stable Ni), nearly 70% was excreted within 72 hours (61% in urine and 6% in feces). All blood and plasma nickel disappeared within 48 hours. After 72 hours, only the kidneys exhibited significant amounts of nickel. The initial excretion of injected nickel differed from nickel that was orally administered in that the orally ingested nickel was preferentially (90%) excreted in the feces, indicating only minimal adsorption

via the intestines. A separate study included injections in both rats and rabbits. The conclusions from this study did not differ substantially from those of the previous study—preferential excretion via urine during the first 48 hours and then more gradual excretion over 3 to 7 days. Because of the long radiological half-life of ^{63}Ni, the effective half-life of ^{63}Ni in the human body is primarily a result of its biological half-life. The biological half-life refers to the amount of time required to remove half of the initial material from the body via a combination of chemical, physical, or biological processes. The effective half-life incorporates both the radiological and biological half-lives. For ^{63}Ni, the biological half-life has been reported as 667 days for the total body, 800 days for bone, and 500 days for the liver.[50] However, Reference 51 of the International Commission of Radiological Protection reports the biological half-life of all nickel isotopes as 1,200 days.

Reference 52 reports a study of the bioaccumulation in the food chain. Researchers noted that ^{63}Ni concentrated mainly in the pancreas and exoskeleton of shrimp and in the skin and viscera of fish. This distribution is consistent with previous studies wherein ^{63}Ni was observed to concentrate in the shell of clams.[53]

In addition, personnel at the Battelle/Marine Research Laboratory studied the biological uptake of ^{63}Ni into the food chain.[52] In this laboratory study, various stainless steel corrosion products, including ^{63}Ni, were introduced into marine sediments. Naturally occurring worms were exposed to these sediments and then fed to shrimp and fish. The accumulation of ^{63}Ni in the soil, worms, shrimp, and fish were then measured. They concluded that ^{63}Ni will be transported from the soil up the food chain, though there was no evidence that this nuclide was biologically concentrated in any of the animals. Indeed, there was some evidence that the ^{63}Ni concentration decreased as it was passed up the food chain. Researchers noticed that ^{63}Ni was more readily bioaccumulated from the sediment than was ^{60}Co, the other radioactive material studied.

In a separate study,[30] male and female rats were injected with nickel-sulfate that contained trace amounts of ^{63}Ni, and the metabolism of the nickel was investigated. The results indicated that the internal organs contained the highest concentrations of nickel and that the principal excretion route (accounting for 50% to 80% of the ingested Ni) was urine. The other two routes were fecal excretion (13% to 30 %) and expiration. The half-life for excretion was dose dependent; ranging from 4.6 hours for the highest dose (1800×10^{-9} moles Ni) to 23 hours at the lowest dose (17×10^{-9} moles Ni).

The annual limits on ^{63}Ni intake (ALI) and the derived air concentration (DAC) for ^{63}Ni are shown in Table 5. 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 for inhalation or ingestion, and DACs for inhalation (or submersion). For a radionuclide whose derived value does not change from the old definition, the DAC is numerically equal to the MPC value in air.[54] The information in Table 5 applies only to ^{63}Ni. In the case of multiple radionuclides released in a mixture, additional guidelines outlined in Title 10 of the Code of Federal Regulations, Part 20[55] must be followed.

Table 5. Annual limits on intake (ALI) and the derived air concentrations (DAC) for ^{63}Ni. (Data obtained from References 54 and 55).

Radionuclide	Component	Ingestion (μCi)	Inhalation[a]
^{63}Ni	ALI	9000[b] μCi	2000/D[b] or 3000/W[b] or 800 [vapor] μCi
	DAC	—[c]	0.7/D or 1.0/W 0.3 [vapor] μCi/m^3

a. Clearance from the lung directly to the blood stream or to the gastrointestinal (GI) tract depends upon the chemical form of the radionuclide and is classified as D, W, and Y, respectively, for clearance times of the order of days, weeks, and years.

b. Removal class is f_1=0.05. A simple model of the lung was used to describe the translocation and retention of material by the body after inhalation. In this model 25% of the inhaled activity was exhaled and 25% was deposited in the lower respiratory tract. The 50% that was deposited in the upper respiratory tract was eventually cleared by means of mucociliary processes and swallowed. What happens then depends on whether the inhaled material was in either a soluble or insoluble chemical form. Any soluble material deposited in the lower respiratory tract is assumed to be transferred directly to the blood stream. The activity cleared from the upper respiratory tract and then swallowed, a fraction (f_1), and in the case of ^{63}Ni f_1=0.05, entered the blood-stream via the GI tract. Additional details of this model are described in Reference 54.

c. Data not available.

CONCLUSIONS

Nickel-63 is not a naturally occurring radionuclide. Its half-life is approximately 100 years and decays by beta particle emission. When it decays, no x-rays or gamma rays are emitted. ^{63}Ni has been used in a small number of biological experiments; however, no specific medical application of ^{63}Ni has been identified.

^{63}Ni is produced in the structural steels of nuclear reactor vessel and internal components from neutron activation of ^{62}Ni, a naturally occurring and stable isotope of nickel. These reactor components serve as the primary low-level radioactive waste material containing ^{63}Ni. The major concern surrounding ^{63}Ni is in limiting the dose received by persons associated with the decommissioning and dismantling of the reactors, primarily for reactors in service for more than 30 years. At that point, the ^{63}Ni activity will become an important activation product with regard to decommissioning activities. Limited amounts of ^{63}Ni can enter the environment through operational wastes from a nuclear reactor (i.e., corrosion of stainless steel surfaces and circulation of coolant).

The chemistry of nickel is dominated by the 4s subshell electrons. While other oxidation states can be produced, the principal form is Ni^{2+}. Nickel 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.

The ability of soil to adsorb ^{63}Ni out of groundwater varies depending on the soil type and water pH. In general, however, nearly all of the ^{63}Ni present in groundwater is expected to be adsorbed by the soil, with only 1% or less of the nickel remaining dissolved in the water. Soil adsorption can be affected, however, by the presence or absence of certain decontamination agents such as EDTA and DPTA, which can significantly reduce the ability of the soil to adsorb the nickel.

When ^{63}Ni comes into contact with the soil and groundwater, most of it will become fixed in the soil and will not migrate appreciably from the original site. Plants and animals can take up ^{63}Ni from the soil and propagate it into the food chain. When this nuclide is ingested in mammals, most of it is immediately excreted within 24 hours. That part that is absorbed tends to be uniformly distributed throughout the body and consequently remains within the body for a long time. The biological half-life of ^{63}Ni in the human body has been reported to be 500 to 1,200 days (total body).

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