

WASTE MANAGEMENT, RADIOACTIVE

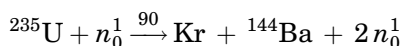
Radioactive wastes are generated in all parts of the fuel cycle supporting nuclear electric power, including mining and milling of uranium ore, chemical conversion, isotope separation, fuel fabrication, nuclear reactor operation, spent fuel storage, and waste disposal. Successful management of wastes from nuclear reactors is vital to continued use of nuclear power, and assurance of waste safety should aid in improving public acceptance. Many reactor wastes are radioactive. Disintegration (decay) of various materials results in the release of high-energy

radiation requiring protective measures. Half-lives of reactor products range from fractions of a second to billions of years.

Classification of wastes may be according to purpose, distinguishing between defense waste related to military applications and commercial waste related to civilian applications. Classification may also be by the type of waste, ie, mill tailings, high-level radioactive waste (HLW), spent fuel, low-level radioactive waste (LLW), or transuranic waste (TRU). Alternatively, the radionuclides and the degree of radioactivity can define the waste. Surveys of nuclear waste management (1,2) and more technical information (3–5) are available.

1. Sources

Three common sources of radiation associated with the nuclear fuel cycle are naturally occurring uranium and thorium, which are mined to produce nuclear fuel, neutron activation, and fission. Activation involves the absorption of a neutron by a stable nucleus to form an unstable nucleus. An example is the reaction of a neutron and cobalt-59 to yield cobalt-60 [10198-40-0], ^{60}Co , a 5.26 year half-life gamma-ray emitter. Another is the absorption of a neutron by uranium-238 [24678-82-8], ^{238}U , to produce plutonium-239 [15117-48-3], ^{239}Pu , as occurs in the fuel of a nuclear reactor. Fission occurs when a neutron is absorbed by uranium-235 [15117-96-1]. One typical reaction is as follows:



Fission of ^{235}U almost always results in two fission fragments plus some neutrons. Most uranium fission fragments are radioactive. Of special interest are technetium-99 [14133-76-7] and iodine-129 [15046-84-1] having half-lives of 2.13×10^5 yr and 1.7×10^7 yr, respectively. Data on all isotopes are found in Ref. 6 (see also RADIOISOTOPES).

Radioactive waste is characterized by volume and activity. Activity is defined as the number of disintegrations per second, and it is measured in becquerels. One disintegration per second is one becquerel. Each radionuclide has a unique half-life, $t_{1/2}$, and corresponding decay constant, $\lambda = 0.693/t_{1/2}$. For a component radionuclide consisting of N atoms, the activity A is defined as

$$A = N\lambda$$

Activities and existing and projected volumes of all types of radioactive waste are listed in Ref. 7.

Most uranium ore has a low, ca 1 part in 500, uranium content. Milling involves physical and chemical processing of the ore to extract the uranium. The mill tailings, which release gaseous radon-222 [13967-62-9], ^{222}Ra , half-life 3.82 d, are placed in large piles and covered to prevent a local health problem.

Uranium oxide [1344-57-6] from mills, whose isotopic concentration is $\sim 0.7\%$ ^{235}U and $\sim 99.3\%$ ^{238}U , is converted into uranium hexafluoride [7783-81-5], UF_6 , for use in gaseous diffusion or centrifuge isotope separation plants (see DIFFUSION

SEPARATION METHODS) where the material is enriched; ie, its ^{235}U concentration is increased. The wastes from these operations are only slightly radioactive. Both uranium-235 and uranium-238 have long half-lives, 7.08×10^8 and 4.46×10^9 years, respectively. Uranium enriched to around 5 wt% is shipped to a reactor fuel fabrication plant (see NUCLEAR REACTORS AND NUCLEAR FUEL RESERVES). There conversion to uranium dioxide is followed by pellet formation, sintering, and placement in tubes to form fuel rods. The rods are put in bundles to form fuel assemblies. Despite active recycling, some low activity wastes are produced.

Uranium dioxide fuel is irradiated in a reactor for periods of one to two years to produce fission energy. Upon removal, the used or spent fuel contains a large inventory of fission products. These are largely contained in the oxide matrix and the sealed fuel rods.

Spent fuel can be stored or disposed of intact, in a once-through mode of operation, practiced by the U.S. commercial nuclear power industry. Alternatively, spent fuel can be reprocessed, ie, treated to separate the uranium, plutonium, and fission products, for reuse of the fuels (see NUCLEAR REACTORS, CHEMICAL REPROCESSING). In the United States, reprocessing is carried out only for fuel from naval reactors. In the nuclear programs of some other countries, especially France and Japan, reprocessing is routine.

Water as coolant in a nuclear reactor contains some radioactive atoms created by neutron irradiation of corrosion products of materials used in reactor construction. Key nuclides and the half-lives in addition to cobalt-60 are nickel-63 [13981-37-8] (100 yr), niobium-94 [14681-63-1] (2.4×10^4 yr), and nickel-59 [14336-70-0] (7.6×10^4 yr). Occasionally small leaks in fuel rods allow fission products to enter the cooling water. Cleanup of the water results in LLW. Another source of low-level waste is the residue from applications of radionuclides in medical diagnosis, treatment, research, and industry. Many of these radionuclides are produced in nuclear reactors, especially in Canada.

Weapons materials from production reactors were accumulated during the Cold War period as a part of the U.S. defense program. Prominent were tritium, ie, hydrogen-3, having a $t_{1/2}$ of 12.3 years, and plutonium-239, $t_{1/2} = 2.4 \times 10^4$ years. The latter constitutes a waste both as a by-product of weapons fabrication in a waste material called transuranic waste (TRU) and as an excess fissionable material if not used for power production in a reactor.

Several legislative actions govern the management of radioactive waste. The Atomic Energy Acts of 1946 and 1954 charged the Atomic Energy Commission with maintaining national nuclear defense and developing peaceful uses of the atom. The National Environmental Policy Act of 1969, which created the U.S. Environmental Protection Agency (EPA), initiated the requirement for an environmental impact statement (EIS) to be prepared for federal facilities. In 1980, the Low-Level Radioactive Waste Policy Act assigned responsibility for LLW disposal to the states. The Nuclear Waste Policy Act (NWPA) of 1982 set forth the schedule and procedure by which high-level waste would be managed by the U.S. Department of Energy (DOE). Each of these acts has been amended (8). The Energy Policy Act of 1992 required the EPA to develop environmental protection standards specifically for the Yucca Mountain high-level waste repository.

2. Treatment

Several modes of waste management are available. One technique is to hold the material for decay. This is applicable to radionuclides of short half-life such as the medical isotope technetium-99m ($t_{1/2} = 6$ h), the concentration of which becomes negligible in a week's holding period. The most common approach to waste management is to concentrate and contain. Various processes are applied to minimize volume and to prevent or delay access of water to the contents of waste containers (9,10).

2.1. Low-Level Waste Treatment. Methods of treatment for radioactive wastes produced in a nuclear power plant include (1) evaporation of cooling water to yield radioactive sludges, (2) filtration using ion-exchange resins, (3) incineration with the release of combustion gases through filters while retaining the radioactively contaminated ashes (see HAZARDOUS WASTE INCINERATORS), (4) compaction by presses, and (5) solidification in cement or asphalt within metal containers.

All processes in a nuclear plant, in a treatment facility, or at a disposal site are governed by rules of the U.S. Nuclear Regulatory Commission (NRC) (11). Radiation protection, ie, the limits on radiation dose to workers and the public, is specified. Exposure is maintained as low as reasonably achievable (ALARA).

Limits on concentration of radioactive content in air and water are specified in Part 20 of Ref. 11. For example, the concentration limit of gamma-emitting cesium-137 [10045-97-3], $t_{1/2} = 30$ years, in water is 0.037 Bq/mL (1×10^{-6} μ Ci/mL). For beta-emitting tritium, $t_{1/2} = 12.3$ years, in air, the limit is 0.0037 Bq/mL (1×10^{-7} μ Ci/mL). The NRC classifies wastes according to half-life and concentration in terms of activity per cubic centimeter. Classes A, B, and C are generally an increasing level of long-term hazard and requirement for integrity of disposal. Materials classed as LLW but of high concentration are designated as greater-than-class-C (GTCC). Mixed wastes, those having a hazardous material or feature plus a radioactive component, are subject to regulation by both the NRC and the EPA. Standards can differ significantly.

Radiation dose limits at a disposal site boundary are specified by the NRC as 25×10^{-5} Sv/year (25 mrem/year), a small fraction of the average radiation exposure of a person in the United States of 360×10^{-5} Sv/year (360 mrem/year). Protection against nuclear radiation is fully described elsewhere (12).

Nuclear utilities have sharply reduced the volume of low-level radioactive waste over the years. In addition to treating wastes, utilities avoid contamination of bulk material by limiting the contact with radioactive materials. Decontamination of used equipment and materials is also carried out. For example, lead used for shielding can be successfully decontaminated and recycled using an abrasive mixture of low-pressure air, water, and alumina.

2.2. Spent Fuel Treatment. Spent fuel assemblies from nuclear power reactors are highly radioactive because they contain fission products. Relatively few options are available for the treatment of spent fuel. The tubes and the fuel matrix provide considerable containment against release of nuclides. Intact assemblies can be encased in metal containers for storage or transportation to reprocessing facilities.

Reprocessing as practiced outside the United States involves chopping fuel rods into small pieces, leaching out the uranium oxide by nitric acid, and applying suitable solvents to separate the uranium, plutonium, and fission products (13,14). Typically, a vitrification process is applied to fission product wastes that are mixed with pulverized glass, heated to melting by an electric current, and the mixture poured into a metal canister for solidification, safe storage, and ultimate disposal.

Special chemical treatment can isolate the nuclides of intermediate half-life, ie, cesium-137 and strontium-90 [10098-97-2], ^{90}Sr , $t_{1/2}$ 29 years (15). These provide most of the radioactivity, radiation, and heat during the early years of a disposal facility. Solid-phase extraction methods using macrocycles and membranes promise to yield waste of low volume and activity (see INCLUSION COMPOUNDS AND MEMBRANE TECHNOLOGY). The separated intensely radioactive chemicals can be placed in separate storage or made available for industrial radiation use.

3. Storage and Transport

3.1. Storage. Storage of spent fuel assemblies in deep water pools at reactor sites serves several safety functions. Cooling by water prevents the fuel from melting from decay heat. The shielding effect of the water provides protection for workers in the vicinity from gamma-radiation. Moreover, adequate separation of assemblies in a pool prevents a chain reaction from occurring. The reinforced concrete pools are designed to withstand earthquakes. Water passes through a heat exchanger to maintain a constant temperature, and the purity of the water is assured by use of a demineralizer.

The pools of most reactors were designed for a limited number of fuel assemblies. As a consequence, pools are filling up. The accumulation of U.S. spent fuel over time for commercial sources is shown in Figure 1. The material awaits

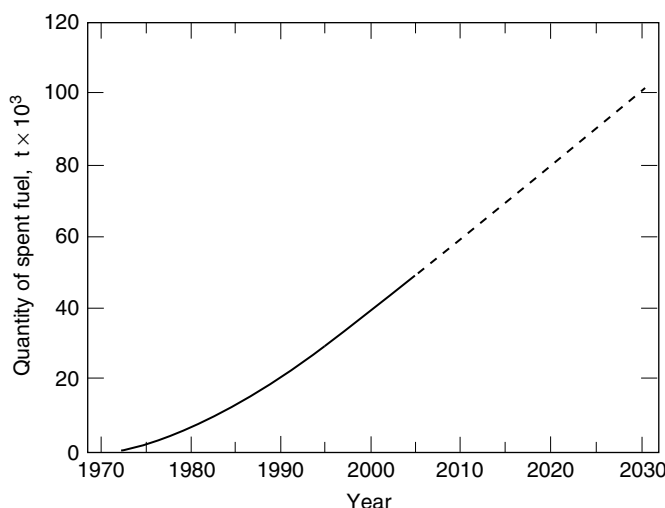


Fig. 1. Spent commercial nuclear fuel in the United States where (— —) represents projected quantities (1). Courtesy of Battelle Press.

permanent disposal. As pools have reached capacity, utilities have moved older, cooler spent fuel to large sealed concrete containers called dry storage casks. The fuel is inserted into the containers, water is drained off, and helium, an inert gas, is added. Arrays of these can be held on a thick concrete pad out in the open.

The NWPA of 1982 specified that the DOE would begin accepting spent fuel from nuclear utilities in 1998. To meet that target date, the DOE would have to use existing storage facilities at federal laboratories and possibly construct additional storage. As a part of the Nuclear Waste Policy Act, consideration was to be given to building a monitored retrievable storage (MRS) facility. Its role would be to receive fuel from utilities, store it for as long as needed, package it for safe burial, and ship it to a disposal site (16). The DOE was unable to identify an acceptable site for an MRS facility. A consortium of eight electric utilities and the Skull Valley Band of the Goshute Indians formed Private Fuel Storage, LLC, to develop an interim storage facility where dry storage casks containing spent fuel could be stored. In June 1997, Private Fuel Storage submitted an application to the NRC for a license for the storage facility. The NRC issued a license February 21, 2006.

Waste by-products of the operation of plutonium-producing reactors beginning in World War II have been stored in underground tanks at Hanford (Washington state) and the Savannah River Plant (South Carolina). Some single-walled tanks (Hanford) leaked, and the contents have been pumped into new double-walled tanks. Plutonium itself, generated by production reactors during the Cold War for weapons purposes, may become a waste by national policy (17). The plutonium would then be disposed of along with other high-level wastes. Alternatively, the plutonium could serve as fuel for reactors that generate electric power (see NUCLEAR REACTORS) or could be bombarded by neutrons produced by high-energy charged particles from an accelerator (see also PLUTONIUM AND PLUTONIUM COMPOUNDS).

Low-level waste with its generally smaller radioactivity level can be stored in suitable containers in buildings. Protective shielding and handling equipment are required.

3.2. Transport. In the United States, waste transportation is regulated by the NRC and the Department of Transportation (DOT). Packaging and shipping must conform with comprehensive rules. Shipping container classes are defined in accordance with the amount of radioactivity involved. A letter code for radioactivity, distinct from that for waste class, is used. Type A containers involve a minimum of protection, whereas type B containers must withstand a series of events, including specified impacts with hard surfaces or spikes, exposure to high-temperature fire, and long immersion in water. Spent fuel casks are of type B. For the movement of spent fuel, computer tracking systems are used. State radiological safety units are informed of shipments of spent fuel and other high-activity radioactive materials so that these units may respond in case of an accident.

A multipurpose canister (MPC) has been considered for the transportation, storage, and disposal of spent fuel, minimizing the amount of handling required. In the design of the container, factors being considered are fuel assembly size, weight, enrichment, amount of burnup, and age (18).

The safety record for transport of radioactive materials including spent fuel and wastes is excellent. Information about transportation of radioactive materials including waste is managed by the DOE. Codes such as RADTRAN that can calculate the public radiation dose owing to the passage of shipments have been developed. The maximum dosage from such shipments is a very small fraction of the typical annual radiation dose from all other sources.

4. Disposal

The disposal of radioactive waste is governed by rules of the NRC and the EPA (19). NRC regulations differ for low-level waste and for high-level waste, including spent fuel (20).

Isolation of radioactive wastes for long periods to allow adequate decay is sought by the use of multiple barriers. These barriers include the waste form itself, the primary containers made of resistant materials, overpacks as secondary layers, buffer materials, concrete vaults, and finally the host rock or soil. Barriers limit water access to the waste and minimize contamination of water supplies. The length of time wastes must remain secure is dependent on the regulatory limit of the maximum radiation exposure of individuals in the vicinity of the disposal site.

Performance assessments are predictions of radioactivity releases, the rate of transfer of contaminants through various media, and the potential for hazard to the public. These are based on a combination of experimental data obtained in the process called site characterization and detailed computations about radionuclides and their effects. The progressive attack on the metal or ceramic waste container, the diffusion of water into the waste form, the leaching of the radioactive compounds, diffusion out, and washing away of radionuclides are all considered.

Relevant hydrological fundamentals are used (21) to take account of the complex interaction of physical and chemical processes involving soil or rock, water, and contaminant. Attention is paid to uncertainties in calculated results.

Models for transport distinguish between the unsaturated zone and the saturated zone, that below the water table. There the underground water moves slowly through the soil or rock according to porosity and gradient, or the extent of fractures. A retardation effect slows the motion of contaminant by large factors in the case of heavy metals. For low-level waste, a variety of dose calculations are made for direct and indirect human body uptake of water. Performance assessment methodology is described in Ref. 22.

4.1. High-Level Waste. Many studies have been made of possible modes of disposal for high-level waste, including spent fuel. Some techniques considered are a very deep hole, a remote island, a mountainside, a subseabed, the Antarctic ice sheet, and delivery to space. As of this writing (ca 2006), the preferred method is deep underground burial in a mined cavity. Several types of rock have been considered as potential hosts to the waste, including granite, salt, basalt, and tuff. A history of high-level waste disposal prior to 1987 is available (23). A report on activities from 1991 to 2001 in programs being conducted in nations around the world to dispose of high-level radioactive waste in geological repositories can be found in Ref. 24.

Regulations on high-level radioactive waste management have traditionally been provided by the NRC, and several specific requirements on the character of a disposal site are spelled out (19,20). The generic regulations planned by the EPA have been delayed by court actions and federal law requirements. Wastes are to be placed at least 300 m below the earth's surface. The waste form must be free of liquid, noncorrosive, and noncombustible. The container is to remain intact for between 300 and 1000 years. The travel time of groundwater prior to waste emplacement is preferred to be greater than 10,000 years but not less than 1000 years. The repository must be placed where there are no attractive resources and far from population centers. Wastes are to be retrievable for a period of 50 years. Finally, releases of radionuclides from the repository must be less than figures specified by the EPA in 40 CFR 197, whose preparation was mandated by the Energy Policy Act of 1992. Typical limits are 3.70×10^{12} Bq/ 10^3 t (100 Ci/ 10^3 t) of heavy metal.

Regulations include guidelines on geologic conditions. Of special interest is the stability of the geology against faulting, volcanic action, and earthquakes. The repository is to be located in an arid region, where the water table is quite low. The host rock is to have a suitable porosity and a low hydraulic conductivity.

Tuff, a compressed volcanic material, is the primary constituent of Yucca Mountain, near Las Vegas, Nevada, the site selected by Congress in 1987 for assessment for spent fuel disposal. The site was studied for more than a decade and in 2002 was declared to be a viable site for the high-level waste repository. Site characterization studies include a surface-based testing program, potential environmental impact, and societal aspects of the repository. Performance assessment considers both the engineered barriers and the geologic environment. Among features being studied are the normal water flow, some release of carbon-14, and abnormal events such as volcanic activity and human intrusion. The DOE expects to submit to the Nuclear Regulatory Commission a license application for the repository in 2007. The application is likely to be under review for many years. The geologic aspects of waste disposal (25,26), proceedings of an annual conference on high-level waste management (27), and one from an annual conference on all types of radioactive waste (28) are available. An alternative to burial is to store the spent fuel against a long-term future energy demand. Uranium and plutonium contained in the fuel would be readily extracted as needed.

4.2. Low-Level Waste. The NRC 10CFR61 specifies the nature of the protection required for waste containers (20). Class A wastes must meet minimum standards, including no use of cardboard; wastes must be solidified; have less than 1% liquid; and not be combustible, corrosive, or explosive. Class B wastes must meet the minimum standards but also have stability; ie, these must retain size and shape under soil weight, and not be influenced by moisture or radiation. Class C wastes must be isolated from a potential inadvertent intruder, ie, one who uses unrestricted land for a home or farm. Institutional control of a disposal facility for 100 years after closure is required.

The traditional method of disposal of low-level radioactive waste has been shallow land burial, consisting of filling a deep trench and covering it with a layer of earth (29). The trend in time of annual volume of LLW per reactor in the United States is shown in Figure 2. Three of the original six commercial sites were closed owing to leaks of radioactivity. Two initiatives resulted: stricter

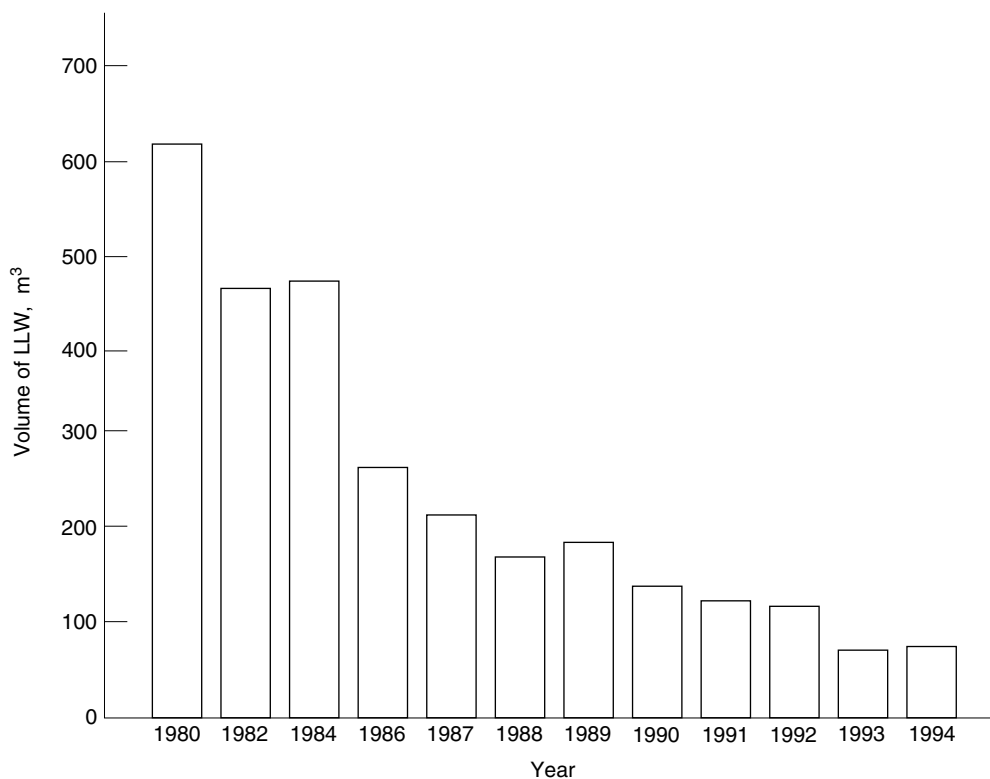


Fig. 2. Volume of low-level radioactive waste per U.S. nuclear power reactor (weighted industry median). The decrease over the period 1980–1994 was more than a factor of seven. Courtesy of Institute of Nuclear Power Operations.

regulations (20) and the 1980 Low Level Radioactive Waste Policy Act, which called for each state to be responsible for wastes generated within its borders, but it recommended the formation of compacts among states to build regional disposal facilities. Figure 3 shows the groupings that were formed. However, the compacts could not find low-level waste disposal sites that were acceptable to the public. The fourth of the original six commercial disposal sites closed in 1992, as was allowed in the Low-Level Radioactive Waste Policy Act. However, two of the original commercial sites (at Barnwell, SC, and Richland, WA) and a private facility, Envirocare, near Clive, UT, remain open. The cost of disposal of low-level waste has increased dramatically since the Low-Level Waste Policy Act was passed in 1980, and low-level waste generators have employed several techniques to reduce their waste volumes and minimize disposal costs. As a result, no new disposal facilities have been required. The historical background of low-level waste management is available (30,31).

One of the most important considerations in operating a low-level radioactive waste disposal facility is to control infiltration of water into the facility. Three techniques employed separately, in sequence, or in conjunction are use of a resistive layer, eg, clay; use of a conductive layer, involving wick action; and bioengineering, using a special plant cover.

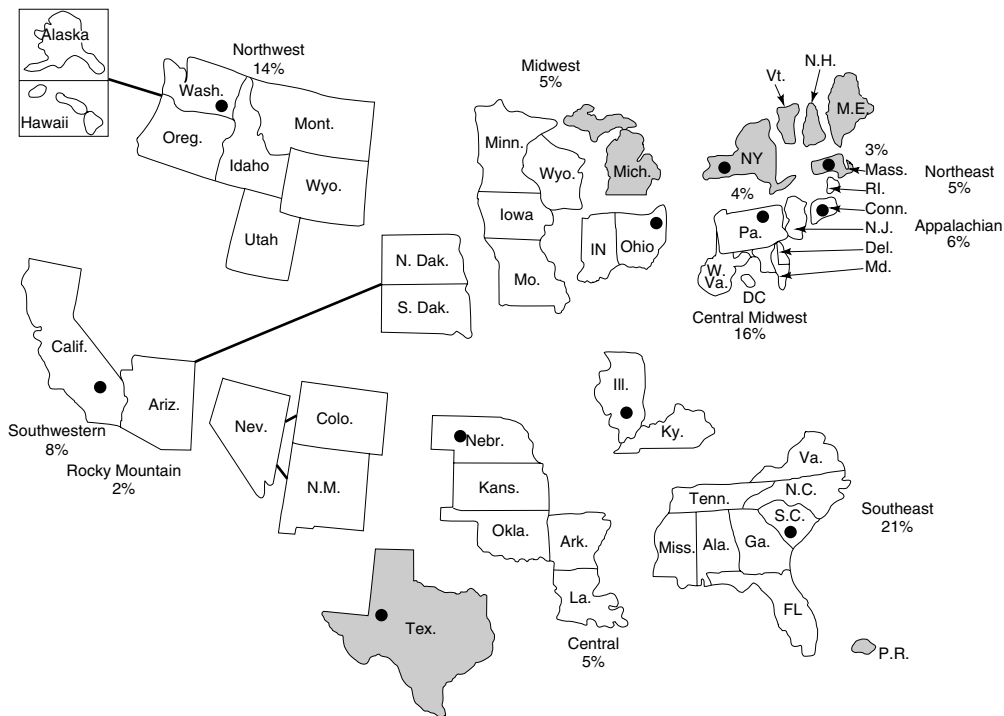


Fig. 3. Interstate compacts for low-level waste management where (○) represents unaffiliated states and (●), host sites. The percentages of total U.S. LLW are also given. Courtesy of Battelle Press.

4.3. Transuranic Waste. Transuranic wastes (TRUs) contain significant amounts [$>3700 \text{ Bq/g}$ (100 nCi/g)] of plutonium or other transuranic elements such as americium or curium. These wastes have accumulated from nuclear weapons production at sites. Since 1999, TRU has been disposed of in the Waste Isolation Pilot Plant (WIPP) near Carlsbad, New Mexico. The geologic medium is rock salt, which has the ability to flow under pressure around waste containers, thus sealing them from water. As of 2006, WIPP was the only operating geological disposal facility in the world.

5. Environmental Issues

Progress toward the disposal of nuclear wastes has been slow for several reasons. One is public opposition, epitomized by the not-in-my-back-yard (NIMBY), syndrome. Much of the public is fearful of nuclear reactors, radioactivity, and radiation (32). Concern was heightened by the Three Mile Island and Chernobyl accidents. Others in the public oppose siting of disposal facilities on economic grounds, believing that business, tourism, and property values would be jeopardized. Some people are concerned with equity when a local population must host a site that benefits the state, region, or nation. Efforts by the nuclear community

to maintain positive dialogue are often successful, especially when the public is involved in discussions at an early stage of a project and is fully and correctly informed of plans and developments (33,34). Educational material on radioactive wastes is available (35,36).

There is an enormous amount of research literature about nuclear waste management, and nuclear scientists and engineers generally are convinced that wastes can be disposed of safely. Delays in disposal can also be attributed to an accepted national policy and to the administration of programs. Until the late 1980s, treatment and disposal of defense wastes were of lower priority than accumulating the needed weapons material. Efforts in the 1970s to find a suitable site for HLW disposal in salt were unsuccessful. Changes in national policy and governmental plans since the 1970s, including the demand for more information about geology and hydrology, have delayed projects. The costs associated with long-term storage of spent fuel or low-level waste are high. Moreover, the nuclear industry recognizes that continued and extended use of nuclear power depends on acceptance of waste treatment by the public and the financial community, which depends on the demonstrated ability of industry and government to meet challenges in a safe and an economical manner (37,38).

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