Chapter 2

RADIOACTIVE WASTE

This chapter is intended to provide readers who have not previously considered the radioactive waste problem with the information they will need to understand the analysis of waste management and regulation that follows. The discussion is written with the non-technical reader in mind.

Radioactivity

Radiation is the emission and propagation of energy through matter or space. Radioactive materials emit energy in the form of electromagnetic radiation, such as gamma or X-rays, or in the form of fast moving sub-atomic particles such as alpha-particles (the nuclei of heluim atoms) and beta particles (electrons), both of which carry an electric charge, and neutrons, which carry no electric charge. As radiation penetrates matter, it interacts with its environment, and energy is tranferred to the surrounding atoms resulting in their ionization. If the radiation is interacting with organic tissue, the ionized atoms acquire new and different properties and may break down or enter into abnormal chemical combinations which may cause the decomposition or synthesis of complex molecules. The net effect is to destroy living cells and to damage the exposed tissue.

The energies of the different emissions listed previously vary widely and so do their penetrating powers. For example, alpha radiation is not very penetrating compared with gamma, and, to a lesser extent, beta. Unlike these two, therefore, it does not constitute a major hazard to man if the radiation source is external. If the

source of radioactivity is inhaled or ingested, however, then all three types of radiation are important, because of the close proximity of living tissue.

We are concerned about radioactive waste because it is a potential radiological hazard to man and other forms of life. A scale against which our concern can be measured is the fact that we all live in a radioactive environment. The biosphere--that thin envelope of soil, water and air which contains and sustains all life on Earth--receives radiation from outer space and from many areas in the Earth's crust. Since the beginning of this century, man has begun to add to this naturally occurring "background" radioactivity. Medical x-rays, normal nuclear power plant operations, and nuclear weapons tests are all sources of ionizing radiation. Depending on where a person lives and what he does, he is exposed to more or less radiation. A person living in Denver, Colorado which is located near uranium ore deposits at an elevation of about 1 mile above sea level, receives more radiation than a person working in a normally functioning nuclear power plant on the James River in Virginia and living nearby.

When a radioactive nucleus* emits radiation--usually an alpha or beta particle or a gamma ray--it "decays" to another nucleus which itself may or may not be radioactive. A sequence of decays involving two or more radioactive nuclei is called a "decay chain". Clearly, every decay chain terminates with a stable, non-radioactive nucleus.

Each type of radioactive nucleus, or radioisotope, has a

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*The nucleus is the positively-charged core of an atom, composed of protons and neutrons.

characteristic fractional decay rate. Of course, it does not make sense to talk about the "fractional decay rate" of an individual nucleus, since at any instant the nucleus in question either will or will not have decayed. But the concept becomes meaningful if, as is invariably the case, we are dealing with a large number of nuclei of the same type. In this situation we can legitimately speak of a collective fractional decay rate. A commonly used measure of decay rate is the radioactive half-life. This may be defined as the length of time required for half of the nuclei in a sample of a radioisotope to decay to another nuclear form.

The half-lives of different radioisotopes span an enormous range from millionths of a second to billions of years. After a period of time equal to 10 half-lives, the radioactivity of a radioisotope has decreased to 0.1 percent of its original level. Table 2.1 indicates the half-lives of some of the major constituents of radioactive waste. In order to calculate the amount of radioactivity from a particular source, it is not sufficient to know only the half-life of the radioisotope involved. It is also necessary to know the quantity present. The level of radioactivity, which is the product of the quantity of a particular radioisotope and the fractional decay rate, is measured in curies.*

We need to know more than the number of curies, however, in order to estimate the biological significance of a volume of material

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*A Curie is a unit which measures the rate of nuclear decay. One curie is defined as being equal to 3.7×10^{10} nuclear decays (or disintegrations) per second. It should be noted that this unit does not distinguish either the energy or type of radiation that is emitted during a decay.

TABLE 2.1 - Half-Lives of Some of the Major Constituents of Radioactive Waste

Radionuclide	Half-life (Years)
Americium - 241	460
Americium - 242	150
Cesium - 135	2,000,000
Cesium - 137	30
Curium - 242	.45
Curium - 243	32
Curium - 244	18
Iodine - 129	16,000,000
Krypton - 85	10.8
Neptunium - 237	2,100,000
Plutonium - 239	24,000
Plutonium - 241	13
Radium - 226	1,600
Strontium - 90	29
Technicium- 99	200,000
Thorium - 230	76,000
Tritium	13

that contains one or more radioisotopes, perhaps mixed with non-radioactive materials. All radioactivity is not the same. As we have seen, different radioisotopes emit different forms of radiation at different energies as they decay.

Furthermore, different living organisms and different organs within the same organism all have varying degrees of sensitivity to radiation. Since many effects of ionizing radiation are cumulative, individual exposure histories are important, and these, together with individual radiation tolerance levels, can vary significantly. For internal sources of radiation, the physical and chemical form of the radioisotope and its route of intake are also important.

If a human being is exposed to excessive amounts of radiation, depending on the circumstances, the harmful effects may be immediate death, life shortened by radiation-induced cancer, radiation-induced genetic change which may affect subsequent generations, or temporary ill-health followed by complete recovery. It is important to note that excessive exposure to harmful levels of ionizing radiation may be caused by contact with a particular radiation source or multiple contacts with many sources. The rate of absorption of radiation is also important, since damaged tissue has more chance to recover from radiation effects if the exposure takes place over an extended period rather than during a sudden burst.

How then do we protect ourselves from the potentially harmful effects of excessive radiation? Although the relation between the magnitude of radiation dose* and the resulting effect on human

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*Dose may be defined as the quantity of energy imparted to a mass of material exposed to radiation. A dose unit is the rad, which is equal to one hundred ergs of absorbed energy per gram of absorbing material. Another dose unit is the rem (roentgen equivalent man) which is the dose of any ionizing radiation that will produce the same biological effect as that produced by 1 roentgen of high voltage X-radiation. It is related to the rad by a quality factor, the relative biological equivalent (RBE).

health is difficult to determine, the government nevertheless establishes dose limits, or Maximum Permissible Doses, which are standards for radioactive emissions from routine activities. On the basis of aggregate values for the breathing rate and the daily consumption of drinking water for an average person, it is possible to calculate the dose to each body organ that would be likely to result from a given, continuous concentration of a particular radionuclide in air or water. These calculations, together with the Maximum Permissible Doses, allow the critical or most sensitive organ to be identified for each radionuclide and method of intake. It is also possible to calculate, for each radionuclide, the concentration in air and water that, if continuously maintained, would result in the Maximum Permissible Dose to the critical organ. This is called the Maximum Permissible Concentration (MPC).

The calculated MPC values are, therefore, a measure of the radiotoxicity of a nuclide and are frequently used in calculations of radiological hazard. The MPC measure is imperfect, however. It accounts only for radiation exposure resulting from breathing contaiminated air and drinking contaminated water. In particular, it does not take into account the effects of biological reconcentration of a radionuclide that has been previously released to the environment. Therefore, a reliable estimate of the radiation dose to the population resulting from the presence of a particular radioactive source requires a detailed study of the particular environment and its demographic characteristics in order to identify the critical pathways to population exposure.

Post-Fission Radioactive Waste

"Radioactive waste" may be defined broadly as being waste material that is contaminated or possibly contaminated by radioactive isotopes. More precisely, radioactive waste is residual material that is removed from the nuclear fuel cycle and held because its release would constitute a hazard to man or the environment. Such waste does not include radioactive emissions, within prescribed limits, which occur during the normal operations of various nuclear facilities. As we use the term, "waste" includes residual material from which certain materials may be sooner or later recovered and recycled.

Post-fission radioactive waste is generated by nuclear fission in power reactors which produce electricity for commercial use, and also in reactors which produce plutonium for nuclear weapons and in propulsion reactors for submarines, missile cruisers, and aircraft carriers. This is the commercial and military post-fission waste that is the subject of the study.

Post-fission radioactive waste may be differentiated into various categories such as high-level (HL), transuranic* contaminated low-level (TRU), and non-TRU waste. Such a categorization does not necessarily provide a key to potential hazards, however, since these also depend on external factors.

HL waste is composed of hundreds of radioisotopes, some in trace amounts and some in very high concentrations, some with short and some with extremely long half-lives. Various isotopes are typi-

*Transuranic means having an atomic number greater than uranium, that is, greater than 92 (e.g., neptunium, plutonium, americium, curium, etc.).

cally gamma or beta or alpha emitters. HL waste is thus a veritable witches' brew, seething with radioactivity. Because of the long half-lives of some of the radioisotopes it contains, HL waste would, if it escaped into the biosphere, constitute a radiological hazard for hundreds of thousands, perhaps a million years. (See Figure 2.1 showing HL waste radioactivity over time.)

TRU waste contains numerous radioisotopes in much lower concentrations than HL waste. It is, however, especially troublesome because it is contaminated with transuranic elements, including plutonium, which have very long half-lives. Indeed, for a given amount of electricity generated with nuclear fuel, roughly the same amount of plutonium will ultimately emerge in TRU as in HL waste.*

Low-level radioactive waste that is not contaminated with transuranic elements is not as hazardous in the long-term as TRU waste. With respect to post-fission waste, it should be recognized that dividing low-level waste into TRU and non-TRU categories, or deciding what is "transuranic contaminated," is a question of judgment based on a complete interpretation of complex data. This report, as noted previously, focuses on HL and TRU wastes.

Post-fission waste originates from spent fuel assemblies that are routinely discharged from a nuclear power reactor. The irradiated fuel rods in these assemblies contain fission products, uranium, whose original uranium-235 content has been depleted by fission, and plutonium, which is produced by neutron capture in uranium-238

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*Assuming that 99.5 per cent of the plutonium is extracted at the reprocessing plant, and recycled in fuel to reactors.

After discharge from the reactor, the materials constituting spent fuel assemblies move through a sequence of operations. Various categories of radioactive waste emerge at different points in the sequence. The operations are outlined briefly below and then discussed further.

Temporary Spent Fuel Storage

Irradiated fuel assemblies are temporarily stored in a cooling pond next to the reactor while radioactive decay proceeds sufficiently to permit safe transport. No plant for reprocessing spent fuel from commercial power reactors is presently operating in the U.S.. Consequently, the available storage capacity of existing cooling ponds at operating power reactors is rapidly decreasing. As a result, additional temporary storage capacity, either at various reactors or at a central location, will be required soon.

Spent Fuel Transport

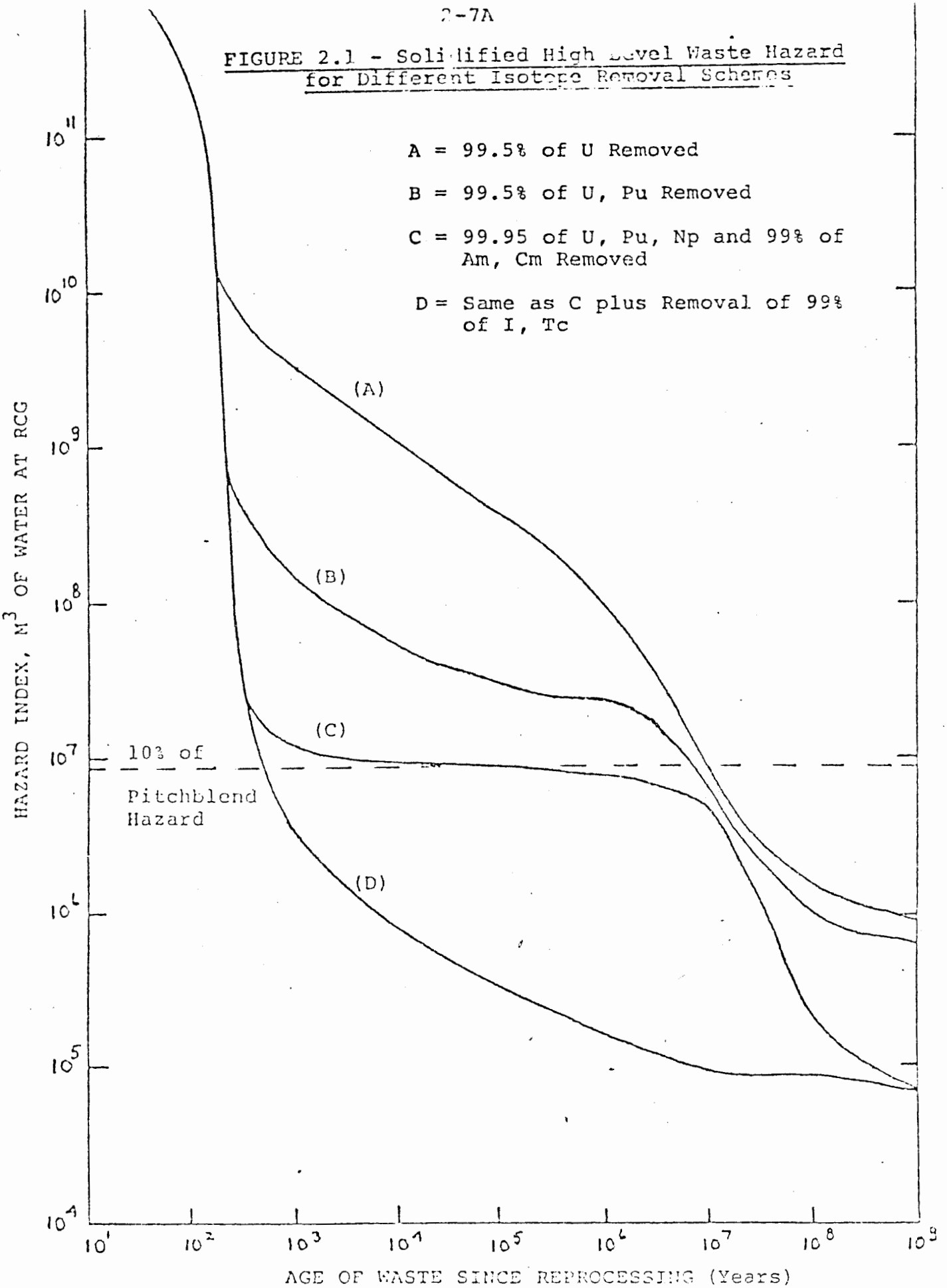
After the cooling period, commercial spent fuel rods, which are still highly radioactive, are placed in massive containers for shipping from the storage pond at the reactor site to a reprocessing plant. In the case of military plutonium production, the production reactor and reprocessing plant are located on the same site.

Reprocessing

Following arrival at a reprocessing plant, the assemblies are disassembled and the spent fuel rods are chopped up. The residual mixture of fuel materials and fission products is dissolved and sent through the plant where a large number of chemical separations are performed. Plutonium and depleted uranium are recovered.

In the commercial nuclear power industry, the depleted uranium

FIGURE 2.1 - Solidified High Level Waste Hazard for Different Isotope Removal Schemes



From Kerry D. Dance, "High-Level Radioactive Waste Management: Past Experience, Future Risks, and Present Decisions," Paper prepared for the Resources and Environment Division of the Ford Foundation, April 1975, p. 21.

Temporary Storage

HL waste emerges in the course of reprocessing in liquid form. The liquid is stored temporarily in tanks adjacent to the reprocessing plant.

While HL waste emerges only as a liquid and primarily at the first plutonium and uranium extraction stage of the reprocessing plant, TRU waste emerges in various forms--liquid, combustible and non-combustible solids--at other points in the nuclear fuel cycle in addition to the reprocessing plant. The main sources of TRU waste are: the fuel cladding hulls and assembly structures from the head-end of the reprocessing plant; the residuals left from the conversion of liquid plutonium nitrate to plutonium oxide powder (commercial use) or to plutonium metal (weapons use); and the spent process material, general trash and failed equipment left from reprocessing, commercial mixed oxide fuel fabrication and military weapon component fabrication. TRU waste is collected, temporarily stored and sometimes mixed with other less toxic wastes at these various fuel cycle stages.

Waste Treatment

Depending on its composition, radioactive waste is treated in various ways: gases may be dissolved, liquids may be solidified, and solids may be incinerated (resulting in gas and less solid). In general, waste treatment is designed to make the waste easier or safer to handle subsequently, but every treatment method has its own set of costs and risks, and almost always results in the generation of secondary waste streams.

Waste Transport

Following treatment, radioactive waste is transported to

can be re-enriched in the uranium-235 isotope, or it can be mixed with plutonium. In either case, the depleted uranium would be recycled in power reactor fuel. The recovered plutonium can be blended with uranium in mixed oxide fuel for recycling in existing light water reactors, or it can be stored for later use as fuel in breeder reactors.

In the weapons program, the recovered plutonium is used in the manufacture of nuclear warheads. In the naval propulsion program, it is unclear what subsequent use, if any, is made of the uranium and the relatively small amounts of plutonium which are recovered form reprocessing.

The radioactive waste from a reprocessing plant includes liquid HL waste, much larger volumes of liquid low-level waste (generally with no more than trace amounts of transuranic contamination), and solid waste -- cladding hulls, failed equipment, spent process materials, trash, etc, -- some of which contains appreciable amounts of the transuranic elements.

One plant for reprocessing commercial power reactor fuels was previously in operation at West Valley, New York. The plant has been temporarily shut down. The possiblity exists that it may not be reopened. Another, larger commercial reprocessing plant is currently under construction at Barnwell, South Carolina.

Plants for reprocessing production reactor fuel for weapons are located at Savannah River, South Carolina, and Richland, Washington. The reprocessing of naval reactor fuel takes place at Idaho Falls, Idaho.

a location where it is more or less permanently disposed of. The importance of transport depends on the method of permanent disposition. No transportation is involved if HL waste is solidified in the bottom of a temporary storage tank and left there indefinitely. Long distance transport involving railroad or truck and ship is necessary for either seabed or ice sheet disposal.

Permanent Disposition

A wide variety of methods for permanent disposition of HL waste is being considered. The most practical appear to be either retrievable storage on or near the surface or geologic disposal in suitable formations either deep underground or under the ocean floor. Other methods under study are ice sheet disposal and, if partitioned, nuclear transmutation or extraterrestrial elimination of the extracted long-lived actinides.

It is generally stated that no method for permanent HL waste disposition has been adopted yet. It could be argued, however, that in the case of the majority of military HL waste a decision may effectively already have been taken and implemented. In this case, liquid waste stored temporarily in tanks has been solidified in situ. It now appears that the task of exhuming these solids will be very difficult and costly, and for this reason the waste might be left there indefinitely.

Some TRU waste has been disposed of, essentially irretrievably, in ocean dumping grounds. Other waste has been deposited in shallow burial grounds. Not all of these land burials will permit retrieval at reasonable cost. The question of the permanent disposition of TRU waste to be generated in the future is unresolved.

Radioactive Waste Quantities

The following is a brief review of the amounts of radioactive wastes that have already been generated in the U.S., together with some predictions of how these amounts will increase over the next twenty to thirty years. The discussion is presented so that it is possible to compare existing quantities of military waste with those produced in the commercial nuclear power program.

High-Level Waste

As a result of military activities from the middle 1940's to the present, the former AEC and its successor, ERDA, have generated about 215 million gallons of liquid HL waste.* Solidification programs have been in operation for some years at the three sites at which this waste is stored: the Hanford reservation near Richland, Washington, the Savannah River Plant in South Carolina, and the Idaho National Engineering Laboratory, near Idaho Falls in Idaho. As a result, over 80 percent of the original liquid has been solidified. The accompanying volume reduction has meant that, as of January 1, 1976, there was an HL waste inventory of about 75 million gallons, half of which was in solid form.

More than 70 percent of this waste is currently stored at Richland, with about 25 percent at Savannah River and 3 percent at Idaho Falls. It is estimated that by the early 1980's when most

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*With the passage of the Energy Reorganization Act of 1974 the former AEC was fissioned into two parts--NRC (nuclear regulation) and ERDA (nuclear and non-nuclear energy research, development and demonstration).

of the waste will have been solidified, there will be a total of more than 450,000 tons of residual HL solids at these three sites.

In contrast, the commercial nuclear power industry has until now only produced 600,000 gallons of HL waste. All of this waste is still in liquid form and is stored at the site of the Nuclear Fuel Services reprocessing plant at West Valley, New York.

Currently, HL waste from ERDA programs is being generated at an annual rate of 7.5 million gallons, but it is not known how long this rate of production will continue. Predictions must be based upon an assumed plutonium demand rate for nuclear weapons. Predictions of future generation rates of commercial HL waste must be based on assumed nuclear power growth rates, and are therefore also subject to considerable uncertainty. It is estimated, however, that the commercial nuclear power industry in the U.S. will have generated 60 million gallons of liquid HL waste by 2000, and that not until 2020 will the commercial power industry have produced as much liquid HL waste as has already been produced by U.S. military programs.

According to recent calculations, the volume of all the solidified HL waste produced by the commercial nuclear power industry through 2000 will be equivalent to a cube 70 feet on each side, whereas the equivalent cube for the military HL waste will have a side measuring approximately 220 feet. Of course, this calculation should not be interpreted to mean that these volumes are all that will be required to store the waste. Much larger storage volumes will be necessary. But the calculation does indicate that the main problem in safely managing HL waste will be one of confining the radioactivity, rather than of finding

enough storage space.

A volumetric comparison alone is not sufficient when comparing military and commercial wastes. One ton of spent fuel from a plutonium production reactor produces a larger volume of HL waste than does one ton of spent commercial power reactor fuel. Furthermore, fuel in plutonium production reactors only receives about a tenth as much irradiation exposure before discharge as commercial power reactor fuel. The amount of radioactivity in spent production fuel is therefore correspondingly lower. Although such information is classified, and in fact may not even be known, one estimate is that existing military HL waste contains 5 billion curies of radioactivity. Nuclear power projections indicate that the inventory of radioactivity in commercial HL waste will not reach that level until the 1990's.

The conclusion that clearly emerges is that the quantity of military HL waste far exceeds the amount generated by the commercial nuclear power industry. Furthermore, this discrepancy is likely to continue at least until the end of the century.

TRU Waste

Through June 1974, 42 million cubic feet of military low-level waste had been buried at five principal land burial sites. Through 1973, over 9 million cubic feet of commercial low-level waste had been buried at six commercially licensed sites.

Until April 1970 there was no distinction made between TRU and non-TRU waste for burial purposes. At that time, TRU waste was defined as waste with a concentration of transuranic activity

greater than 10 nanocuries per gram.* It was also decided that all TRU waste subsequently delivered to Federal burial grounds should be stored above ground in retrievable form. About 952 kilograms of plutonium are contained in the military TRU waste buried at the five major Federal burial sites. Of this amount, 740 kilograms were buried before 1970, and the remaining 212 kilograms have been stored retrievably. Through 1973, TRU waste containing some 80 kilograms of plutonium has been buried at the six commercial sites. There is likely to be no further shallow land burial of commercial TRU waste if proposed regulatory changes are adopted. All future military and commercial TRU (and that which is stored presently) will probably be disposed of more carefully.

Military activities are currently generating low-level solid waste at the rate of 1.3 million cubic feet per year. (The fraction of this total generation rate that is TRU contaminated has not yet been established.) It is expected that this rate will gradually decrease in the future. Recent projections of commercial waste generation indicate that by 2000 there will be over 50 million cubic feet of TRU waste, including about 1 million cubic feet of spent fuel cladding waste.

In addition to the above quantities, unspecified amounts of low-level waste, military and commercial, have been placed in cannisters and discharged into the sea at U.S. dumping sites off

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*One nanocurie = one billionth of a curie.

the Atlantic and Pacific coasts. The proportion that is TRU contaminated is unknown. Ocean dumping of U.S. low-level waste was suspended in 1970. However, other countries are continuing this practice.

The existing amount of military TRU waste thus exceeds the current amount of commercial waste, but the dominance is not as great as in the case of HL waste.

Radioactive Waste Leakage

AEC, ERDA's predecessor as manager of military waste, intentionally discharged or accidentally released large quantities of HL and TRU waste. Between 1956 and 1958, about 31 million gallons of radioactive waste containing 1.3 million curies (excluding strontium and cesium, but including plutonium) was poured into soil on the Hanford reservation. In addition, the Hanford Plutonium Finishing Plant has discharged liquid effluent containing large quantities of plutonium to sub-surface trenches that are not isolated from the soil. Indeed, in the case of one particular trench, over 100 kilograms of plutonium was released in this way, creating a potentially severe radiological hazard and the possibility of a spontaneous fission reaction.* Roughly 740 kilograms of plutonium have been buried irretrievably at five principal Federal burial sites. The liquid HL waste tank farms at the Hanford reservation have developed eighteen leaks amounting to the loss of over 430,000 gallons of waste.** The largest leak so far occurred in 1973 and

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*Note that the HL waste generated by a typical U.S. power reactor (i.e., a 1000 MWe light water reactor) during its entire operating lifetime (about 30 years) would contain about 30 kilograms of plutonium.

**The 430,000 gallons contained 50,000 curies of radioactivity including 4 curies of plutonium.

caused the escape of 115,000 gallons of HL waste into the surrounding soil. Eight leaks have occurred at the Savannah River Plant, but only one of about 100 gallons is known to have resulted in contamination of the surrounding soil. These leaks have neither killed nor injured any one to date. Nonetheless their hazard will remain for hundreds of thousands of years.

Potential Causes of Harmful Effects

Many events may cause the release of radioactivity from waste management activities. They can be classified as accidents due to human failure, intentional acts, or natural phenomena.

Accidents can be categorized according to their causes--usually either inadequate construction or improper operation. An example of the first category would be the leaks which occurred in carbon steel, military liquid HL waste tanks. The surrounding ground and the food chain will delay, distribute over time, and, if there is enough time, instigate the actual biological consequences of these leaks.

Intentional acts against waste management facilities may be characterized as acts of either warfare or sabotage. For instance, the temporary HL waste tanks adjacent to a reprocessing plant might be sabotaged for the purpose of achieving long-term local contamination. Surface waste management activities might be targeted with nuclear weapons to increase the degree, duration and coverage of contamination after a nuclear war, but the same weapons would be much more effective if they were used initially in the destruction of cities or military targets.

Finally, various natural events can abruptly or chronically initiate releases of radioactive materials to the environment. Abrupt events include floods, earthquakes, volcanoes, and, conceivably, meteors. They may result in immediate environmental contamination or they may expose the final waste form to slow but continual degradation and subsequent seepage into the biologically active media of soil and water. In chronic release scenarios, the essential question is how much waste will reach the soil or watershed during its period of toxicity. Chronic events include the possible shifts of water tables and river courses, the glacial erosion caused by Ice Ages, slow but continual geologic faulting, leaching processes, and surface ice flows and underground lake migration in Antarctica.

Conclusions

From the preceding outline of the basic facts of the post-fission radioactive waste problem in the U.S., several conclusions may be drawn:

1. As a consequence largely of nuclear weapons programs, the U.S. government has already committed itself to a radioactive waste management responsibility of major proportions which is still growing.

2. Because of the maturing nuclear power industry, the U.S. is generating a rapidly increasing volume of radioactive waste. The bulk of this waste remains in the form of spent fuel elements in temporary storage ponds at commercial reactor sites.

3. Radioactive wastes must be safely managed to prevent contamination of the biosphere and possible radiological hazards to life.

4. Thus far, the U.S. government's record of management has been marred in a sufficient number of instances to be a cause of concern.

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An inevitable legacy of our nuclear age, radioactive waste constitutes a potential Nth century hazard.