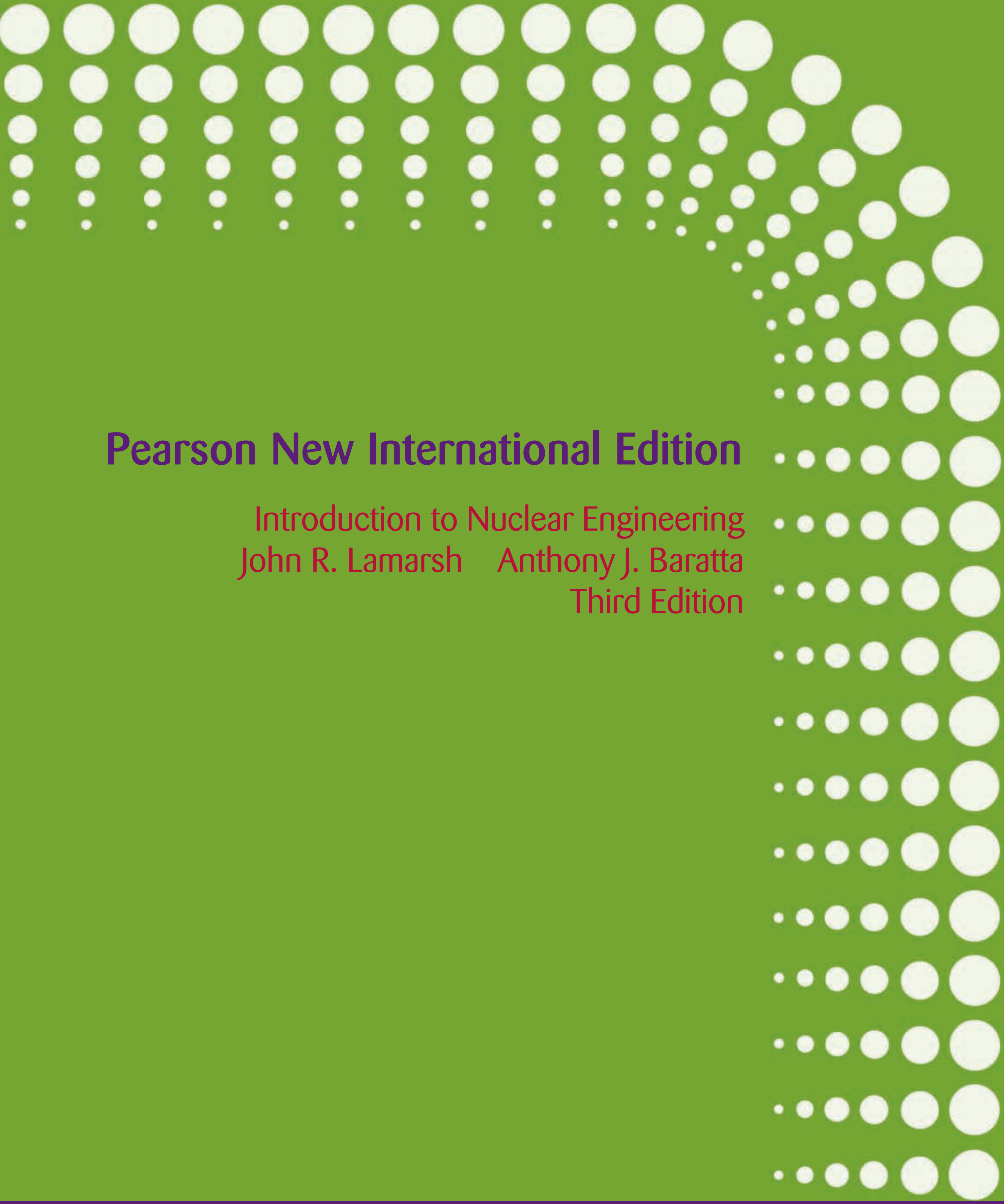

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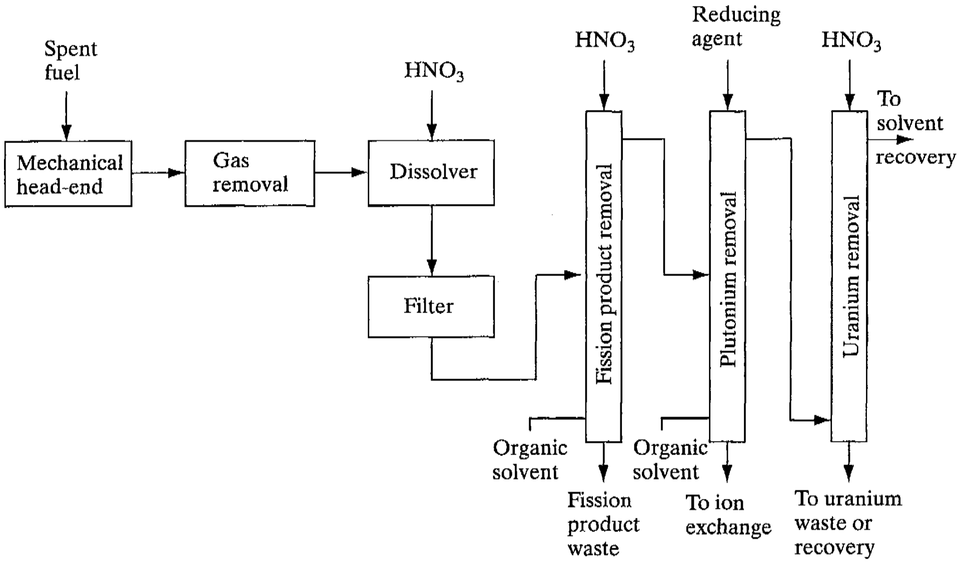

Figure 4.55 Simplified flow diagram of a PUREX reprocessing plant.

whereas the aqueous solution exiting at the bottom holds most of the fission products and very little uranium or plutonium.

The organic solution passes next into a second column, where it counterflows against a dilute solution of a chemical-reducing agent (a ferrous compound is often used), which reduces the plutonium to the 3^+ state, but leaves the uranium in the 6^+ state. Since the plutonium is no longer soluble in the TBP, it passes into the aqueous solution before leaving the column.

The uranium is stripped from the organic solvent in a third column, where it passes into a counterflowing stream of dilute nitric acid. The solvent leaving the top of the column, from which most of the plutonium, uranium, and fission products have now been removed, is piped to a recovery plant for purification and reuse. The uranium exits the column in aqueous solution.

To further purify the uranium and plutonium fractions, their respective solutions can be processed through additional extraction columns. However, the plutonium is often purified and concentrated by ion exchange. This process involves passing the plutonium solution into an ion exchange resin and then eluting the plutonium with dilute acid. The concentration of the purified plutonium can then be increased by partially evaporating the solution, taking care not to approach criticality. This is the usual form of the plutonium output from a fuel reprocessing plant—a highly purified solution of plutonium nitrate. It is an easy matter to transform the plutonium to the oxide PuO_2.

4.9 RADIOACTIVE WASTE DISPOSAL

Radioactive wastes in several different forms are produced at various points in the fuel cycle of a nuclear power plant: during the mining of uranium, the manufacture of the fuel, the operation of the reactor, and the processing and recycling of the fuel (if this is part of the cycle). Wastes are also produced when the plant is ultimately decommissioned and dismantled. The nuclear power industry is not the sole purveyor of radioactive wastes, however. Hospitals have become a major source of such wastes due to the widespread use of radiopharmaceuticals in medicine. Radioactive wastes are also a significant by-product of nuclear weapons programs.

It is usual to classify waste in four categories. *High-level waste* consists of spent fuel, if this is discarded as waste, and any wastes generated in the first stages of a fuel reprocessing plant (since this waste contains the bulk of the fission products). *Transuranic (TRU) waste* consists mostly of the isotopes of plutonium at concentrations in excess of 10^{-9} Ci/g; TRU wastes are generated by fuel reprocessing, plutonium fuel fabrication, and manufacturing of nuclear weapons. *Low-level waste* contains less than 10^{-9} Ci/g of TRU nuclides. It also includes material that is free of TRU and requires little or no shielding, but it is still potentially dangerous. *Mine and mill tailings* consist of residues from uranium mining and milling operations; such residues contain low concentrations of naturally occurring radionuclides.

A perusal of the chart of nuclides reveals that the vast majority of the several hundred fission products are very short-lived. Only five have half-lives between 1 and 5 years; two–namely, ^{90}Sr and ^{137}Cs–have half-lives of about 30 years, and three—^{93}Zr, ^{129}I, and ^{135}Cs–have half-lives in excess of a million years, and hence are effectively stable. In 100 years, the activity of a 5-year nuclide decreases by a factor of 10^6, and the shorter lived nuclides disappear altogether. Over the long-term, therefore, the fission product activity of high-level waste is due only to ^{90}Sr and ^{137}Cs. The latter radionuclide decays into stable ^{137}Ba. However, ^{90}Sr decays to ^{90}Y, which decays with a 64-hr half-life to stable ^{90}Zr. Thus, high-level waste ultimately contains three fission products, ^{90}Sr, ^{90}Y and ^{137}Cs.

The half-lives of many of the TRU nuclides tend to be considerably longer. For instance, the half-life $T_{1/2}$ of ^{239}Pu is 24,000 years. Therefore, the activity of these nuclides dies off more slowly than that of the fission products. In the spent fuel from a typical LWR, the TRU activity exceeds the fission product activity after approximately 700 years.

The total high-level activity from spent fuel, including both the fission products and the TRU nuclides, depends on the nature of the reactor fuel cycle. With the once-through cycle, the activity of the fuel persists for hundreds of thousands of years owing to the presence of TRU material. However, with a closed cycle,

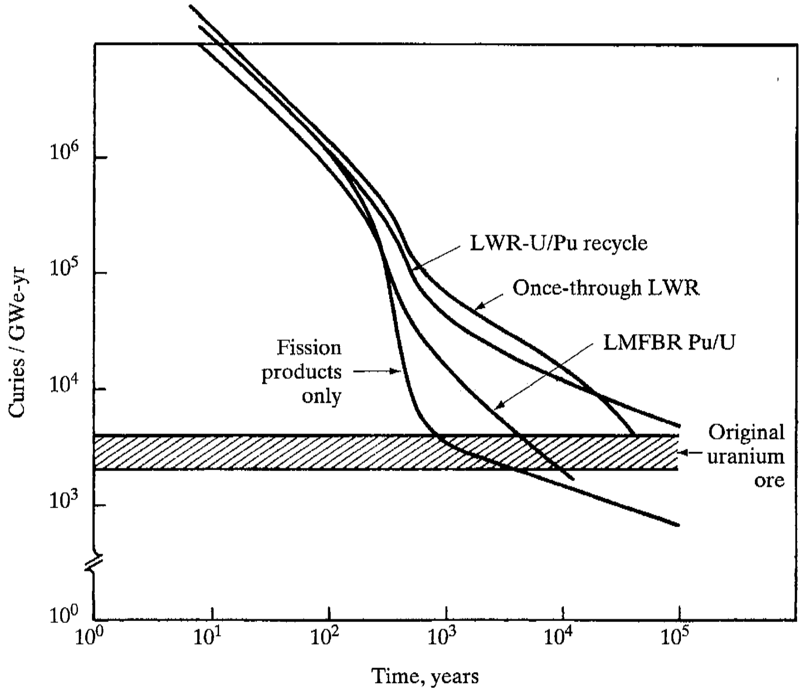

Figure 4.56 High-level waste activities from different reactor fuel cycles. (Based on *Nuclear Proliferation and Civilian Nuclear Power,* U.S. Department of Energy report DOE/NE-0001/9, Volume 9, 1980.)

the plutonium isotopes are returned to the reactor where, on fissioning, they are transformed into short-lived fission products.

This situation is illustrated in Fig. 4.56, where the activity of high-level waste generated in 1 GWE year is shown for the LWR with and without recycling; for the LMFBR (with recycling, of course) and for a special case in which LWR spent fuel is reprocessed, the fission products are treated as waste and the plutonium is merely stored. Except in the early and late years, the activity in this last cycle is due almost entirely to ^{90}Sr, ^{90}Y, and ^{137}Cs. Also shown in the figure is the range of activities of the uranium ore required to produce the original fuel. It is observed that the high-level activity associated with the fuel cycles approaches within an order of magnitude of the ore in about 1,000 years.[18]

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[18] A more fair comparison of ore and high-level waste should take into account the relative toxicity of the heavy elements, which decay in large part by α-emission, with that of fission products, which emit β-rays. The toxicity of α-emitters is about 10 times greater than that of β-rays (see Section 9.2).

Most of the high-level waste in the world today is a by-product of nuclear weapons programs. For the most part, pending permanent disposal, this waste is either stored in liquid form in large tanks or is being solidified to reduce the possibility of leakage. Increasing amounts of high-level waste are also accumulating in the spent-fuel pools of nuclear power plants. Commercial reprocessing of spent fuel is occurring in a number of foreign countries. However, because of proliferation concerns, the United States is not participating.

How commercial high-level waste is disposed of clearly depends on whether or not spent fuel is reprocessed. With no reprocessing, the fuel may be disposed of intact. Presumably this would be done by placing the spent fuel assemblies in suitable containers and burying these containers in some stable geological setting. Historically, stable rock formations have been considered for this purpose.

If the fuel is reprocessed and the plutonium is recycled (or stored), the disposal problem becomes more manageable and susceptible to unique technological solution. Since fission products represent a small fraction of the mass of the fuel, reprocessing substantially reduces the volume of the waste. The waste, in liquid form, can then be calcinated—that is, dried at high temperature; mixed with *frit*, the substance from which glass is made; and then vitrified—that is, made into glass. Another newer technique involves surrounding beads of waste with layers of ceramic as in the fabrication of HTGR fuel. These and similar methods for solidifying the waste effectively immobilize the radioactive particles, something that cannot be done as easily with unprocessed spent fuel. The glass or ceramic complexes are finally placed in cannisters and deposited in stable geological formations.

If a canister holding either a whole fuel assembly or solidified waste should disintegrate, even soon after its emplacement in a repository, there is good reason to believe that the fission products and TRU nuclides would not diffuse far into the environment. Strong support for this contention is furnished by what has become known as the *Oklo phenomenon.* Oklo is the name of a uranium mine in the African nation of Gabon, where France obtains much of the uranium for its nuclear program. When uranium from this mine was introduced into a French gaseous diffusion plant, it was discovered that the feed uranium was already depleted below the 0.711 w/o of ordinary natural uranium. It was as if the uranium had already been used to fuel some unknown reactor.

And so it had. French scientists found traces of fission products and TRU waste at various locations within the mine. These observations were puzzling at first because it is not possible to make a reactor go critical with natural uranium, except under very special circumstances with a graphite or heavy-water moderator, neither of which could reasonably be expected to have ever been present in the vicinity of Oklo. The explanation of the phenomenon is to be found in the fact that the half-life of ^{235}U, 7.13×10^8 years, is considerably shorter than the half-life of ^{238}U, 4.51×10^9 years. Since the original formation of the earth, more ^{235}U has therefore decayed than ^{238}U. This, in turn, means that the enrichment of natural uranium was greater years ago than it is today. Indeed, it is easy to show (see Problem 2.37) that about 3 billion years ago this enrichment was in the neighborhood of 3 w/o—sufficiently high to form a critical assembly with ordinary water, which is known to have been present near Oklo at that time. The relevance of the Oklo phenomenon to present-day disposal of radioactive wastes is that neither the fission products (identified by their stable daughters) nor the plutonium migrated from the Oklo site in the billions of years since the reactor was critical.

Compared with high-level waste, low-level waste represents more of a nuisance than a hazard. However, it comprises a much larger volume. A nominal 1,000 MWe LWR power plant produces the order of 100 m^3 of solid low-level waste, mostly contaminated laundry wastes, protective clothing, glassware, tools, containers, and so on, and somewhat higher level waste in the form of spent resins from reactor coolant demineralizers, air filters, and so on. Such wastes are normally placed in drums and shipped off-site to waste depositories, where the drums, several hundred per GWe-year, are buried. Liquid low-level wastes are usually solidified or retained for decay, diluted, and discharged to the environment.

In terms of total activity, a typical large hospital generates more low-level waste than a nuclear power plant. The bulk of this activity is due to ^{3}H and ^{14}C; the remainder consists of short-lived nuclides that quickly decay. Fortunately, the total amount of ^{3}H and ^{14}C originating in medical institutions is trivial compared with their natural production rates in the atmosphere from cosmic rays (see Section 9.7). At least in principle, these wastes can safely be incinerated, although, for political reasons, this procedure has not been generally adopted.

The major source of concern from uranium mining and mill tailings is the increased release of the radioactive gas radon—in particular, the isotope ^{222}Rn, which has a half-life of 3.8 days. This nuclide is one of the products in the long decay chain beginning with ^{238}U and is the immediate daughter of the decay of ^{226}Ra. Chemically, radon is a noble gas, and therefore it readily diffuses out of solid materials containing uranium or radium. Although radon does not present a health hazard, its longer lived daughters do, especially ^{210}Pb ($T_{1/2} = 19.4$ years). When these daughter products, formed by the decay of radon in the atmosphere, are inhaled, they may become attached to the tissues at the base of the bronchial network. Their subsequent decay can lead to lung cancer (see Chap. 9 for further discussion of this process). Disposal of such tailings is either by placement underground, the preferred but more costly method, or by covering the tailings with no less than 3 m of earth and then planting vegetation to prevent erosion.

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