
Nuclear Fuel Cycle

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9.1 Characteristics of the Nuclear Fuel Cycle

9.1.1 Types of Fuel Cycle

The *nuclear fuel cycle* is the progression of steps in the utilization of fissile materials, from the initial mining of the uranium (or thorium) through the final disposition of the material removed from the reactor. It is called a cycle because in the general case, some of the material taken from a reactor may be used again, or “recycled.”

Fuel cycles differ in the nature of the fuel used, the fuel’s history in the reactor, and the manner of handling the fuel that is removed from the reactor at the end of the fuel’s useful life (known as the *spent fuel*). For uranium-fueled reactors—which means virtually all commercial reactors—a key difference is in the disposition of the plutonium and other actinides that are produced in a chain of neutron captures and beta decays that starts with neutron capture in ^{238}U to produce ^{239}Pu (see Section 7.4).¹ The actinides are important because (1) some, especially ^{239}Pu , are fissile and can be used as nuclear fuel in other reactors or in bombs, and (2) many of the actinides have long half-lives, complicating the problems of nuclear waste disposal. The three broad fuel cycle categories are as follows:

¹ The *actinides* are the elements with atomic numbers Z greater than or equal to that of actinium ($Z = 89$). (The terminology is not uniform and, sometimes, actinium is not included among the “actinides.”) Neptunium ($Z = 93$), americium ($Z = 95$), and curium ($Z = 96$) are referred to as *minor actinides* in view of their low abundance in spent fuel compared to uranium ($Z = 92$) and plutonium ($Z = 94$). Elements with atomic numbers greater than 92 are termed *transuranic* elements.

- ◆ *Once-through fuel cycle.* This is sometimes called an *open fuel cycle* or a “throw-away” cycle. It is not really a cycle, in that the spent fuel is treated as waste when it is removed from the reactor and is not used further. The ^{239}Pu and other actinides are part of these wastes.
- ◆ *Reprocessing fuel cycle.* In the present standard reprocessing fuel cycle, plutonium and uranium are chemically extracted from the spent fuel. The plutonium is used to make additional fuel, often by mixing it with uranium oxides to produce *mixed-oxide fuel* (MOX) for use in thermal reactors. This provides additional energy and changes the nature of the wastes. In potential variants of the reprocessing fuel cycle, the minor actinides would also be extracted, and they and the plutonium would be incorporated in fresh fuel for fast reactors (see Section 9.4.3).
- ◆ *Breeding cycle.* For this cycle, the reactor is designed so that there is more fissile material (mostly ^{239}Pu) in the spent fuel than there was in the fuel put into the reactor (see Section 8.3). As in the reprocessing fuel cycle, the plutonium can be removed and be used in another reactor. With a sequence of such steps, fission energy is in effect extracted from a substantial fraction of the ^{238}U in uranium, not just from the small ^{235}U component, increasing the energy output from a given amount of uranium by a factor that could, in principle, approach 100.

It may be noted that uranium accounts for most of the mass of the nuclear wastes in the once-through cycle. It is separated out in the reprocessing and breeding cycles for possible reuse in reactor fuel.

At present, all U.S. commercial reactors and the majority of reactors worldwide are operating with a once-through fuel cycle, although some countries, particularly France, have large-scale reprocessing programs with use of plutonium in the form of MOX fuel. It should be noted, of course, that even in the once-through fuel cycle, the potential for eventually using the fuel in a reprocessing cycle remains until the fuel is disposed of irretrievably. No country is employing a breeder cycle at this time, although France appeared on the verge of attempting such a program with its Phenix and Superphenix reactors—but this effort has been abandoned, at least for the time being (see Section 8.3.3).

Although virtually all of the world’s commercial reactors have used uranium fuel, there is continuing interest in the use of thorium fuel.² In a thorium fuel cycle, the thorium (all ^{232}Th in nature) serves as the fertile fuel. Neutron capture and beta decay result in the production of ^{233}U , which has favorable properties as a fissile fuel. To start the thorium cycle, a fissile material such as ^{235}U or ^{239}Pu is needed, but once begun, it can be sustained if enough ^{233}U is produced to at least replace the initial fissile material. It is often argued that a thorium cycle is preferable to a uranium cycle, because if ^{233}U is ex-

² The Fort St. Vrain high-temperature, gas-cooled, graphite-moderated reactor in Colorado, which was shut down in 1989, is one of several exceptions to the exclusive use of uranium, having used thorium for part of its fuel [1, p. 41].

tracted from the spent fuel, it can be “denatured” by mixing it with natural uranium to make a fuel that cannot be used in a bomb. Bomb material could be obtained only after the isotopic separation of ^{233}U . In contrast, bomb material can be obtained from a uranium-fueled reactor by chemical separation of the plutonium (see Chapter 17). Isotopic separation is technically more difficult than chemical separation; thus, a thorium fuel cycle could be more proliferation resistant than a uranium fuel cycle unless, in the latter case, the plutonium is well protected from diversion or theft.

9.1.2 Steps in the Nuclear Fuel Cycle

A schematic picture of the fuel cycle is shown in Figure 9.1, which indicates alternative paths, with and without reprocessing [2]. The steps in the fuel cycle that precede the introduction of the fuel into the reactor are referred to as the *front end* of the fuel cycle. Those that follow the removal of the fuel from the reactor comprise the *back end* of the fuel cycle. At present, there is only a truncated back end to the fuel cycle in the United States, as virtually all commercial spent fuel is accumulating in cooling pools or storage casks at the reactor sites.

Implementation of a spent fuel disposal plan, or of a reprocessing and waste disposal plan, would represent the “closing” of the fuel cycle. This closing is viewed by many to be an essential condition for the increased use of nuclear power in the United States—and perhaps even for its continued use beyond the next several decades.

Key aspects of the fuel cycle will be surveyed in the remainder of this chapter. The fuel cycle will be discussed particularly in the context of light water reactors, in view of their dominance among world nuclear reactors. The main aspects are relevant to other types of reactor as well. A more extensive treatment of the crucial step of waste disposal will be given in Chapters 10–13.

9.2 Front End of the Fuel Cycle

9.2.1 Uranium Mining and Milling

Uranium Deposits in the Earth’s Crust

The concentration of uranium varies greatly among geological formations. The average concentration in the Earth’s crust is about 3 parts per million (ppm) by weight, but extremes extend from under 1 ppm to something in the neighborhood of 500,000 ppm.³

Uranium resources are widely distributed, with substantial uranium production in many countries, including Australia, Canada, Kazakhstan, Namibia,

³ For example, one deposit in Canada is identified as having zones of “over 50% U_3O_8 ,” which translates to over 42% uranium [3].

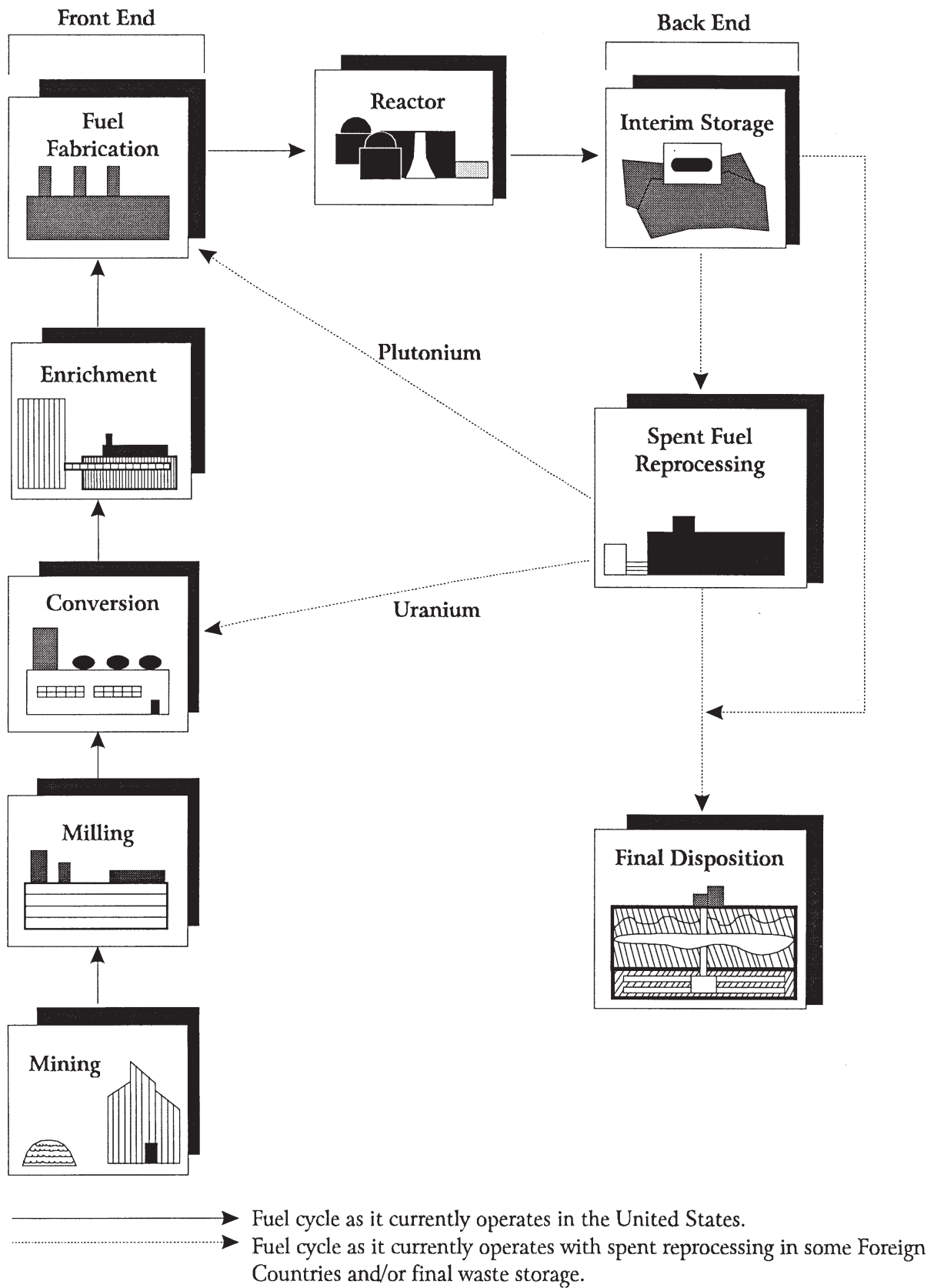


Fig. 9.1. Schematic of the nuclear fuel cycle. (From Ref. [2, p. 45]).

Niger, the Russian Federation, South Africa, the United States, and Uzbekistan [4, p. 36]. Most of the uranium now used in the United States is imported, with Canada being the largest supplier. Through 2000, the United States had been the world's leader in cumulative production of uranium, with Canada a close second. However, the U.S. share of production declined by the 1990s, and now Canada and Australia are the world's main suppliers of uranium. Together, they accounted for about 50% of world production in 2000 [4].

Uranium in rocks is mostly in the form of a uranium oxide, U_3O_8 . In "conventional" mining, the rock is extracted from open pit or underground mines, the U_3O_8 is then extracted in the milling process by crushing the rock and leaching with acid, and the U_3O_8 is then recovered from the liquid and dried. The concentrated U_3O_8 is known as *yellowcake*.⁴ In an "unconventional" method, appropriate for only certain types of uranium deposits, U_3O_8 is extracted by in situ leaching (i.e., by pumping a leaching agent through the ore without physical removal of the rock).⁵

At low concentrations, the uranium content is expressed in terms of the uranium grade, given in percent by weight of either uranium or U_3O_8 . Thus, ore which is 1000 ppm of uranium corresponds to grades of 0.100% U or 0.118% U_3O_8 .⁶ In an extensive 1983 study of U.S. uranium ores, deposits were listed with U_3O_8 grades ranging from under 0.01% to over 1.8%, with a median of about 0.1% [6, p. 39]. The higher the grade, the less the amount of ore that must be extracted, which, in general, leads to lower costs. Ores below a grade of 0.05% are considered low-grade ores and have not been widely needed. Of course, the ultimate criterion is overall cost, not grade per se, and at one time open-pit mining utilized ores down to 0.04% [7, p. 411].

During 2001, most of the uranium extraction in the United States was done by in situ leaching, using ores ranging in grade from 0.09% to about 0.20% U_3O_8 [8]. Worldwide, conventional mining dominates but is now economically practical only at higher uranium grades ("above a few tenths of a percent") [1, p. 25].

Radon Exposures from Uranium Mining and Mill Tailings

In the early days of uranium mining, little attention was paid to radiation safety. In the Middle Ages, long before uranium had been identified as an element, metal miners in southern Germany and Czechoslovakia contracted lung ailments, called *Bergkrankheit* ("mountain sickness"). Modern scientists have attributed the ailment to lung cancer caused by a high uranium concentration

⁴ U_3O_8 is not yellow in its pure form. Yellowcake is about 85% U_3O_8 [5, p. 241], and the yellow color results from another uranium compound in the ore.

⁵ The designations "conventional" and "unconventional" correspond to those, for example, of Ref. [1, p. 25].

⁶ In international usage, "grade" usually refers to U content, whereas in U.S. DOE documents it refers to U_3O_8 content. Note: U_3O_8 is 84.8% uranium and 15.2% oxygen, by weight.

that, by chance, was in the rock formations being mined. The decay of the radionuclides in the uranium series proceeds from ^{238}U through several steps to ^{226}Ra and then to radon gas (^{222}Rn) and its radioactive progeny. Inhalation of these “radon daughters” can lead to lung cancer (see Section 3.5.1).

As one would expect, the problem of radon exposure is more extreme in uranium mines than in other sorts of mine. It became a particularly serious problem in a number of countries—for example, in the United States, Czechoslovakia, and Canada—when large-scale uranium mining was begun in the 1940s to meet the demands of nuclear weapon and nuclear power programs. By the late 1950s, steps were initiated in the United States to reduce radon exposures, mainly through better ventilation, and by the 1970s, the average exposures of uranium miners had become quite low (lower than that from indoor radon in many homes). However, a good deal of damage had already been done, and there is unambiguous evidence of increased lung cancer fatalities among uranium miners.

The residues of the milling operation, representing the remainder of the ore after extraction of the U_3O_8 , are the *mill tailings*. All of the uranium progeny, starting with ^{230}Th , are present in the tailings.⁷ The radionuclide ^{230}Th has a half-life of 75,400 years and thus sustains the remainder of the uranium series for a long period of time. This results in the continuous production of radon, some escaping to the atmosphere. Of course, these steps do not increase the rate of radon production above what it would have been without mining, but the radon in the tailings can more readily reach the atmosphere than can radon in underground ore. At one time, this was viewed by some as constituting an important environmental hazard, and it is still deemed necessary to take remedial measures to limit radon emissions from the tailings (using overlying layers of material to impede radon escape). However, interest in the issue has diminished as it has become obvious that exposures from “normal” indoor radon pose a much more serious problem, in terms both of the number of people impacted and the magnitudes of the radon concentrations to which they are exposed.⁸

9.2.2 Enrichment of Uranium

Preparation for Enrichment: Conversion

There are a variety of approaches to the enrichment of uranium, each taking advantage of the small mass difference between ^{235}U and ^{238}U . In the most used of these processes, it is necessary to have the uranium in gaseous form. For that purpose, the U_3O_8 is chemically converted to gaseous uranium hexafluoride, UF_6 . This is the compound of choice, because UF_6 is a gas at lower

⁷ The ^{234}U remains in the yellowcake and the radionuclides between ^{238}U and ^{234}U in the uranium series are short-lived.

⁸ For a comparison of the hazards from mill tailings and indoor radon, see, for example, Ref. [9].

temperatures than can be reached by any other uranium compound in gaseous form [10, p. 589].

Degrees of Enrichment

Natural uranium has an isotopic abundance by *number of atoms* of 0.0055% ^{234}U , 0.720% ^{235}U and 99.275% ^{238}U .⁹ In the remainder of the discussion of uranium isotopic enrichment, we will follow the standard practice of describing the ^{235}U fraction in terms of *mass* rather than, as is common in many other scientific applications, of number of atoms.¹⁰ For natural uranium, the ^{235}U abundance by mass is 0.711%. The presence of the small amount of ^{234}U is often ignored, because corrections on the order of 10^{-4} or less are irrelevant.

The fissile nuclide in thermal reactors is ^{235}U . For reactors that require uranium with a higher fraction of ^{235}U than is found in natural uranium, *enrichment* is necessary. This is, of course, the case for light water reactors (LWRs). Fuel used in LWRs in past years has been enriched to ^{235}U concentrations ranging from under 2% to over 4%. The anticipated average for the United States, for cumulative production up until about 2010, is 3.0% for BWR fuel and 3.75% for PWR fuel.¹¹

The material used in LWRs is known as slightly enriched uranium, in contrast to the highly enriched uranium used for nuclear weapons and submarine reactors. Within the core of a given reactor, enrichments vary with the location of the fuel assemblies. As discussed later in the context of the burnup of fuel, there is a general trend toward using fuel with higher initial enrichments.

The products of the enrichment process are the enriched material itself and the depleted uranium, sometimes called *enrichment tails*. Typically, enrichment tails have in the neighborhood of 0.2% to 0.35% ^{235}U remaining [12, p. 7]. As one goes to lower concentrations of ^{235}U in the tails, the consumption of uranium ore is reduced, but the cost of enrichment is increased. Thus, there is a trade-off.

The depleted uranium is sometimes used in special applications. Its use in armor-piercing shells, where the high density of uranium is advantageous ($\rho \approx 19 \text{ g/cm}^3$), has led to some public concern about the resulting environmental risks. However, depleted uranium has a lower specific activity than

⁹ The ^{234}U arises as a member of the ^{238}U series, with an abundance relative to ^{238}U that is inversely proportional to the half-lives of the two isotopes ($2.45 \times 10^5 \text{ yr}$ and $4.468 \times 10^9 \text{ yr}$, respectively).

¹⁰ These descriptions of isotopic abundance are related by the expression $w = [(1 - \delta)/(1 - x\delta)]x$, where, specialized to the case of uranium, w is the ratio of ^{235}U mass to total uranium mass, x is the ratio of the number of ^{235}U atoms to the total number of uranium atoms, and δ is the ratio of the difference between the ^{238}U and ^{235}U atomic masses to the ^{238}U atomic mass. For low enrichments (with $\delta = 0.0126$ for uranium), $w \doteq 0.987x$, and there is little difference between the two formulations. For natural uranium, $x = 0.00720$ and $w = 0.00711$.

¹¹ This is the planning basis for the Yucca Mountain nuclear waste repository [11, p. 3–13].

does natural uranium, and there is no evidence of appreciable radiation hazards except for occupants of a closed vehicle that has been struck by a shell that partially vaporizes within it.¹²

Methods for Enrichment

The leading enrichment methods in terms of past or anticipated future use are as follows:¹³

- ◆ *Gaseous diffusion.* The average kinetic energy of the molecules in a gas is independent of the molecular weight M of the gas and depends only on the temperature. At the same temperature, the average velocities are therefore inversely proportional to \sqrt{M} . For uranium in the form of UF_6 , the ratio of the velocities of the two isotopic species is 1.0043.¹⁴ If a gas sample streams past a barrier with small apertures, a few more ^{235}U molecules than ^{238}U molecules pass through the barrier, slightly increasing the ^{235}U fraction in the gas. The ratio of $^{235}\text{U}/^{238}\text{U}$ before and after passing the barrier is the enrichment ratio α . Its ideal or maximum value is given by the velocity ratio $\alpha = 1.0043$. However, one cannot calculate the number of stages of diffusion needed to achieve a given enrichment merely in terms of powers of α , because the ideal value is not achieved in practice and because it is necessary to continually recycle the less enriched part of the stream. Typically, if one starts with natural uranium (0.71%) and with tails depleted to 0.3%, it is found that about 1200 enrichment stages are required to achieve an enrichment of 4% [15, p. 36].
- ◆ *Centrifuge separation.* Any fluid—liquid or gaseous—can be separated in a high-speed centrifuge. The centrifugal action causes the heavier component to become more highly concentrated at large radii. As in gaseous diffusion, only a small gain is made in any one stage, and high enrichments of the UF_6 are reached using multiple centrifuge stages, with the slightly enriched output of one stage serving as the input to the next one. The centrifuges used for uranium enrichment are rotating cylinders. Uranium that is slightly enriched in ^{238}U (and depleted in ^{235}U) can be extracted from the outer region of the cylinder and returned to an earlier stage in the centrifuge cascade. Uranium slightly enriched in ^{235}U can be extracted from regions near the center and used as input to the next higher stage in the array of centrifuge units. High enrichments of the UF_6 are reached using multiple centrifuge stages. The power requirement for a given degree of enrichment is much less for centrifuge separation than for diffusion separation.
- ◆ *Aerodynamic processes.* These processes exploit the effects of centrifugal forces, but without a rotating centrifuge. Gas—typically UF_6 mixed with

¹² In this case, direct damage from the shell is a still greater concern.

¹³ Detailed discussions of these methods are given in, for example, Refs. [13] and [14].

¹⁴ The atomic mass of fluorine (F) is 19.00 u.

hydrogen—expands through an aperture, and the flow of the resulting gas stream is diverted by a barrier, causing it to move in a curved path. The more massive molecules on average have a higher radius of curvature than do the lighter molecules, and a component enriched in ^{235}U is preferentially selected by a physical partition. The process is repeated to obtain successively greater enrichments. The gas nozzle process was developed in Germany as the *Becker* or *jet nozzle* process. A variant with a different geometry for the motion of the gas stream, the so-called *Helikon* process, has been developed and used in South Africa.

- ◆ *Electromagnetic separation.* When ions in the same charge state are accelerated through the same potential difference, the energy is the same and the radius of curvature in a magnetic field is proportional to \sqrt{M} . Thus, it is possible to separate the different species magnetically. This separation can be done with ions of uranium and, so, conversion to UF_6 is not, in principle, necessary. Overall, this approach gives a low yield at a high cost in energy, but it has the advantage of employing a relatively straightforward technology.
- ◆ *Laser enrichment.* The atomic energy levels of different isotopes differ slightly.¹⁵ This effect can be exploited to separate ^{235}U from ^{238}U , starting with uranium in either atomic or molecular form. For example, in the atomic vapor laser isotope separation (AVLIS) method, the uranium is in the form of a hot vapor. Lasers precisely tuned to the appropriate wavelength are used to excite ^{235}U atoms, but not ^{238}U atoms, to energy levels that lie several electron volts above the ground state. An additional laser is used to ionize the excited ^{235}U atoms.¹⁶ The ionized ^{235}U atoms can be separated from the un-ionized ^{238}U atoms by electric and magnetic fields. An alternative to the AVLIS method is the SILEX process (separation of isotopes by laser excitation). It is based on the selective dissociation of UF_6 (a gas) into UF_5 (a solid) [16]. The costs in energy of laser enrichment are lower than those of other enrichment methods, but a sophisticated laser technology is required, and, to date, there are no commercial facilities for laser enrichment of uranium. Once mastered, the laser technique is expected to be relatively inexpensive. On the negative side, there have been fears that if the technique develops sufficiently, laser separation may make it easy for small countries or well-organized terrorist groups to enrich uranium for nuclear weapons.

¹⁵ This “isotope effect” was responsible for the discovery of ^2H . It arises for two reasons: (1) the atomic energy levels depend on the reduced mass of the electrons, which differs from the mass of a free electron by an amount proportional to m_e/M , where m_e is the electron mass and M the atomic mass, and (2) the energy levels of heavy atoms depend in a small measure on the overlap between the wave functions of the innermost electrons and the nucleus, with differences between isotopes due to differences in their nuclear radii.

¹⁶ The ionization energy to remove an electron from uranium in its unexcited (ground) state is 6.2 eV.

Adopted Enrichment Practices

During World War II, not knowing which method would be the most effective, the United States embarked on both diffusion and electromagnetic separation, as well as still another method that was later discarded (namely thermal diffusion, which exploits temperature gradients). The electromagnetic separation technique was abandoned in the United States after World War II and was widely considered to be obsolete. However, it was found in 1991, after the Gulf War, that Iraq had been secretly using this approach in an attempt to obtain enriched uranium for nuclear weapons.

In the United States since World War II, the enrichment program has relied on gaseous diffusion, as did early European programs. Since the 1950s, the DOE (and its predecessor agencies, starting with the AEC) operated two large gaseous diffusion enrichment facilities—one in Paducah, Kentucky and one in Portsmouth, Ohio. In 1999, operation of these facilities was privatized under the management of the United States Enrichment Corporation (USEC), and in 2001, the Portsmouth plant—the smaller of the two—was shut down. Outside the United States, there are major enrichment facilities in France and Russia and smaller ones in a number of other countries, including the United Kingdom, Netherlands, Germany, Japan, and China [1, p. 33]. In recent years (1999–2001), most of the enrichment for fuel used in U.S. reactors has been carried out abroad, particularly in Russia [17, p. 28]. The gaseous diffusion method is used in France and the United States, whereas the centrifuge method is used almost everywhere else.

A 2001 OECD report anticipated that most new plants will use the centrifuge method [1, p. 85]. The USEC, which has been the sole U.S. company pursuing enrichment, plans to replace its diffusion plant with a “second-generation” centrifuge plant [18]. To that end, it submitted to the NRC in February 2003 a license application for a preliminary demonstration facility scheduled to be on-line in 2005. A larger, full-scale plant—the so-called American Centrifuge—is planned for later in the decade [19]. In addition, the Louisiana Energy Services Partnership—an organization that includes, among others, Urenco (a major European enrichment company), the Westinghouse Electric Company, and several U.S. utilities is seeking to build a centrifuge enrichment facility in New Mexico [20].

USEC has also worked on developing laser enrichment technology as a “third-generation” option. It originally focused on the AVLIS method and later on the SILEX process, but as of Spring 2003, USEC decided to concentrate on its centrifuge projects to the exclusion of laser options [21].

Separative Work

In a ^{235}U enrichment process, there are three streams of material: the input or *feed*, the output or *product*, and the residue or *tails*. The system operates

with a cascade of steps, with the enrichment of the product increasing successively in each step.¹⁷ As the enrichment cascade progresses, the tails from an intermediate stage have a higher ^{235}U concentration than the original feed material, and these tails can profitably be returned to the cascade. There are different strategies for reusing the tails of successive steps to maximize the efficiency of the process, including an “ideal cascade” (see, e.g., Ref. [5]).

The difficulty of carrying out uranium enrichment, as measured, for example, by the relative energy required in the diffusion process, is described by a quantity known as the *separative work*.¹⁸ Separative work has the dimensions of mass and is specified in *separative work units* (SWU), as kg-SWU or tonne-SWU. Figure 9.2 shows the separative work required to produce 1 kg of enriched uranium product and 1 kg of ^{235}U in the form of enriched uranium, for different degrees of final enrichment.

As seen in Figure 9.2, it requires more separative work per kilogram of ^{235}U to enrich uranium from 0.7% to 5% than to carry it the rest of the way to 95% enrichment. Even 3% enriched uranium fuel is more than “halfway” to 95% enrichment. This could make the slightly enriched uranium produced for reactors a somewhat attractive initial material for the production of uranium for weapons (see Chapter 17).

Although the formalism was developed in the context of gaseous diffusion enrichment methods, it is used for other processes as well. Separative work serves as a general measure of what is achieved in the enrichment. For an individual process, it serves also as a measure of relative energy consumption. Different processes vary greatly in their energy consumption. For example, gaseous diffusion uses 2.5 MWh/kg-SWU, while the gas centrifuge uses about 1/50th as much energy and laser isotope separation methods still less [12, p. 530].

It is of interest, in the spirit of what is known as “net energy analysis,” to compare the energy required to enrich uranium with the energy obtained from it. The production by the diffusion process of 1 kg of ^{235}U in the form of

¹⁷ The logical structure of the system is similar to that of fractional distillation, and some of the formalism was developed in the 19th century by Lord Rayleigh [5, p. 649].

¹⁸ In a separation process, the masses are in the ratios:

$$\frac{M_F}{M_P} = \frac{w_P - w_T}{w_F - w_T} \quad \text{and} \quad \frac{M_T}{M_P} = \frac{w_P - w_F}{w_F - w_T},$$

where M_F , M_P , and M_T are the masses and w_F , w_P , and w_T are the ^{235}U concentrations (by weight) of the feed, product, and tails, respectively. This result follows from the conservation of total mass and ^{235}U mass: $M_F = M_P + M_T$ and $w_F M_F = w_P M_P + w_T M_T$. The separative work in an isotopic enrichment process is

$$\Delta V = M_P V_P + M_T V_T - M_F V_F,$$

where V is the *value function*, defined as $V = (1 - 2w) \ln[(1 - w)/w]$. (For the derivation leading to this result see, e.g., Ref. [5, Chapter 12].)

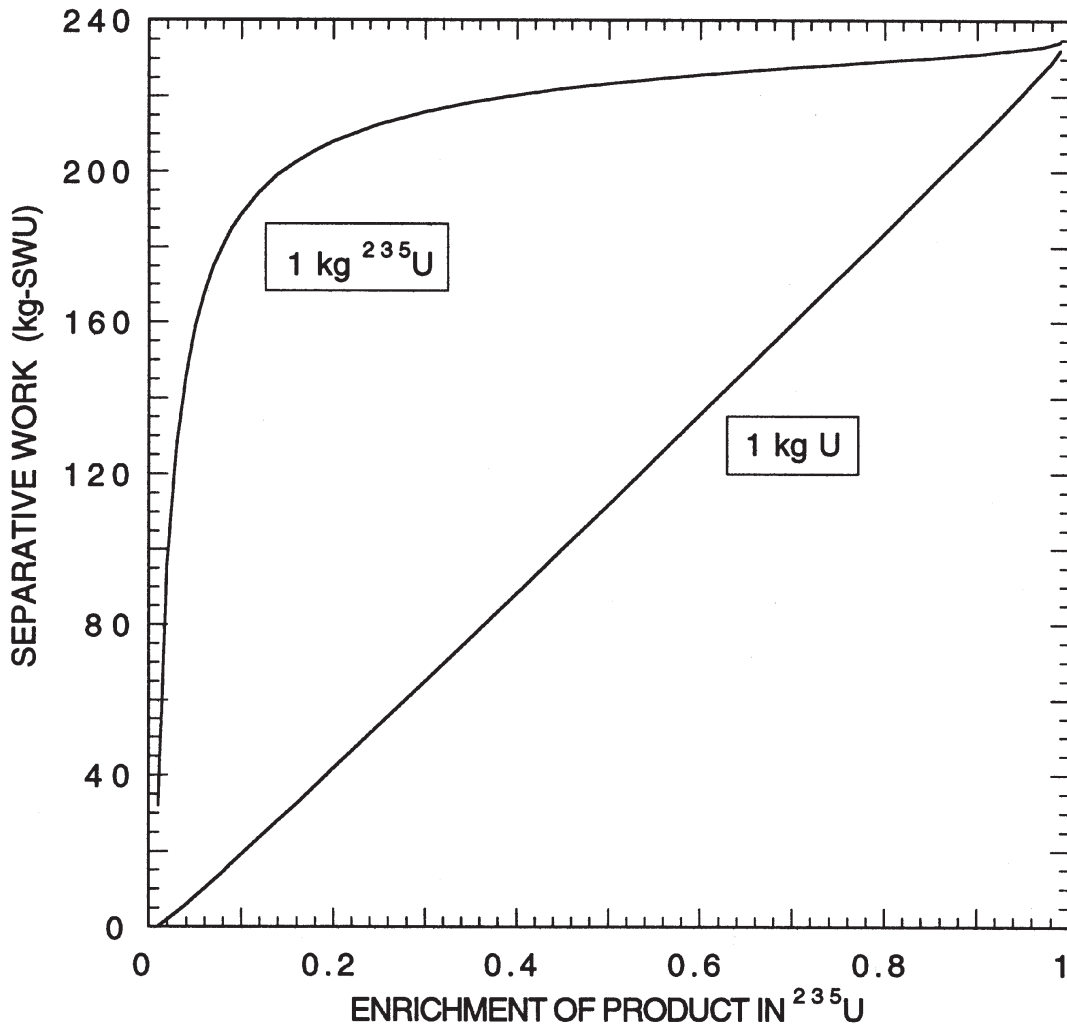


Fig. 9.2. Separative work (in kg-SWU) as a function of ^{235}U enrichment for the production of 1 kg of U and 1 kg of ^{235}U . (The initial feed concentration is $w_F = 0.00711$ and the assumed tails concentration is $w_T = 0.0025$.)

uranium enriched to 3.75% in ^{235}U , with a ^{235}U concentration in the tails of 0.25%, requires 142 kg-SWU (see Figure 9.2) or about 355 MWh. The reactor output from 1 kg of ^{235}U is about 1 MWyr or 8760 MWh (see Section 9.3.2). Therefore, the relatively energy-intensive diffusion process requires roughly 4% of the energy output of a reactor. Although this is the largest single energy input to the production of nuclear power, it does not significantly reduce the net positive energy balance from nuclear power.

9.2.3 Fuel Fabrication

Most nuclear fuel used in light water reactors is in the form of uranium dioxide (UO_2), also sometimes called “uranium oxide.” This is not a single compound, but a mixture of oxides (UO_n), where n typically ranges from 1.9 to 2.1 [22, p. 226]. The UO_2 is produced by chemical conversion of the enriched UF_6 . It is

then processed into a fine powder and compacted and sintered to form rugged pellets. During sintering, the oxygen content of the fuel can be adjusted. The pellets are corrected in size, to close tolerances, by grinding.¹⁹ The pellets are loaded into zircaloy fuel pins that are arranged in a matrix to form the fuel assembly. As discussed in Section 8.2.3, the reactor core consists of a large number of such assemblies.

9.2.4 Other Fuel Types

The focus here has been on UO_2 , which is the usual fuel for LWRs. Other fuel types are of interest, however, even if not widely used at present (see, e.g., Ref. [1]). Possibilities include the following:

- ◆ *Mixed-oxide fuel (MOX)*. MOX fuel, a mixture of uranium and plutonium oxides, uses plutonium in order to exploit its energy content, reduce the stocks of potential weapons materials, or both (see Section 9.4.2).
- ◆ *Metal alloy fuels*. Metallic fuel, in the form of alloys of uranium, provide an alternative to oxide fuels that is easier to reprocess.
- ◆ *Microsphere fuel particles*. High-temperature gas-cooled reactors utilize uranium or thorium oxide in the form of very small spheres, with multiple layers of outer protection. These are the so-called TRISO fuel particles (see Section 16.4.3).
- ◆ *Thorium fuels*. A fuel cycle based on thorium-232 as the fertile fuel and uranium-233 as the fissile fuel could be used to supplement uranium resources. It has the advantage of producing little plutonium and thereby lessening waste disposal and proliferation problems.
- ◆ *Molten salt*. Although all operating commercial reactors presently use fuel in solid form, it is also possible to have the fuel as a liquid uranium fluoride, mixed with other liquid fluorides, as would be done in the proposed molten salt reactor (see Section 16.6.1).

9.3 Fuel Utilization

9.3.1 Burnup as a Measure of Fuel Utilization

Thermal Efficiency of U.S. Reactors

The *thermal efficiency* of a reactor is the ratio of the electrical energy produced to the total heat energy produced. Since 1973, the average thermal efficiency of U.S. reactors has ranged between 30.6% and 32.1%, according to DOE compilations [23, Table A6]. There has been a gradual improvement with time, and since 1985 it has been above 31.5%, reaching 32.1% for the years 1996–2002. We will use the approximate figure of 32% as the nominal average efficiency of LWRs.

¹⁹ For a discussion of the details of these processes, see Ref. [22, Section 7.5].

Basic Unit for Burnup: GWDT per MTHM

A useful measure of the performance in the nuclear fuel cycle is the energy obtained per unit mass of fuel, known as the fuel's *burnup*. The burnup is commonly specified in megawatt-days or gigawatt-days of thermal output per metric tonne of heavy metal (MWDt/MTHM or GWDT/MTHM). This is a cumbersome notation for repeated use, and we represent GWDT/MTHM in a more compact form as GWd/t (gigawatt-days per tonne). In standard energy units, $1 \text{ GWd} = 8.64 \times 10^{13} \text{ joules (J)}$.

For U.S. reactors, as well as most reactors elsewhere, the “heavy metal” in the original fuel is uranium.²⁰ The fuel is in the form of uranium oxide (UO_2). About 12% of the mass of the fuel is oxygen and, therefore, there is a distinction between the mass of heavy metal and the mass of the fuel. The heavy metal in the spent fuel removed from the reactor is still primarily uranium, but it also includes isotopes of plutonium and—to a small extent—other transuranic elements. Typically, the mass of heavy metal is about 3% or 4% less in the spent fuel than in the initial fuel due to fission (including fission of plutonium isotopes).²¹

A 1000-MWe reactor, operating at a typical thermal efficiency of 32%, produces energy at the rate of 3125 MWt (where it is explicitly indicated that this is the thermal output). One gigawatt-year of electric power therefore represents a thermal output of 1141 GWd(t). If, for example, the average burnup in a reactor is 40 GWd/t, the fuel consumption is 28.5 tonnes of enriched uranium per gigawatt-year.

Trends in Burnup of LWR Fuel

Average burnup values for past years are shown in Table 9.1 along with the average projected for the fuel to be deposited at the Yucca Mountain waste repository. Overall, there has been a trend with time toward higher burnup, on average roughly doubling in the 25 years from 1973 to 1998 and projected to continue to rise. Thus, in a 1993 DOE projection, it was expected that the median PWR fuel burnup for standard assemblies would be about 43 GWd/t in the year 2000—a value actually achieved in 1998—and 51 GWd/t

²⁰ The main exception is for reactors that use a mixture of uranium and plutonium in *mixed-oxide* fuels (see Section 9.4.2).

²¹ The designation “metric tons of heavy metal” (MTHM) commonly appears in discussions of the utilization and disposal of nuclear fuel. Here, MTHM refers to the heavy metal mass of the initial fuel. Alternatively, this can be made explicit by using the designation “metric tonnes of *initial* heavy metal” (MTIHM). In effect, “MTHM” and “MTIHM” are used interchangeably and mean the same thing. Thus, the U.S. spent fuel inventory at the end of 1995 is given as 31926 MTHM in the 2002 Yucca Mountain EIS [24, Table A-7] and as 31952 MTIHM in a 1996 report [25, Table 1.2]. (The 0.08% difference is insignificant compared to the difference of several percent in the actual heavy metal contents of the initial and spent fuel.)

Table 9.1. Average burnup of U.S. spent fuel, for BWRs and PWRs.

Year Discharged	Burnup (GWd/t)	
	BWRs	PWRs
Annual		
1973	12.4	23.7
1978	19.8	26.4
1983	26.7	30.1
1988	24.1	33.4
1993	30.3	38.9
1998	36.4	43.3
Average		
1968–1998	25.1	33.4
Yucca Mountain ^a	33.6	41.2

^aThis includes all commercial spent fuel slated for Yucca Mountain (i.e., all of the fuel discharged through about 2010).

Source: Refs. [26, Table 2], and [11, Table 3–6].

in 2010 [27, p. 144]. It should be noted that Table 9.1 gives only averages. In any year, there is a wide disparity among reactors. Thus, in 1993, when the average PWR burnup was 38.9 GWd/t, 12% of the spent fuel had a burnup in excess of 45 GWd/t [28, p. 25].

An estimated average for the fuel expected to be deposited in the Yucca Mountain repository (i.e., all of the fuel generated in the United States from the start of nuclear power until about 2010) is given in Yucca Mountain planning documents. The indicated average burnups for BWRs and PWRs are 33.6 GWd/t and 41.2 GWd/t, respectively, with average initial ²³⁵U enrichments of 3.03% and 3.75% [11, p. 3–13]. The corresponding weighted average for all LWR fuel is 38.6 GWd/t, with an enrichment of 3.5%.

The burnup depends on the power density in the fuel and the length of time the fuel is kept in the reactor. Unless the conversion ratio is very large, a high burnup requires a high initial enrichment in ²³⁵U. For example, model calculations carried out at Oak Ridge National Laboratory included a fuel cycle that gives a burnup of 60 GWd/t, with a 4.7% ²³⁵U concentration and the fuel kept in the reactor for three 600-day cycles (i.e., 1800 days) [29, p. 2.4–3]. Looking to the future, the term “high burnup” is sometimes used to refer to a burnup in excess of 70 GWd/t [1, p. 88].

High enrichment alone is not sufficient for high burnup. In addition, the fuel and cladding must be able to withstand the added neutron bombardment and the buildup of fission gases. This depends both on the fuel itself and on the composition of the alloy used for the cladding. Further, to compensate for the initial high reactivity with high enrichment, burnable poisons are used with the fuel.

With higher burnup, the mass and volume of spent fuel required for a given energy output are reduced. The consumption of uranium is also reduced, because a larger fraction of the ^{235}U is consumed and more ^{239}Pu is produced and consumed. In addition, the time intervals between refueling operations can be longer, meaning that less reactor time is lost for refueling. High burnup fuel is also less proliferation-prone than fuel with lesser burnup because the concentrations of ^{240}Pu and ^{242}Pu are higher and the fuel is harder to handle due to its high level of radioactivity (see Chapter 17).

9.3.2 Uranium Consumption and Plutonium Production

In uranium-fueled reactors, there is a continual destruction of ^{235}U , through fission and neutron capture, and buildup of plutonium isotopes through neutron capture and beta decay. The plutonium sequence starts with ^{239}Pu , following neutron capture in ^{238}U , and continues to include plutonium isotopes up to ^{242}Pu , as well other heavy radionuclides—for example, ^{241}Am (atomic number $Z = 95$), which is produced primarily from the beta decay of ^{241}Pu ($T = 14.39$ yrs). Similarly, radionuclides of atomic mass numbers 236 and 237 (primarily, ^{236}U and ^{237}Np), are formed through neutron capture in ^{235}U and, for ^{237}Np , subsequent neutron capture and beta decay.

Most of the reactor's energy output comes from the fission of ^{235}U . However, as discussed earlier, production and fission of ^{239}Pu also play a significant part in the energy economy of the reactor. Thus, the total energy output is the sum of the energy from the fissile plutonium isotopes (^{239}Pu and ^{241}Pu) plus the energy from ^{235}U and, to a much lesser extent, ^{238}U .

Table 9.2 gives the masses of the main actinide isotopes in the spent fuel when it is discharged from the reactor, for a representative PWR case, namely UO_2 fuel with a 3.75% ^{235}U enrichment, a burnup of 40 GWd/t, and a residence time in the reactor of a little over 3 years. The results in Table 9.2 were calculated with the ORIGEN program, developed at the Oak Ridge National Laboratory, which traces the production and consumption of the nuclides as a function of time, taking into account the simultaneous nuclear processes for all the nuclides, including fission, neutron capture, and radioactive decay.²² Important qualitative features of the results include the following:

- ◆ Fission products. The fission product production is about 40 kg, as seen from the 40.4-kg reduction in the heavy metal mass.²³ It may seem sur-

²² I am indebted for the calculation of the activities to Dr. Edwin Kolbe, Project Manager for Radioactive Materials at the Swiss National Cooperative for the Disposal of Radioactive Waste (NAGRA) [31]. The mass of each radionuclide was calculated from the ratio of its activity to its specific activity [see Eq. (3.4)]. These ORIGEN results are also used in Chapter 10.

²³ The precise number is slightly less than 40.4 kg due, in part, to neutron escape from the reactor. The mass equivalent of the energy produced in the reactor is 0.04 kg, representing a very small additional “correction” to the calculated fission product mass.

Table 9.2. Activity and mass of actinides in PWR spent fuel (per MTHM), for burnup of 40 GWd/t and ^{235}U enrichment of 3.75%.

Nuclide	Half-life (years)	Activity (Bq)	Mass (kg)	Isotopic Percent
Input fuel				
^{234}U	2.46×10^5		0.05	0.005
^{235}U	7.04×10^8		37.5	3.75
^{238}U	4.47×10^9		962.4	96.24
Total HM ^a			1000	
Spent fuel				
^{234}U	2.46×10^5	4.70×10^{10}	0.2	0.02
^{235}U	7.04×10^8	6.85×10^8	8.6	0.90
^{236}U	2.34×10^7	1.21×10^{10}	5.1	0.53
^{238}U	4.47×10^9	1.16×10^{10}	934.4	98.54
Total U			948.2	100
^{237}Np	2.14×10^6	1.49×10^{10}	0.57	87
^{239}Np	0.0065	7.18×10^{17}	0.08	13
Total Np			0.65	100
^{238}Pu	87.7	1.38×10^{14}	0.22	2.1
^{239}Pu	2.41×10^4	1.28×10^{13}	5.56	53.2
^{240}Pu	6564	2.07×10^{13}	2.46	23.6
^{241}Pu	14.29	5.93×10^{15}	1.54	14.8
^{242}Pu	3.73×10^5	9.66×10^{10}	0.66	6.3
Total Pu			10.4	100
^{241}Am	432.2	6.41×10^{12}	0.05	24
^{243}Am	7370	1.18×10^{12}	0.16	76
Total Am			0.21	100
^{244}Cm	18.1	1.54×10^{14}	0.05	
Total HM			959.6	
Decrease in HM			40.4	

^aThe initial total heavy metal (HM) mass is the uranium mass.

Source: Half-lives from Ref. [30]; activities are based on ORIGEN calculation (see text) [31]. The mass is calculated from the activity and half-life.

prising that the fission product mass exceeds the mass of the destroyed ^{235}U . However, in addition to fission in ^{235}U , there is fission of ^{239}Pu and ^{241}Pu .

- ◆ ^{236}U . Neutron absorption in ^{235}U leads to capture in 14% of the events (see Table 7.1). Therefore, the destruction of ^{235}U is accompanied by the production of a significant amount of ^{236}U . The isotope ^{236}U has a modest capture cross section at thermal energies ($\sigma_\gamma = 5.1$ b) and a negligible fission cross section. Its capture product, ^{237}U , decays with a rather short half-life (6.75 days) to ^{237}Np ($T = 2.14 \times 10^6$ years), which has a large

capture cross section and a considerably smaller fission cross section. This produces ^{238}Np , followed by beta decay to ^{238}Pu [32]. Overall, ^{236}U is more a weak poison rather than a fissile or fertile fuel.

- ◆ ^{238}Pu . With a half-life of 88 years, ^{238}Pu is useful as an energy source for use in “radioisotope thermoelectric generators” (RTGs), in which heat from the radionuclide is used to produce electricity. Its intermediate half-life means that the output from ^{238}Pu changes little in a decade or two, yet its specific activity is relatively high. RTGs based on ^{238}Pu were at one time used in heart pacemakers, but, more recently, their main use has been in the space program (e.g., in the Cassini mission to Saturn launched in 1997).
- ◆ ^{239}Pu . There is continuous production of ^{239}Pu following neutron capture in ^{238}U , and also continuous destruction of ^{239}Pu , primarily by fission but also by neutron capture. The final ^{239}Pu abundance reflects the net effect of production and destruction. The relatively low value of its isotopic abundance (53%) is characteristic of “reactor-grade” plutonium (see Section 17.4.1).
- ◆ ^{240}Pu . The capture cross section is relatively high in ^{239}Pu , so that neutron absorption results in the production of ^{240}Pu as well as in fission. ^{240}Pu acts as a fertile fuel for the production of ^{241}Pu , but the absorption cross section for ^{240}Pu is less than that for ^{239}Pu , and ^{240}Pu is not consumed as rapidly as is ^{239}Pu .
- ◆ Other Pu isotopes. Neutron capture on ^{240}Pu produces ^{241}Pu , which is fissile but which also has an appreciable branch for neutron capture to ^{242}Pu . The buildup of plutonium isotopes stops here because ^{243}Pu , the next in the series, decays with a half-life of 5.0 h to ^{243}Am and further neutron captures and beta decays moves the chain to atomic numbers higher than that of plutonium.

The plutonium isotopes through atomic mass number $A = 242$ have half-lives that are long compared to normal exposure periods in the reactor (the shortest is 14.3 years for ^{241}Pu). Considering those with $A > 238$ as a group, they are fed by neutron capture in ^{238}U (quickly followed by beta decay to ^{239}Np and ^{239}Pu) and are depleted primarily by the fission of ^{239}Pu and ^{241}Pu and the beta decay of ^{243}Pu .

9.3.3 Energy from Consumption of Fuel

Energy per Unit Mass from Fission of ^{235}U

The fission of a ^{235}U nucleus corresponds on average to the release of 200 MeV (3.20×10^{-11} J), including the associated contributions from neutron capture and the decay of fission fragments (see Section 6.4.2). The number of ^{235}U atoms per gram of uranium is wN_A/M , where w is the fraction of ^{235}U in the uranium (by weight), N_A is Avogadro’s number, and M is the atomic mass of ^{235}U . Thus, 1 kg of natural uranium ($w = 0.00711$) has 1.822×10^{22} nuclei

Table 9.3. Energy per unit mass from fission of uranium, at different degrees of enrichment.

Category	Enrichment, w (%)	Energy per Unit Mass ^a		
		J/kg	GWd/t	Tonne/GWyr(e) ^b
Natural U	0.711	5.84×10^{11}	6.8	169
Enriched U	3.2	2.63×10^{12}	30.4	38
Enriched U	3.5	2.87×10^{12}	33.3	34
Enriched U	3.75	3.08×10^{12}	35.6	32
Enriched U	5.0	4.10×10^{12}	47.5	24
Pure ^{235}U	100	8.21×10^{13}	950	1.2

^aThermal energy, assuming fission of *all* ^{235}U , a release of 200 MeV per ^{235}U fission, and no fission of other nuclides.

^bAssuming a thermal conversion efficiency of 32%.

of ^{235}U and their complete fission would release 5.8×10^{11} J. The available fission energy per kilogram of uranium, for different degrees of enrichment, is given in Table 9.3 for a few illustrative cases, assuming fission of all ^{235}U and ignoring losses due to capture in ^{235}U and gains from fission in ^{239}Pu and other nuclides.

As indicated earlier, 1 GWyr of electric power corresponds to a thermal output of 1141 GWd(t) or 9.86×10^{16} J. Assuming complete fission of all the ^{235}U and ignoring fission in plutonium, this corresponds to a fuel requirement of 1.20 tonnes of ^{235}U per GWyr(e). For the specific case of a 3.75% enrichment in ^{235}U (as in Table 9.2), the requirement translates to 32 tonnes of uranium. The corresponding thermal burnup is 35.6 GWd/t, as indicated in Table 9.3.

Energy per Unit Mass of Fuel

The discussion in the preceding subsection is incomplete, because it omits many crucial factors that significantly modify the amount of ^{235}U required by a reactor. These include the following:

1. Not all of the ^{235}U is consumed in the reactor. For example, for the case described in Table 9.2, the ^{235}U content per MTHM is 37.5 kg in the fresh fuel and 8.6 kg in the spent fuel (i.e., a consumption of only 77% of the ^{235}U).
2. About 14% of the thermal neutron-absorption reactions in ^{235}U result in capture rather than fission.
3. Fission in ^{239}Pu (and, to a lesser extent, in ^{241}Pu) provides a substantial additional energy source. This reduces the ^{235}U required for a given energy production.
4. There is a small contribution from fast-neutron fission in ^{238}U .

These effects all change the number of fission events, reducing the number of fissions in ^{235}U and adding fission in plutonium isotopes and even ^{238}U . The overall consequence can be crudely estimated by comparing the decrease in the total heavy metal mass—which results almost entirely from fission of uranium and plutonium isotopes—to the original ^{235}U mass. The ratio of these quantities is 1.081, which would suggest that there are about 8.1% more fission events than would be given by complete fission of ^{235}U . Applying this 8.1% “correction” raises the ^{235}U burnup of 35.6 GWd/t to a revised value of 38.5 GWd/t. This estimate still ignores the differences in fission energy yields and atomic masses between ^{235}U and the heavier actinides. Roughly 40% of the fissions are in ^{239}Pu and ^{241}Pu , not ^{235}U . Taking this into account adds roughly 0.6 GWd/t. Gamma-ray emission following neutron capture in the actinides adds further. Together, these approximate corrections bring the total close to the burnup of 40 GWd/t indicated in Table 9.2.

A value of 40 GWd/t is a good representation of recent LWR performance, although average PWR burnups are now higher and future ones are expected to be still higher (see Table 9.1).²⁴ The burnup in GWd/t can be translated into the fuel requirement per year. For example, for a burnup of 40 GWd/t the total uranium requirement is 28.5 tonnes per GWyr, or 1.07 tonnes of ^{235}U for an enrichment of 3.75%.

The precise uranium requirements for a given energy output depend on details of the fuel cycle and reactor operation. Nonetheless, the following equivalence, as found for the above example, is useful for approximate estimates of the general magnitudes for a once-through LWR fuel cycle:

$$1 \text{ tonne of } ^{235}\text{U} \rightarrow 1 \text{ GWyr(e)} \quad (\text{approximate}).$$

9.3.4 Uranium Ore Requirement

The amount of uranium ore required to operate a reactor depends on the burnup achieved, the initial enrichment, and the amount of ^{235}U lost in the enrichment process. The mass M_F of natural uranium used as feed input to the enrichment facility is related to the mass M_P of enriched uranium produced in the fuel by the expression $M_F/M_P = (w_P - w_T)/(w_F - w_T)$, where w_P , w_T , and w_F are the enrichments of the fuel, the tailings, and natural uranium, respectively. For $w_P = 3.75\%$, $w_T = 0.2\%$, $w_F = 0.711\%$, and $M_P = 28.5$ tonnes/GWyr, the natural uranium requirement is 198 tonnes/GWyr. Thus, in round numbers, a once-through LWR fuel cycle requires about 200 tonnes

²⁴ The average burnup is systematically less in BWRs than in PWRs because burnup in the former is less uniform along the length of the fuel rods. The water at the bottom of the BWR tank has a high density and is a better moderator than the steam–water mixture at the top. This means that the maximum burnup is achieved at the bottom of the rod, with a smaller burnup higher up on the rod. The maximum acceptable burnups are about the same for the PWR and BWR, but the PWR has a more uniform profile along the length of the fuel rod and therefore a greater average burnup.

of natural uranium per gigawatt-year. Present world demand for uranium is 60,000 tonnes/yr, corresponding to the requirements for the present annual generation by nuclear power plants of about 300 GWyr [1, p. 17].

9.4 Back End of Fuel Cycle

9.4.1 Handling of Spent Fuel

Initial Handling of Reactor Fuel

Periodically, a portion of the fuel in the reactor is removed and replaced by fresh fuel. In typical past practice, an average sample of fuel remained in the reactor for 3 years, and approximately one-third of the fuel was removed each year, with a shutdown time for refueling and maintenance of up to about 2 months. The trend is now to extend the interval between refueling operations and to reduce the time for refueling.²⁵ Currently, time intervals of 18 months and shutdowns of 1 month are typical.

When the spent fuel is first removed from the reactor, the level of radioactivity is very high, due to the accumulation of radioactive fission products and radioactive nuclei formed by neutron capture. Each radioactive decay involves the release of energy, which immediately appears as heat, so the fuel is thermally hot as well as radioactively “hot.” Independent of the reprocessing question, the first stage is the same, namely allowing the fuel to cool both thermally and radioactively. The cooling of the fuel normally takes place in water-filled cooling pools at the reactor site.

Originally, it was planned to keep the spent fuel at the reactor for roughly 150 days and then to transfer it to handling facilities at other locations. The nature of the next step, in principle, depends on whether the fuel is to be disposed of as waste or reprocessed. However, as yet, this “next step” has been much delayed in the United States because no off-site facilities have been developed. Instead, almost all of the fuel has remained at the reactor sites—in many cases for more than 20 years.

In the absence of alternatives, some U.S. utilities are transferring older fuel rods from cooling pools to air-cooled (dry storage) casks at the reactor site. This may provide a workable temporary solution to the long delay in implementing a national waste disposal program. However, it is only a stopgap because the reactor operator cannot be counted on to be willing and able to supervise the spent fuel for prolonged periods of time (see Section 11.1.3).

Disposal or Storage of Spent Fuel

For many years, it had been assumed that all U.S. civilian nuclear waste would be reprocessed, but U.S. reprocessing plans have been abandoned. Instead,

²⁵ An annual refueling shutdown of 2 months would mean a *maximum* capacity factor of 83%, which is well below the present U.S. average.

official plans now call for disposing of the spent fuel directly, while retaining for many decades the option of retrieving it. The fuel is to remain in solid form and the fuel assemblies eventually placed in protective containers and ultimately moved in secure casks to either a permanent or an interim repository site. In the latter case, the waste would be moved to a permanent repository at a later time.

A distinction is sometimes made between “disposal” and “storage.” The former suggests permanence, whereas the latter suggests the possibility that the spent fuel might be later retrieved. This possibility is made explicit in *retrievable* storage systems, where the permanent sealing of the repository is deferred, allowing the spent fuel to be recovered should this be desired at a later time.²⁶ In this case, the reprocessing option is not foreclosed, and the spent fuel may ultimately not be a “waste.”

There are several motivations for maintaining retrievability: (a) It allows for remedial action in case surprises are encountered in the first decades of waste storage that require modifying the fuel package or the repository; (b) it keeps open the option of recovering plutonium from the fuel; and (c) it allows the recovery of other materials deemed useful—for example, fission products for use in medical diagnosis and therapy or in the irradiation of food or sewage sludge. When the placement becomes irreversible, with no prospect of retrieving the fuel, this becomes final disposal.

9.4.2 Reprocessing

Extraction of Plutonium and Uranium

The alternative to disposing of the spent fuel is to reprocess it and extract at least the uranium and plutonium. In reprocessing, the spent fuel is dissolved in acid and the plutonium and uranium are chemically extracted into separate streams, for use in new fuel. The most widely used method for this is the suggestively named PUREX process.

Most early U.S. plans for reprocessing assumed that 99.5% of the U and Pu would be removed. The remainder constitutes the high-level waste. In the traditional plans, the wastes include almost all of the nonvolatile fission products, 0.5% of the uranium and plutonium, and almost all of the minor actinides [i.e., neptunium ($Z = 93$), americium ($Z = 95$), and curium ($Z = 96$)]. The uranium represents most of the mass of the spent fuel, but the fission products contain most of the radioactivity.

Extraction can be more complete than contemplated in the original U.S. thinking. The French program has exceeded the 99.5% goal, separating out more than 99.9% of the uranium and 99.8% of the plutonium [33, p. 28]. There is no essential reason to limit extraction to plutonium and uranium,

²⁶ Plans for the Yucca Mountain repository call for it to remain open for perhaps as much as several hundred years, but the preservation of the reprocessing option does not now appear to be the major motivating factor (see Section 12.2.1).

although the former represents the valuable fuel and the latter represents the bulk of the mass. It is possible to extract other radioisotopes as well, either because they are deemed pernicious as components of the waste or because they are useful in other applications. The minor actinides have been of particular interest. They include long-lived products whose removal would decrease the long-term activity in the waste. One option is to separate them and return them to a reactor where they would be transmuted in neutron reactions.

Of course, if the chief goal is safety, it is necessary to balance the benefits from decreased activity in the wastes against the increased hazards of handling and processing them when they are still very hot. At present, this further separation option has not been adopted in the major reprocessing programs in France and the United Kingdom, and the minor actinides remain with the fission products [34, p. 149].

The residue of reprocessing constitutes the wastes. They are to be put in solid form for eventual disposal. The standard method is to mix the high-level waste with molten borosilicate glass and contain the solidified glass in metal canisters. Although other solid waste forms have been suggested, borosilicate glass has been used in the French nuclear program and had figured prominently in the original U.S. plans for reprocessing commercial wastes. It is being used for the sequestering of already reprocessed U.S. military wastes at the Savannah River site in South Carolina and is planned for the wastes at the Hanford reservation in Washington state.

Status of Reprocessing Programs

Until the late 1970s, reprocessing had been planned as part of the U.S. nuclear power program. A reprocessing facility at West Valley, New York was in operation from 1966 to 1972, with a capacity of 300 MTHM/yr. This is enough, roughly speaking, for the output of 10 large reactors. There were plans for further facilities at Morris (Illinois) and Barnwell (South Carolina) which would have substantially increased the reprocessing capacity. However, all these plans have been abandoned.²⁷

In part, the abandonment was impelled by technical difficulties. There had been high radiation exposures of workers at West Valley and the plant was shut down in 1972; plans to remodel and expand it were later aborted. When the Morris plant was first tested with nonradioactive materials, it did not perform reliably, and the General Electric Co., which was building the plant, decided there were serious difficulties. The Barnwell plant moved ahead until the early 1980s, but it faced problems of meeting increasingly strict standards on permissible radioactive releases.

These difficulties might have been surmounted had there been a belief that reprocessing was needed. However, the fundamental motivation for re-

²⁷ For a discussion of this history, see Ref. [35].

processing began to slip away. There was no prospective near-term shortage in uranium supply, and uranium prices were low enough to remove the economic incentive for reprocessing. Further, an important body of opinion had developed in the United States against reprocessing, on the grounds that it might make plutonium too readily available for diversion into destructive devices.

This view was expressed in the 1977 report *Nuclear Power Issues and Choices*, sponsored by the Ford Foundation and authored by an influential group of national science policy leaders. In its conclusions on reprocessing, the report stated:

[T]he most severe risks from reprocessing and recycle are the increased opportunities for the proliferation of national weapons capabilities and the terrorist danger associated with plutonium in the fuel cycle.

In these circumstances, we believe that reprocessing should be deferred indefinitely by the United States and no effort should be made to subsidize the completion or operation of existing facilities. The United States should work to reduce the cost and improve the availability of alternatives to reprocessing worldwide and seek to restrain separation and use of plutonium. [36, p. 333]

Consistent with this thinking, the Carter administration decided in 1977 to “defer indefinitely the commercial reprocessing and recycling of the plutonium produced in U.S. nuclear power programs” [37, p. 54]. Work on U.S. reprocessing plants for commercial fuel was phased out, culminating in the closing of Barnwell at the end of 1983 [35, p. 124].

Nonetheless, reprocessing has been pursued in other countries. The largest reprocessing programs are in France and the United Kingdom, both of which completed major expansions of reprocessing capacity in 1994 to handle both domestic and foreign fuel. In addition, a large facility is being built in Japan. France has the most fully developed fuel cycle. Although much of its present reprocessing capacity is devoted to foreign orders, it also has a program of reprocessing and plutonium recycle of domestic fuel.

Table 9.4 lists the reprocessing plants in operation or under construction. It omits plants that were closed down before 2002, including plants in Belgium, France, Germany, the United Kingdom, and the United States.

Use of Mixed-Oxide Fuel

The fuel manufactured from the output of the reprocessing phase is generally a mixture of plutonium oxides and uranium oxides, with 3% to 7% PuO₂ and the remainder UO₂. It is called a *mixed-oxide fuel* or MOX. At the higher ²³⁹Pu enrichments, a burnable poison would be added to the fuel to reduce its initial reactivity. Due to differences in the nuclear properties of ²³⁹Pu and ²³⁵U, most LWRs are limited to using only about a one-third fraction of MOX

Table 9.4. Reprocessing plants for commercial nuclear fuel in operation or under construction, 2002.

Country	Location	Year of Start-up	Capacity (MTHM/yr) ^a
In operation			
France	La Hague (UP2) ^a	1976	800
France	La Hague (UP3)	1989	800
India	Tarapur	1974	100
India	Kalpakkam	1998	100
Japan	Tokai-mura	1977	90
Russia	Chelyabinsk	1984	400
United Kingdom	Sellafield (B205)	1964	1500
United Kingdom	Sellafield (Thorp)	1994	1200
Under construction			
China	Diwopu	2002 (?)	25–50
Japan	Rokkasho-Mura	2005	800

^aUP2 was upgraded and redesignated as UP2-800, with full capacity reached in 1994 [40].

Sources: Refs. [1, p. 46]; [38, Table 4]; and [39, p. 119].

in the reactor core, with the remainder ordinary uranium-oxide fuel [41, 42].²⁸ Some LWRs, however, have been designed to accommodate a full load of MOX fuel.²⁹

By 2001, about 20 PWRs in France (out of 58) were using MOX for one-third of their fuel [34, p. 138]. In the United States, the interest in MOX fuel has been motivated by the need to dispose of plutonium from dismantled nuclear weapons (see Section 18.3.3). Toward this end, the DOE is planning to build facilities for conversion of plutonium into MOX fuel at its Savannah River site. At least one nuclear plant operator (Duke Energy) has made a

²⁸ It is more difficult to control a thermal reactor using plutonium than one using uranium. Contributing reasons include (a) the delayed neutron fraction, β , is smaller for ^{239}Pu than for ^{235}U and (b) the fission cross section resonance in ^{239}Pu near 0.3 eV (see Figure 6.1) leads to a positive feedback if the reactor temperature rises. In addition, with ^{239}Pu , the neutron and gamma-ray spectra are more energetic than with ^{235}U , causing more radiation damage [42, p. 119]. As a result, it is necessary to have design changes, including more control rods, if a full load of MOX fuel is used in place of uranium fuel. This cannot be readily accomplished in most LWRs. However, it is possible in the so-called System-80 PWRs. Three such reactors are in operation at the Palo Verde nuclear plant in Arizona, but, at present, no U.S. LWR is licensed by the NRC to operate with MOX fuel.

²⁹ See Section 18.3 for a further discussion of MOX fuel, in the context of the burning of plutonium from dismantled nuclear weapons.

commitment to use MOX fuel in some of its reactors, beginning in 2007 if plans proceed according to the initial schedule [43].

9.4.3 Alternative Reprocessing and Fuel Cycle Candidates

Advanced Aqueous Process

In the widely used PUREX process, the plutonium and uranium are extracted and the fission products and minor actinides constitute the wastes. The advanced aqueous process is a modification of the PUREX process in which the minor actinides are recovered as well. Uranium is crystallized out at an early stage to reduce the bulk of the material that must be dealt with in the further chemical processing [44, p. 60]. The two product streams, one of uranium and the other of plutonium and the minor actinides, are used to fabricate fuel for use in either thermal or fast reactors.

The UREX Process

An alternative to the advanced aqueous process is the uranium extraction process (UREX and UREX+). It differs in the means of separating out the uranium. Several output streams are specifically identified in this process [45, p. II-3]:

1. Uranium. The uranium is extracted in very pure form (“at purity levels of 99.999 percent”). This leaves it free of highly radioactive contaminants and makes it easy to handle for disposal or reuse in a reactor.
2. Plutonium and minor actinides. Neptunium, americium, and curium are retained with the plutonium. These elements can be incorporated into the reactor fuel.
3. Long-lived fission products. Long-lived fission products (in particular, iodine-129 and technetium-99) are separately extracted, for destruction in a reactor (see Section 11.3.3).
4. Other fission products. These become the wastes. The waste disposal problem is simplified because the long-lived radionuclides have, for the most part, been removed.

Pyroprocessing

The above-discussed chemical reprocessing processes are known as *aqueous* processes. An alternative approach, under active exploration for use in conjunction with future reactors, is the *pyroprocess* or electrorefining process. In this method, the spent fuel is dissolved at very high temperatures in molten cadmium, creating an “electrolytic bath.” Groups of chemical elements are separately extracted on the basis of differences in the potentials at which they dissolve and ionize. In particular, ions of the actinides, including uranium,

plutonium, and the minor actinides, are attracted to cathodes and are extracted. The actinides are then incorporated in the fabrication of new fuel elements.

Full Actinide Recycle

A fuel cycle based on the nearly complete extraction of plutonium and minor actinides (collectively, the transuranic elements) and their consumption by fission in fast reactors has been sketched in the MIT report *The Future of Nuclear Power* [46]. This cycle envisages a global nuclear economy in 2050 with a capacity of 1500 GWe based on a balanced combination of thermal and fast reactors. The thermal reactors are assumed to be LWRs, fueled by enriched uranium oxide. The fast reactors, undefined as to type, are fueled by transuranics obtained from the LWR spent fuel. Pyroprocessing is used to extract the transuranics from both the thermal and fast reactor spent fuel. For each load of fresh fuel in the fast reactors, 20% of the transuranics are consumed in fission and the remaining 80% are available for recycle.

A balanced system is one in which the spent fuel from the thermal reactors provides the transuranics needed to make up for those consumed in the fast reactors. With the assumptions made in the MIT analysis, this is achieved by having slightly more capacity in the thermal reactors than in the fast reactors (815 GWe and 685 GWe, respectively). A variety of choices exist for the fast reactors, including some of the Generation IV reactors discussed in Section 16.6.

The uranium requirement for the entire fuel cycle is the amount needed to provide for the 815 GWe of LWRs, which is 54% of the amount needed if LWRs accounted for the full 1500-GWe capacity. A further, and perhaps even more important, benefit is the almost complete elimination of plutonium and minor actinides from the stream of wastes that require permanent disposal.

General Features of Reprocessing Options

Any fuel cycle that recycles the fissile components of the spent fuel (mainly the remaining ^{235}U and the plutonium isotopes ^{239}Pu and ^{241}Pu), increases the energy obtained from the existing uranium resources. If the minor actinides are included with the uranium and plutonium in the new fuel, the wastes will have much less long-term radioactivity than wastes in the once-through fuel cycle. The mass of the spent fuel is greatly reduced if the uranium is either returned to the reactor or is separated from other radionuclides to become low-activity depleted uranium. The fission products then constitute the waste product that requires long-term disposal. This greatly reduces the mass of the waste product and the period during which it must be kept isolated from the environment.

All of the reprocessing fuel cycles that have been described in this section achieve these resource extension and waste reduction benefits. The PUREX process accomplishes much of this, but in its standard form the minor actinides are not removed.

A long-standing objection to reprocessing is based on the increased proliferation risks if ^{239}Pu is in wide circulation. The above-described methods lessen the risks in two ways: (1) The presence of the minor actinides increases the activity of the fuel and makes it more difficult to handle, and (2) the reprocessing and fuel fabrication facilities can be located adjacent to the reactor, making theft or diversion of the fuel very difficult without the collaboration of the plant operators. The collocation aspect was particularly stressed in planning documents for the Integral Fast Reactor (see Section 16.5.1) and combined facilities were tested on a small scale using fuel from the experimental breeder reactor in Idaho (EBR-II).

A further objection is based on economics. Given present demand and prices, it is more expensive to reprocess spent fuel than to obtain fuel from newly mined uranium. This is a cogent objection at the present scale of nuclear power use. The advantages of these reprocessing approaches become more relevant in the context of a possible major expansion of nuclear power.

At present, the reprocessing approaches discussed here are in the development and study stage, except for the long-used PUREX process. In general, the pyroprocessing technique is more suitable for use with fuel in metallic form, while the aqueous processes are more suitable for oxide fuels. Thus pyroprocessing was originally studied for use with metallic fuel from a sodium-cooled fast reactor. However, either class of process could be used with a wide variety of fuel forms, given appropriate pretreatment stages.

9.4.4 Waste Disposal

All countries with announced plans for disposing of high-level radioactive wastes are planning on eventual disposal in deep geologic repositories, typically made by excavating caverns or holes in favorable environments. Many of the plans for these permanent disposal facilities include a period during which the waste could still be retrieved.

Deep geologic disposal has been the favored course in U.S. thinking since the first attempts to formulate plans. There have been continuing efforts to locate and design a suitable facility. A site at Yucca Mountain in Nevada was selected in 1987 as the candidate for a U.S. repository and it has been under intense study since. The DOE in 2002, with the subsequent concurrence of the president and Congress, recommended going ahead with the Yucca Mountain project. The announced goal is to have a facility ready to receive wastes by 2010, subject to approval by the Nuclear Regulatory Commission. (Much more extensive discussions of nuclear wastes are presented in Chapters 10–13.)