# 2. OBJECTIVES

12. Three safety objectives are defined for nuclear power plants. The first is very general in nature. The other two are complementary objectives that interpret the general objective, dealing with radiation protection and technical aspects of safety respectively. The safety objectives are not independent; their overlap ensures completeness and adds emphasis.

# 2.1. GENERAL NUCLEAR SAFETY OBJECTIVE

13. Objective: To protect individuals, society and the environment by establishing and maintaining in nuclear power plants an effective defence against radiological hazard.

14. Each viable method of production of electricity has unique advantages and possible detrimental effects. In the statement of the general nuclear safety objective, radiological hazard means adverse health effects of radiation on both plant workers and the public, and radioactive contamination of land, air, water or food products. It does not include any of the more conventional types of hazards that attend any industrial endeavour. The protection system is effective as stated in the objective if it prevents significant addition either to the risk to health or to the risk of other damage to which individuals, society and the environment are exposed as a consequence of industrial activity already accepted. In this application, risk is defined as the arithmetic product of the probability of an accident or an event and the adverse effect it would produce. These health risks are to be estimated without taking into account the countervailing and substantial benefits which the nuclear and industrial activities bestow, both in better health and in other ways important to modern civilization. When the objective is fulfilled, the level of risk due to nuclear power plants does not exceed that due to competing energy sources, and is generally lower. If another means of electricity generation is replaced by a nuclear plant, the total risk will generally be reduced. The comparison of risks due to nuclear plants with other industrial risks to which people and the environment are exposed makes it necessary to use calculational models in risk analysis. To make full use of these techniques and to support implementation of this general nuclear safety objective, it is important that quantitative targets, 'safety goals', are formulated.

15. It is recognized that although the interests of society require protection against the harmful effects of radiation, they are not solely concerned with the radiological

safety of people and the avoidance of contamination of the environment. The protection of the resources invested in the plant is of high societal importance and demands attention to all the safety issues with which this report is concerned. However, the main focus of this document is the safety of people. What follows is therefore expressed in these terms solely, but this is not to imply that INSAG has no regard for other factors.

# 2.2. RADIATION PROTECTION OBJECTIVE

16. Objective: To ensure in normal operation that radiation exposure within the plant and due to any release of radioactive material from the plant is kept as low as reasonably achievable and below prescribed limits, and to ensure mitigation of the extent of radiation exposures due to accidents.

17. Radiation protection is provided in nuclear power plants under normal conditions and separate measures would be available under accident circumstances. For planned plant operating conditions and anticipated operational occurrences, compliance with radiation protection standards based on ICRP recommendations<sup>2</sup> ensures appropriate radiation protection. That is, the ICRP's system of dose limitation provides appropriate protection for planned situations anticipated to occur once or more in the lifetime of a plant.

18. The aforementioned radiation protection standards have been developed to prevent harmful effects of ionizing radiation by keeping exposures sufficiently low that non-stochastic effects are precluded and the probability of stochastic effects is limited to levels deemed tolerable. This applies to controlled circumstances. In the event of any accident that could cause the source of exposure to be not entirely under control, safety provisions in the plant are planned and countermeasures outside the plant are prepared to mitigate harm to individuals, populations and the environment.

<sup>&</sup>lt;sup>2</sup> For example INTERNATIONAL ATOMIC ENERGY AGENCY, Basic Safety Standards for Radiation Protection (1982 edn), Safety Series No. 9, IAEA, Vienna (1982).

# 2.3. TECHNICAL SAFETY OBJECTIVE

19. Objective: To prevent with high confidence accidents in nuclear plants; to ensure that, for all accidents taken into account in the design of the plant, even those of very low probability, radiological consequences, if any, would be minor; and to ensure that the likelihood of severe accidents with serious radiological consequences is extremely small.

20. Accident prevention is the first safety priority of both designers and operators. It is achieved through the use of reliable structures, components, systems and procedures in a plant operated by personnel who are committed to a strong safety culture.

21. However, in no human endeavour can one ever guarantee that the prevention of accidents will be totally successful. Designers of nuclear power plants therefore assume that component, system and human failures are possible, and can lead to abnormal occurrences, ranging from minor disturbances to highly unlikely accident sequences. The necessary additional protection is achieved by the incorporation of many engineered safety features into the plant. These are provided to halt the progress of an accident in the specific range of accidents considered during design and, when necessary, to mitigate its consequences. The design parameters of each engineered safety feature are defined by a deterministic analysis of its effectiveness against the accidents it is intended to control. The accidents in the spectrum requiring the most extreme design parameters for the safety feature are termed the design basis accidents for that feature.

22. Attention is also directed to accidents of very low likelihood but more severe than those considered explicitly in the design (accidents 'beyond the design basis'). Some of these severe accidents could cause such deterioration in plant conditions that proper core cooling cannot be maintained, or that fuel damage occurs for other reasons. These accidents would have a potential for major radiological consequences if radioactive materials released from the fuel were not adequately confined. As a result of the accident prevention policy, they are of low probability of occurrence.

23. Since these accidents could nonetheless occur, other procedural measures are provided for managing their course and mitigating their consequences. These additional measures are defined on the basis of operating experience, safety analysis and the results of safety research. Attention is given in design, siting, procedures and training to controlling the progression and consequences of accidents. Limitation of

accident consequences requires measures to ensure sale situation, ensure cooling, adequate confinement integrity and off-site emergency preparedness. High consequence severe accidents are therefore extremely unlikely because they are effectively prevented or mitigated by defence in depth.

24. In the safety technology of nuclear power, risk is defined (as in Section 2.1) as the product of the likelihood of occurrence of an accident and its potential radiological consequences. The technical safety objective for accidents is to apply accident prevention, management and mitigation measures in such a way that overall risk is very low and no accident sequence, whether it is of low probability or high probability, contributes to risk in a way that is excessive in comparison with other sequences.

25. The target for existing nuclear power plants consistent with the technical safety objective is a likelihood of occurrence of severe core damage that is below about  $10^{-4}$  events per plant operating year. Implementation of all safety principles at future plants should lead to the achievement of an improved goal of not more than about  $10^{-5}$  such events per plant operating year. Severe accident management and mitigation measures should reduce by a factor of at least ten the probability of large off-site releases requiring short term off-site response.

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26. A number of concepts are general in application, bearing in many important ways on the nature and application of the specific safety principles enunciated later. These important concepts are here called fundamental safety principles and they are identified in Section 3. They are of three kinds, relating to management, defence in depth and technical issues.

### 3.1. MANAGEMENT RESPONSIBILITIES

27. Three fundamental management principles are identified. They are connected with the establishment of a safety culture, the responsibilities of the operating organization, and the provision of regulatory control and verification of safety related activities.

# 3.1.1. Safety culture

28. Principle: An established safety culture governs the actions and interactions of all individuals and organizations engaged in activities related to nuclear power.

29. The phrase 'safety culture' refers to a very general matter, the personal dedication and accountability of all individuals engaged in any activity which has a bearing on the safety of nuclear power plants. The starting point for the necessary full attention to safety matters is with the senior management of all organizations concerned. Policies are established and implemented which ensure correct practices, with the recognition that their importance lies not just in the practices themselves but also in the environment of safety consciousness which they create. Clear lines of responsibility and communication are established; sound procedures are developed; strict adherence to these procedures is demanded; internal reviews are performed of safety related activities; above all, staff training and education emphasize the reasons behind the safety practices established, together with the consequences for safety of shortfalls in personal performance.

30. These matters are especially important for operating organizations and the staff directly engaged in plant operation. For the latter, at all levels, training emphasizes the significance of their individual tasks from the standpoint of basic understanding

sis on the reasons underlying safety limits and the safety consequences of violations. Open attitudes are required in such staff to ensure that information relevant to plant safety is freely communicated; when errors of practice are committed, their admission is particularly encouraged. By these means, an all pervading safety thinking is achieved, allowing an inherently questioning attitude, the prevention of complacency, a commitment to excellence, and the fostering of both personal accountability and corporate self-regulation in safety matters.

# 3.1.2. Responsibility of the operating organization

31. Principle: The ultimate responsibility for the safety of a nuclear power plant rests with the operating organization. This is in no way diluted by the separate activities and responsibilities of designers, suppliers, constructors and regulators.

32. Once the operating organization accepts possession, it is in complete charge of the plant, with full responsibility and commensurate authority for approved activities in the production of electric power. Since these activities also affect the safety of the plant, the operating organization establishes policy for adherence to safety requirements, establishes procedures for safe control of the plant under all conditions, including maintenance and surveillance, and retains a competent, fit and fully trained staff. The operating organization ensures that responsibilities are well defined and documented and that the resources and facilities for the tasks of its staff are in place.

33. The operating organization also has responsibilities in certain areas where its control is less direct. By using its own staff and resources, or through agencies acting on its behalf, the operating organization institutes rigorous reviews, audits and, as necessary, approval processes to ensure that the factors which determine the safety of the plant are given the necessary attention. This applies, for example, to site investigation, design, manufacturing, construction, testing and commissioning.

34. This principle of the operating organization's overriding safety responsibility is a prime one. The responsibilities of other parties are also significant for safety as well as for financial and legal matters. Variations in national practices make it difficult to define the formal responsibilities of the other parties, but clearly designers, manufacturers and constructors are required as a minimum to provide a sound design and equipment that meets its specifications in terms of both engineering detail and performance of the intended function, meeting or exceeding quality standards commensurate with the safety significance of components or systems. The technical societies and the scientific community generally carry responsibilities for high standards of performance of individuals in the professional sense, and for maintaining and strengthening the basis on which the safety of nuclear power plants. stands. The responsibilities of the regulators are discussed in Section 3.1.3.

- 3.1.3. Regulatory control and independent verification
- 35. Principle: The government establishes the legal framework for a nuclear industry and an independent regulatory organization which is responsible for licensing and regulatory control of nuclear power plants and for enforcing the relevant regulations. The separation between the responsibilities of the regulatory organization and those of other parties is clear, so that the regulators retain their independence as a safety authority and are protected from undue pressure.

36. A legally constituted regulatory organization provides governmental licensing, regulation and surveillance of the operation of nuclear power plants in respect of their safety. Activities of the regulatory organizations cover the following functional areas:

- specification and development of standards and regulations for safety;
- issue of licences to operating organizations, following appropriate safety assessments;
- inspection, monitoring and review of the safety performance of nuclear plants and operating organizations;
- requiring corrective actions of an operating organization where necessary and taking any necessary enforcement actions, including withdrawal of licence, if acceptable safety levels are not achieved;
- advocacy of safety research, as discussed in Section 3.3.6; and
- dissemination of safety information (also discussed in Section 3.3.6).

37. The regulatory organization acts independently of designers, constructors and operators to the extent necessary to ensure that safety is the only mission of the regulatory personnel. The resources of the regulatory organization are sufficient for it to accomplish its functions without adversely affecting construction schedules or energy production, except where warranted for the assurance of safety. Expertise in a sufficiently wide range of nuclear technologies is available to the regulatory organization.

38. To fulfil its functions effectively, the regulatory organization has the necessary legal authority, and it is provided with free access to facilities and to relevant information in the possession of the operating organization.

# 3.2. STRATEGY OF DEFENCE IN DEPTH

39. 'Defence in depth' is singled out amongst the fundamental principles since it underlies the safety technology of nuclear power. All safety activities, whether organizational, behavioural or equipment related, are subject to layers of overlapping provisions, so that if a failure should occur it would be compensated for or corrected without causing harm to individuals or the public at large. This idea of multiple levels of protection is the central feature of defence in depth, and it is repeatedly used in the specific safety principles that follow.

40. Two corollary principles of defence in depth are defined, namely, accident prevention and accident mitigation. These corollary principles follow the general statement of defence in depth.

# 3.2.1. Defence in depth

41. Principle: To compensate for potential human and mechanical failures, a defence in depth concept is implemented, centred on several levels of protection including successive barriers preventing the release of radioactive material to the environment. The concept includes protection of the barriers by averting damage to the plant and to the barriers themselves. It includes further measures to protect the public and the environment from harm in case these barriers are not fully effective.

42. The defence in depth concept provides an overall strategy for safety measures and features of nuclear power plants. When properly applied, it ensures that no single human or mechanical failure would lead to injury to the public, and even combinations of failures that are only remotely possible would lead to little or no injury. Defence in depth helps to establish that the three basic safety functions (controlling the power, cooling the fuel and confining the radioactive material) are preserved, and that radioactive materials do not reach people or the environment.

43. The principle of defence in depth is implemented primarily by means of a series of barriers which should in principle never be jeopardized, and which must be violated in turn before harm can occur to people or the environment. These barriers are physical, providing for the confinement of radioactive material at successive locations. The barriers may serve operational and safety purposes, or may serve safety purposes only. Power operation is only allowed if this multibarrier system is not jeopardized and is capable of functioning as designed.

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44. The reliability of the physical barriers is enhanced by applying the concept of defence in depth to them in turn, protecting each of them by a series of measures. Each physical barrier is designed conservatively, its quality is checked to ensure that the margins against failure are retained, its status is monitored, and all plant processes capable of affecting it are controlled and monitored in operation. Human aspects of defence in depth are brought into play to protect the integrity of the barriers, such as quality assurance, administrative controls, safety reviews, independent regulation, operating limits, personnel qualification and training, and safety culture. Design provisions including both those for normal plant systems and those for engineered safety systems help to prevent undue challenges to the integrity of the physical barriers, to prevent the failure of a barrier if it is jeopardized, and to prevent consequential damage of multiple barriers in series. Safety system designers ensure to the extent practicable that the different safety systems protecting the physical barriers are functionally independent under accident conditions.

45. All of the components of defence are available at all times that a plant is at normal power. Appropriate levels are available at other times. The existence of several components of defence in depth is never justification for continued operation in the absence of one component. Severe accidents in the past have been the result of multiple failures, both human and equipment failures, due to deficiencies in several components of defence in depth that should not have been permitted.

46. System design according to defence in depth includes process controls that use feedback to provide a tolerance of any failures which might otherwise allow faults or abnormal conditions to develop into accidents. These controls protect the physical barriers by keeping the plant in a well defined region of operating parameters where barriers will not be jeopardized. Care in system design prevents cliff edge effects which might permit small deviations to precipitate grossly abnormal plant behaviour and cause damage.

47. Competent engineering of the barriers and the measures for their protection coupled with feedback to maintain operation in optimal ranges leads to a record of smooth, steady performance in producing electricity on demand. This indicates the proper implementation of the most important indicator of the success of defence in depth, which is operation with little or no need to call on safety systems.

48. The multibarrier system protects humans and the environment in a wide range of abnormal conditions. Preplanned countermeasures are provided, as a further component of defence in depth, against the possibility that radioactive material might still be released from the plant.

49. The Appendix presents a discussion of the means by which the separate components of defence in depth protect and complement each other. The importance of prevention and mitigation of accidents in defence in depth is treated in the following two corollaries.

#### 3.2.2. Accident prevention

50. Principle: Principal emphasis is placed on the primary means of achieving safety, which is the prevention of accidents, particularly any which could cause severe core damage.

51. The design, construction, operation and maintenance of nuclear power plants has as its primary objective the generation of electricity reliably and economically. In accordance with the general safety management principle on safety culture, the safety implications of decisions in all these areas must be borne in mind. The following is concentrated on these safety aspects.

52. The first means of preventing accidents is to strive for such high quality in design, construction and operation of the plant that deviations from normal operational states are infrequent. Safety systems are used as a backup to feedback in process control to prevent such deviations from developing into accidents. Safety systems make use of redundancy and diversity of design and the physical separation of parallel components, where appropriate, to reduce the likelihood of the loss of a vital safety function. Systems and components are inspected and tested regularly to reveal any degradation which might lead to abnormal operating conditions or inadequate safety system performance. Abnormal conditions possibly affecting nuclear safety are promptly detected by monitoring systems that give alarms and in many cases initiate corrective actions automatically. The operators are trained to recognize readily the onset of an accident and to respond properly and in a timely manner to such abnormal conditions. They have also been well trained in appropriate operating procedures, with which they have become familiarized.

53. Thus the prevention of accidents depends on conservatively designed equipment and good operational practices to prevent failure, quality assurance to verify the achievement of the design intent, surveillance to detect degradation or incipient failure during operation, and steps to ensure that a small perturbation or incipient failure would not develop into a more serious situation.

54. A number of probabilistic safety assessments have been made for a range of nuclear power plant designs in different countries. They show that sufficiently low probabilities of severe core damage are attainable. When effective preparation for accident management and for mitigation of the effects of severe accidents is taken into account, the results of these probabilistic safety assessments are consistent with the general nuclear safety objective in Section 2.1.

55. Probabilistic safety assessment also guides design and operation by identifying potential accident sequences that could contribute excessively to risk. Measures can then be taken to reduce this contribution.

#### 3.2.3. Accident mitigation

56. Principle: In-plant and off-site mitigation measures are available and are prepared for that would substantially reduce the effects of an accidental release of radioactive material.

57. Provisions for accident mitigation extend the defence in depth concept beyond accident prevention. The accident mitigation provisions are of three kinds, namely, accident management, engineered safety features and off-site countermeasures.

58. Accident management includes preplanned and ad hoc operational practices which, in circumstances in which the design specifications of the plant are exceeded, would make optimum use of existing plant equipment in normal and unusual ways to restore control. This phase of accident management would have the objective of restoring the plant to a safe state with the reactor shut down, continued fuel cooling assured, radioactive material confined and the confinement function protected. In such circumstances, engineered safety features would act to confine any radioactive material released from the core so that discharge to the environment would be minimal. These engineered safety features include physical barriers, some of which have the single purpose of confining radioactive material. Off-site countermeasures are available, going beyond the level of protection provided in most human endeavours, to compensate for the remote possibility that safety measures at the plant might fail. In such a case, the effects on the surrounding population or the environment would be mitigated by protective actions, such as sheltering or evacuation of the population, and by prevention of the transfer of radioactive material to man by food-chains and other pathways.

# 3.3. GENERAL TECHNICAL PRINCIPLES

59. There are several underlying technical principles which are essential to the successful application of safety technology for nuclear power plants.

# 3.3.1. Proven engineering practices

60. Principle: Nuclear power technology is based on engineering practices which are proven by testing and experience; and which are reflected in approved codes and standards and other appropriately documented statements.

61. Systems and components are conservatively designed, constructed and tested to quality standards commensurate with the safety objectives. Approved codes and standards are used whose adequacy and applicability have been assessed and which have been supplemented or modified if necessary. If opportunities for advancement or improvement over existing practices are available and seem appropriate, such changes are applied cautiously.

62. Numerous codes and standards have been adopted for nuclear use, after formulation by the professional engineering community and approval by the appropriate agencies. Some existing codes and standards have been modified from an original form to take into account unique features of their use for nuclear plants and the elevated importance assigned to the safety of nuclear plants. Approved codes have the simultaneous objectives of reliability and safety. They are based on principles proven by research, past application, testing and dependable analysis<sup>3</sup>.

63. Well established manufacturing and construction methods are used. Dependence on experienced and approved suppliers contributes to confidence in the performance of important components. Deviations from previously successful manufacturing and construction practices are approved only after demonstration that the alternatives meet the requirements. Manufacturing and construction quality is ensured through the use of appropriate standards and by the proper selection, training and qualification of workers. The use of proven engineering continues throughout the plant's life. When repairs and modifications are made, analysis is conducted and review is made to ensure that the system is returned to a configuration covered in the safety analysis and technical specifications. Where new and unreviewed safety questions are posed, new analysis is conducted.

64. The design and construction of new types of power plants are based as far as possible on experience from earlier operating plants or on the results of research programmes and the operation of prototypes of an adequate size.

# 3.3.2. Quality assurance

65. Principle: Quality assurance is applied throughout activities at a nuclear power plant as part of a comprehensive system to ensure with high confidence that all items delivered and services and tasks performed meet specified requirements.

66. The comprehensive system referred to in the principle begins with analysis and design in accordance with the preceding principle on proven engineering, and it continues into the use of quality assurance methods. Other fundamental technical safety principles are also important in this respect, particularly those on safety assessment and verification and on operating experience and safety research.

<sup>&</sup>lt;sup>3</sup> The IAEA's NUSS series of documents has been developed in accordance with this principle.

28 Busic Theory

# Prerequisite Concepts

Energy Forms Basic Chemistry Basic SI Units

Appendix II

The characteristics of the radioactive decay and nuclear reaction processes have been the driving forces behind nearly every unique design feature of nuclear energy systems. Thus, familiarity with some of the fundamental principles of nuclear physics is essential to understanding nuclear technology.

Radioactive decay and nuclear reactions are unique in that they

1. provide clear evidence that mass and energy can be interconverted

- 2. involve a variety of particles and radiations, which often have discrete, or quantized, energies
- 3. require descriptive formulations based on laws of probability

Such characteristics have prompted the development of many new experimental and analytical methods.

# THE NUCLEUS

The atom is the basic unit of matter. As first modeled by Niels Bohr in 1913, it consists of a heavy central *nucleus* surrounded by orbital *electrons*. The nucleus, in turn, consists of two types of particles, namely *protons* and *neutrons*. Table 2-1 compares the atom and its constituents in terms of electric charge and mass.<sup>†</sup>

The proton and electron are of exactly opposite charge. A complete atom has the same number of protons and electrons, each given by the *atomic number* Z. The electrostatic [Coulomb] attractive forces between the oppositely charged particles is

<sup>†</sup>Unless otherwise noted, data here and in the remainder of the book are based on the General Electric Chart of the Nuclides (GE/Chart, 1989). The definition for the "amu" is provided at the end of this section.

#### TABLE 2-1

Characteristics of Atomic and Nuclear Constituents

Constituent	Churge (e) <sup>†</sup>	Mass (anu)‡	Radius (m)
Electron		5.5 × 10 <sup>-4</sup>	· ·
Proton	+1	1,007276	
Neutrun	0	1.008665	
Nucleus	+ 2 \$	~ 41	~ 10 1
Alom	0	~ A	~ 10 11

<sup>†</sup>e = 1.6022 × 10<sup>−19</sup> C.

 $t_{amu} = 1.6606 \times 10^{-22} \text{ kg}.$ 

 $\frac{1}{2}$  = atomic number,

 $\P_A$  = atomic mass number.

the basis for the electron orbits (in a manner similar to the way solar system orbits result from gravitational forces).

Although the atom is electrically neutral, the number and resulting configuration of the orbital electrons uniquely determine the chemical properties of the atom, and hence its identity as an *element*. It may be recalled that the atomic number is the basis for alignment in the periodic table of elements.

# Structure

Very strong, short-range forces override the Coulomb repulsive forces to bind the positively charged protons (along with the uncharged neutrons) into the compact nucleus. The protons and neutrons in a nucleus are collectively referred to as *nucleons*. Since each nucleon has roughly the same mass, the nucleus itself has a mass that is nearly proportional to the *atomic mass number A*, defined as the total number of nucleons. The electrons are very light compared to the particles in the nucleus, so the mass of the atom is nearly that of the nucleus.

The characteristic dimensions of the nucleus and atom are listed in Table 2-1. The latter value is based on the effective radius of the outer electron orbits in arrays of atoms or molecular combinations.

A useful shorthand notation for nuclear species or *nuclides* is  $\frac{1}{2}X$ , where X is the chemical symbol, Z is the atomic number, and A is the atomic mass number. (An alternative formulation,  $_{Z}X^{A}$ , is also found, although current practice favors retention of the upper right-hand location for charge-state information.) The subscript Z is actually redundant once the chemical element has been identified; its use is discretionary.

Different nuclides of a single chemical element are called *isotopes*. For example, uranium isotopes  ${}^{233}_{92}U$ ,  ${}^{235}_{92}U$ , and  ${}^{238}_{92}U$  were mentioned in the last chapter. Each has 92 protons and electrons with 141, 143, and 146 neutrons, respectively. Another important isotope group is the hydrogen family— ${}^{1}_{1}H$ ,  ${}^{2}_{1}H$ , and  ${}^{3}_{1}H$ . The latter two are the only isotopes often given separate names and symbols—deuterium  ${}^{2}_{1}D$  and tritium  ${}^{1}_{1}T$ , respectively.

# **Binding Energy**

One of the most startling observations of nuclear physics is that the mass of an atom is less than the sum of the masses of the individual constituents. When all parts are assembled, the product atom has "missing" mass, or a mass defect  $\Delta$ , given by

$$\Delta = [Z(m_{\rm p} + m_{\rm e}) + (A - Z) m_{\rm n}] - M_{\rm strong}$$
(2-1)

where the masses  $m_p$ ,  $m_e$ , and  $m_n$  of the proton, electron, and neutron, respectively, are multiplied by the number present in the atom of mass  $M_{atom}$ .

The defect mass is converted into energy at the time the nucleus is formed.<sup>†</sup> The conversion is described by the expression

$$E = mc^2 \tag{2-2}$$

<sup>†</sup> Mass changes also occur with chemical binding (e.g., electrons with a nucleus to form an atom and atoms with each other to form molecules), but they are so small as to defy measurement.



Binding energy per nucleon as a function of mass number.

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for energy E, mass m, and proportionality constant  $c^2$ , where c is the speed of light in a vacuum. This simple-appearing equation (one of the world's most famous!) was developed by Albert Einstein with the "theory of relativity."

The energy associated with the mass defect is called the *binding energy*. It is said to put the atom into a "negative energy state" since positive energy from an external source would have to be supplied to disassemble the constituents. (This is comparable to the earth-moon system, which could be separated only through an addition of outside energy.) The binding energy [BE] for a given nucleus may be expressed as

$$BE = [M_{atom} - Z(m_{p} + m_{e}) - (A - Z)m_{n}]c^{2} = -\Delta c^{2}$$
(2-3)

As the number of particles in a nucleus increases, the BE also increases. The rate of increase, however, is not uniform. In Fig. 2-1, the BE in MeV\* per nucleon is plotted as a function of atomic mass number. The nuclides in the center of the range are more tightly bound on the average than those at either very low or very high masses.

The existence of the fission process is one ramification of the behavior shown in Fig. 2-1. Compared to nuclei of half its mass, the <sup>235</sup>U nucleus is bound relatively lightly. Energy must be released to split the loosely bound <sup>235</sup>U into two tightly bound fragments. A reasonably good estimate for the energy released in fission can be obtained by using data from the curve in Fig. 2-1.

Energy production in our sun and other stars is based on the fusion process, which combines two very light nuclei into a single heavier nucleus. As shown by Fig. 2-1, two deuterium nuclei [ ${}^{2}_{1}$ H or  ${}^{2}_{1}$ D], for example, could release a substantial amount of energy if combined to form the much more tightly bound helium [ ${}^{4}_{1}$ He]. A number of fusion reactions are considered as potential terrestrial nuclear energy sources in Chap. 21.

# Mass and Energy Scales

The masses and energies associated with nuclear particles and their interactions are extremely small compared to the conventional macroscopic scales. Thus, special units are found to be very useful.

The atomic mass unit [amu] is defined as 1/12 of the mass of the carbon-12 [ ${}^{12}$ C] atom.<sup>†</sup> The masses in Table 2-1 are based on this scale.

When an electron moves through an electrical potential difference of 1 volt [V], it acquires a kinetic energy of 1 *electron volt* [eV]. This unit (equal to  $1.602 \times 10^{-19}$  J, as noted in App. II), along with its multiples keV for a thousand and MeV for a million, is very convenient for nuclear systems.<sup>‡</sup>

Mass and kinetic energy, as noted previously, may be considered equivalent through the expression  $E = mc^2$ . Thus, for example, it is not uncommon to express mass differences in MeV or binding energies in anu based on the conversion 1 amu = 931.5 MeV, (Other useful factors are contained in App. 11.)

# **RADIOACTIVE DECAY**

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The interactions among the particles in a nucleus are extremely complex. Some combinations of proton and neutron numbers result in very tightly bound nuclei, while others yield more loosely bound nuclei (or do not form them at all).

Whenever a nucleus can attain a more stable (i.e., more tightly bound) configuration by emitting radiation, a spontaneous disintegration process known as *radioactive decay* may occur. (In practice this "radiation" may be actual electromagnetic radiation or may be a particle.) Examples of such processes are delayed briefly to allow for an examination of important basic principles in the following paragraphs.

#### **Conservation Principles**

Detailed studies of radioactive decay and nuclear reaction processes have led to the formulation of very useful *conservation principles*. For example, electric charge is conserved in a decay; the *total* amount is the same before and after the reaction even

<sup>†</sup>Two earlier muss scales defined the arru as 1/16 the mass of elemental oxygen and of oxygen-16, respectively. Each of these differs from the current standard, so caution is advised when using data from multiple sources {e.g., Kaplan (1962) is based on the <sup>16</sup>O scale, while the Chart of the Nuclides (GE/Chart, 1989) employs the <sup>15</sup>C scale}.

feV is usually pronounced as "ee-vee," keV as "kay-ee-vee," etc.

<sup>&</sup>quot;The MeV is a convenient unit for energy, which is defined shortly.

though it may be distributed differently among entirely different nuclides and/or particles. The four principles of most interest here are conservation of

J. charge

2. mass number or number of nucleons

total energy

4. linear and angular momentum

Conservation of electric charge implies that charges are neither created nor destroyed. Single positive and negative charges may, of course, neutralize each other. Conversely, it is also possible for a neutral particle to produce one charge of each sign.

Conservation of mass number does not allow a net change in the number of nucleons. However, the conversion of a proton to a neutron is allowed. Electrons follow a separate particle conservation law (which is beyond the scope of this discussion). By convention, a mass number of zero is assigned to electrons.

The total of the kinetic energy and the energy equivalent of the mass in a system must be conserved in all decays and reactions. This principle determines which outcomes are and are not possible.

Conservation of linear momentum is responsible for the distribution of the available kinetic energy among product nuclei, particles, and/or radiations. Angular momentum considerations for particles that make up the nucleus play a major role in determining the likelihood of occurrence of the outcomes that are energetically possible. (This latter consideration is substantially beyond the scope of this book.)

#### Natural Radioactivity

A wide range of radioactive nuclides, or *radionuclides*, exist in nature (and did so before the advent of the nuclear age). Artificial radionuclides produced by nuclear reactions are considered separately.

The naturally occurring radioactive decay processes may produce any of three radiations. Dating back to the time of their discovery and identification, the arbitrary names *alpha*, *beta*, and *gamma* are still employed.

# Alpha Radiation

Alpha radiation is a helium nucleus, which may be represented as either  $\frac{4}{2}$ He or  $\frac{4}{2}\alpha$ . An important alpha-decay process with  $\frac{23}{2}$ U is written in the form

where  ${}^{230}_{33}$ Th (thorium-231) is the decay or *daughter* product of the  ${}^{232}_{35}$ U parent nucleus. It may be noted that the reaction equation demonstrates conservation of mass number A and charge (or equivalently, atomic number Z) on both sides of the equation. Thus, once any two of the three constituents are known, the third may be determined readily.

Most alpha-emitting species have been observed to generate several discrete kinetic energies. Thus, to conserve total energy, the product nuclei must have correspondingly different masses. The discrete or *quantum* differences in energy are related to a complex energy level structure within the nucleus. (Explanation of such phenomena

is delegated to the field of quantum mechanics, which again is outside the scope of this book.)

#### Beta Radiation

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Beta radiation is an electron of nuclear, rather than orbital, origin. Since, as noted earlier, the electron has a negative charge equal in magnitude to that of the proton and has a mass number of zero, it is represented as  $_{1}^{\circ}e$  or  $_{1}^{\circ}\beta$ . A beta-decay reaction, which is important to production of plutonium (e.g., in a breeder reactor), is

$$^{2}_{33}^{3}N_{p} \rightarrow ^{2}_{34}^{39}Pu + _{1}^{0}\beta + _{0}^{0}\nu^{*}$$

where  ${}^{239}_{55}Np$  [neptunium-239] is the parent of the  ${}^{239}_{64}Pu$ , and  ${}^{9}_{6}\nu^{*}$  is an uncharged, massless "particle" called an *antineutrino*.<sup>†</sup> As required by conservation principles, the algebraic sums of both charge and mass number on each side of the equation are equal.

The nuclear basis for beta decay is

 $\ln \rightarrow \ln + \ln e + \delta \nu^*$ 

where the uncharged neutron emits an electron and an antineutrino while leaving a proton (and a net additional positive charge) in the nucleus. The slight mass difference between the neutron and proton may be noted from Table 2-1 to be sufficient to allow for electron emission as well as a small amount of kinetic energy.

The nature of the antineutrino defies the human senses. Because it has neither charge nor mass, the antineutrino does not interact significantly with other materials and is not readily detected. However, it does carry a portion of the kinetic energy that would otherwise belong to the beta particle. In any given decay, the antineutrino may take anywhere from 0 to 100 percent of the energy, with an average of about twothirds for many cases.

Analogously to alpha decay, the beta process in a given radionuclide may produce several discrete transition energies based on the energy levels in the nucleus. Here, however, a range of beta energies from the transition energy down to zero are actually observed, due to the sharing with antineutrinos.

#### Gamma Radiation

Gamma radiation is a high-energy electromagnetic radiation that originates in the nucleus. It is emitted in the form of *photons*, discrete bundles of energy that have both wave and particle properties.

Gamma radiation is emitted by *excited* or *metastable* nuclei, i.e., those with a slight mass excess (which may, for example, result from a previous alpha or beta transition of less than maximum energy). In one gamma-decay reaction

 $^{60}$   $^{77}$  Co  $\rightarrow$   $^{69}$  Co +  $^{8}$  Y

<sup>†</sup> There is also a neutrino  ${}^{0}_{0}\nu$ , which is associated with *positron* and *electron capture* decay processes. Because neutrinos are not significant in the context of commercial nuclear energy, they are not considered here. The interested reader may consult a textbook on nuclear physics.

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an excited nucleus is transformed into one that is more "stable" (although, in this case, still radioactive). Such processes increase the binding energy but do not affect either the charge or the mass number of the nucleus.

The gamma-ray energies represent transitions between the discrete energy levels in a nucleus. As a practical matter, nuclides can often be readily identified or differentiated from each other on the basis of their distinctive gamma energies.

#### **Decay Probability**

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The precise time at which any single nucleus will decay cannot be determined. However, the average behavior of a very large sample can be predicted accurately by using statistical methods.

An average time dependence for a given nuclide is quantified in terms of a *decay* constant  $\lambda$ —the probability per unit time that a decay will occur. The activity of a sample is the average number of disintegrations per unit time. For a large sample, the activity is the product of the decay constant and the number of atoms present, or

$$Activity = \lambda n(t) \tag{2-4}$$

where n(t) is the concentration, which changes as a function of the time t. Because  $\lambda$  is a constant, the activity and the concentration are always proportional and may be used interchangeably to describe any given radionuclide population.

It has been typical to quote activities in units of the Curie {Ci}, defined as  $3.7 \times 10^{10}$  disintegrations per second, which is roughly the decay rate of 1 g of radium (the material studied by Marie Curie in her pioneering studies of radioactivity). The currently favored S1 unit is the *Becquerel*, which is 1 disintegration per second.

The rate at which a given radionuclide sample decays is, of course equal to the rate of decrease of its concentration, or

Activity = rate of decrease

Mathematically, this is equivalent to

$$\lambda n(t) = -\frac{dn(t)}{dt}$$

By rearranging terms,

 $\lambda = -\frac{dn(t)/n(t)}{dt}$ (2-5a)

or  $\frac{dn(t)}{n(t)} = -\lambda dt$ 

where Eq. 2-5a shows that the decay constant  $\lambda$  is the fractional change in nuclide concentration per unit time.

$$n(t) = n(0) e^{-\lambda_t}$$
(2-6)

where n(0) is the radionuclide concentration at time t = 0. As a consequence of the exponential decay, two useful times can be identified:

1. The mean lifetime  $\tau$ —the average [statistical mean] time a nucleus exists before undergoing radioactive decay. Because it may be shown that

$$\tau = \frac{1}{\lambda}$$

this lifetime is also the amount of time required for the sample to decrease by a factor of e (see Eq. 2-6).

2. The half-life  $T_{1/2}$ —the average amount of time required for sample size or activity to decrease to one-half of its initial amount.

The half-life, mean lifetime, and decay constant are found to be related by

$$T_{1/2} = \ln 2\tau = \frac{\ln 2}{\lambda}$$

or equivalently

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(2-5b)

$$T_{1/2} \approx 0.693\tau = \frac{0.693}{\lambda}$$

The basic features of decay of a radionuclide sample are shown by the graph in Fig. 2-2. Assuming an initial concentration n(0), the population may be noted to decrease by one-half of this value in a time of one half-life. Additional decreases occur such that whenever one half-life elapses, the concentration drops to one-half of whatever its value was at the beginning of that time interval.

An example of an important application of radioactive decay is in the management of radioactive wastes (a subject considered in detail in Chap. 19). The fission products <sup>85</sup>Kr [krypton-85] and <sup>87</sup>Kr, which have half-lives of roughly 11 years and 76 min, respectively, are generally both present in LWR cooling water. In 10 half-lives, each would be reduced in population and activity to about 0.1 percent (actually 1/1024). Thus, <sup>87</sup>Kr would essentially disappear of its own accord in a little over one-half day. The <sup>83</sup>Kr, on the other hand, would be of concern for on the order of a hundred years. (The latter, for example, was a problem following the accident at the Three Mile Island reactor, discussed in Chap. 15.)





# NUCLEAR REACTIONS

A majority of all known radionuclides are produced when nuclear particles interact with nuclei. The "man-made"<sup>†</sup> or artificial nuclides of interest in nuclear reactors span nearly all elements.

A simple reaction is depicted in Fig. 2-3. It may be represented in equation form as

$$X + x \rightarrow (C)^* \rightarrow Y + y$$

for target nucleus X, projectile particle x, compound nucleus  $(C)^*$ , product nucleus Y, and product particle y. Common shorthand notations for the reaction are

$$X(x, y)Y$$
 or simply  $X(x, y)$ 

where conservation principles allow the latter simplification. The implied designations are actually quite arbitrary since, for example, the target and projectile may both be moving (and, occasionally, are even the same nuclear species) and the "product" may consist of several nuclei and/or particles.

<sup>1</sup>This term is still commonly used (with apologies to women).



# **Compound Nucleus**

The compound nucleus temporarily contains all of the charge and mass involved in the reaction. However, it is so unstable in an energy sense that it only exists for on the order of  $10^{-14}$  s (a time so short as to be insignificant, and undetectable, on a scale of human awareness). Because of its instability, a compound nucleus should never be considered equivalent to a nuclide that may have the same number of protons and neutrons.

Nuclear reactions are subject to the same conservation principles that govern radioactive decay. Based on conservation of charge and mass number alone, a very wide range of reactions can be postulated. Total energy considerations determine which reactions are feasible. Then, angular momentum (and other) characteristics fix the relative likelihood of occurrence of each possible reaction. Equations for a number of important reactions are considered at the end of this section. Reaction probabilities are the subject of the last section of this chapter.

Conservation of total energy implies a balance, including both kinetic energy and mass. The simple reaction in Fig. 2-3 must obey the balance equation

$$E_{X} + M_{X}c^{2} + E_{x} + M_{x}c^{2} = E_{Y} + M_{Y}c^{2} + E_{y} + M_{y}c^{2}$$
(2-7)

where  $E_i$  and  $M_ic^2$ , respectively, are the kinetic and mass-equivalent energies of the *i*th participant in the reaction, Rearranging the terms of Eq. 2-7 shows that

$$[(E_{Y} + E_{y}) - (E_{X} + E_{x})] = [(M_{X} + M_{x}) - (M_{Y} + M_{y})]c^{2}$$
(2-8)

where the left-hand bracket is the Q-value for the reaction.

When Q > 0, the kinetic energy of the products is greater than that of the initial reactants. This implies that mass has been converted to energy (a fact that may be verified by examining the right-hand side of Eq. 2-8). Such a reaction is said to be *exothermal* or *exoergic* because it produces more energy than that required to initiate it.

For cases with  $Q \le 0$ , the reaction reduces the kinetic energy of the system and is said to be *endothermal* or *endoergic*. These reactions have a minimum *threshold energy*, which must be added to the system to make it feasible (i.e., to allow the mass increase required by Eq. 2-8).

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A different type of energy threshold exists in reactions between charged particles of like sign (e.g., an alpha particle and a nucleus) due to the repulsive Coulomb forces. In this case, however, the reaction may still be exoergic as long as Q > 0 or, equivalently, as long as the product kinetic energy exceeds the energy required to override the electrostatic forces. The fusion reactions considered in Chap. 21 are an important example of threshold reactions of this latter type.

# **Reaction Types**

A very wide range of nuclear reactions have been observed experimentally. Of these, the reactions of most interest to the study of nuclear reactors are the ones that involve neutrons.

When a neutron strikes  ${}^{235}_{92}U$ , for example, a compound nucleus  $({}^{236}_{92}U)^*$  is formed, as sketched in Fig. 2-4. The compound nucleus then divides in one of several possible ways. These reactions and several others are discussed in the remainder of this section. (It should be noted that charge Z and mass number A are conserved in each case.)

# Scattering

A scattering event is said to have occurred when the compound nucleus emits a single neutron. Despite the fact that the initial and final neutrons do not need to be (and likely are not) the same, the net effect of the reaction is as if the projectile neutron had merely "bounced off," or scattered from, the nucleus.

The scattering is *elastic* when the kinetic energy of the system is unchanged by the reaction. Although the equation for elastic scattering,  $^{233}U(n, n)$ , or

 $^{235}_{92}U + ^{1}_{0}n \rightarrow (^{236}_{92}U)^{*} \rightarrow ^{235}_{92}U + ^{1}_{0}n$ 

looks particularly uninteresting, the fact that the neutron generally changes both its





kinetic energy and its direction is significant. The former change, in particular, may substantially alter the probability for further reactions.

If the kinetic energy of the system decreases, the scattering is *inelastic*. The equation  $\frac{235}{2}U(n, n')$ , or

$$^{235}_{32}U + ^{1}_{0}n \rightarrow (^{236}_{92}U)^{*} \rightarrow ^{235}_{23}U + ^{1}_{0}n$$

represents the fact that kinetic energy is not conserved and that, thus, the product nucleus is left in an excited state. This extra loss of kinetic energy makes inelastic scattering significant for certain applications to neutron slowing down. The excited  ${}^{23}_{62}^{*}$ U nucleus quickly decays to its more stable form by emitting gamma radiation.

# Radiative Capture

The reaction  $^{235}_{92}U(n, \gamma)$  or

 $\label{eq:constraint} \begin{array}{c} {}^2\mathfrak{z}\mathfrak{z}\mathfrak{z}\mathfrak{U} \ + \ \mathfrak{z}\mathfrak{n} \ \rightarrow \ ({}^2\mathfrak{z}\mathfrak{z}\mathfrak{u}\mathfrak{U}\mathfrak{)}^* \ \rightarrow \ {}^2\mathfrak{z}\mathfrak{z}\mathfrak{u}\mathfrak{U} \ + \ \mathfrak{g}\gamma \end{array}$ 

is known as *radiative capture* or simply n,  $\gamma$ . The *capture gamma* in this case has an energy of about 6 MeV (corresponding roughly to the binding energy per nucleon on Fig. 2-1 for this extra neutron).

The same reaction occurring in materials other than the fuel is often called *activation*. When sodium coolant in an LMFBR gains a neutron in the reaction

$$^{23}_{11}Na + ^{1}_{0}n \rightarrow (^{24}_{11}Na)^* \rightarrow ^{24}_{11}Na + ^{0}_{0}\gamma$$

the <sup>21</sup>/<sub>1</sub>Na is radioactive (i.e., the sodium has been activated). The desire to eliminate the possibility of radioactive sodium contacting water is responsible for the introduction into the LMFBR steam cycle (Fig. 1-5) of an intermediate loop of "clean" sodium.

On the more positive side, the entire field of *activation analysis* is based on producing artificial radionuclides by neutron bombardment of an unknown sample. Then, by using gamma-ray information to determine the identity and quantity of each species, the composition of the initial material can be determined, often to a high degree of accuracy.

#### Multiple Neutron

The compound nucleus may also deexcite by emitting more than one neutron. The reaction  $^{235}_{35}U(n, 2n)$  or

$$^{2}32U + \sqrt{n} \rightarrow (^{2}32U)^{*} \rightarrow ^{2}32U + 2\sqrt{n}$$

evolves a pair of neutrons. Reactions that produce three or more neutrons are also possible.

The multiple neutron reactions are generally endoergic. For example, above a neutron threshold energy of about 6 MeV, the reaction

$${}^{2}\mathfrak{J}_{2}^{3}\mathbb{U} + {}^{1}\mathfrak{g}_{n} \rightarrow ({}^{2}\mathfrak{J}_{2}^{4}\mathbb{U})^{*} \rightarrow {}^{2}\mathfrak{J}_{2}^{3}\mathbb{U} + {}^{2}\mathfrak{g}_{n}^{n}$$

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is possible. The relatively short half-life of <sup>232</sup>U can have a significant impact on operations in <sup>233</sup>U-thorium fuel cycles (described in Chap. 6).

# Fission

The typical fission reaction  ${}^{2}\frac{35}{2}U(n, f)$  or

 ${}^{235}_{92}U + {}^{1}_{0}n \rightarrow ({}^{236}_{92}U)^{\bullet} \rightarrow F_1 + F_2 + {}^{0}_{0}\gamma's + {}^{1}_{0}n's$ 

yields two fission-fragment nuclei plus several gammas and neutrons. As considered in the next section, fission produces many different fragment pairs.

#### **Charged** Particles

Although not common for <sup>235</sup>U, there are many neutron-initiated reactions that produce light charged particles. An important example is the  ${}^{10}B(n, \alpha)$  reaction

$${}^{10}_{3}B + {}^{1}_{0}n \rightarrow ({}^{1}_{3}B)^* \rightarrow {}^{3}Li + {}^{4}_{2}\alpha$$

where boron-10 is converted to lithium-7 plus an alpha particle. On the basis of this reaction, boron is often used as a "poison" for removing neutrons when it is desired to shut down the fission chain reaction.

Other neutron-induced reactions yield protons or deuterons. Multiple chargedparticle emissions, possibly accompanied by neutron(s) and/or gamma(s), have also been observed.

#### Neutron Production

Two other types of reactions are of interest because their product is a neutron. The reaction  ${}^{2}_{1}Be(\alpha, n)$  or

$${}_{4}^{9}\text{Be} + {}_{2}^{4}\alpha \rightarrow ({}_{6}^{13}\text{C})^{*} \rightarrow {}_{6}^{12}\text{C} + {}_{0}^{1}\text{n}$$

can occur when an alpha emitter is intimately mixed with beryllium. Plutoniumberyllium [Pu-Be] and radium-beryllium [Ra-Be] sources both employ this reaction to produce neutrons. Either may be used to initiate a fission chain reaction for startup of a nuclear reactor.

High-energy gamma rays may interact with certain nuclei to product *photoneu*trons. One such reaction with deuterium,  ${}_{1}^{2}D(\gamma, n)$  or

$$^{2}D + ^{0}\gamma \rightarrow (^{2}D)^{*} \rightarrow ^{1}H + ^{1}n$$

occurs in any system employing heavy water. A similar reaction occurs with  ${}_{4}^{3}$ Be in research reactors that have beryllium components. Both reactions have energy thresholds based on overcoming the "binding energy of the last neutron" (i.e., the binding energy difference between the isotopes with A - Z neutrons and A - Z - 1 neutrons, respectively).

# NUCLEAR FISSION

The fission reaction is the basis for current commercial application of nuclear energy. Several of the important features of the process are discussed in this section.

# Nature of Fission

According to a very simple qualitative model, a nucleus may be considered as a liquid drop that reacts to the forces upon and within it. The nucleus, then, assumes a spherical shape when the forces are in equilibrium. When energy is added, the nucleus is caused to oscillate from its initially spherical shape. If the shape becomes sufficiently elongated, it may neck down in the middle and then split into two or more fragments. A large amount of energy is released in the form of radiations and fragment kinetic energy (e.g., see Fig. 1-1).

Almost any nucleus can be fissioned if a sufficient amount of excitation energy is available. In the elements with Z < 90, however, the requirements tend to be prohibitively large. Fission is most readily achieved in the heavy nuclei where the threshold energies are 4-6 MeV or lower for a number of important nuclides.

Certain heavy nuclides exhibit the property of *spontaneous fission* wherein an external energy addition is not required. In californium-252  $\binom{252}{98}Cf$ , for example, this process occurs as a form of radioactive decay with a half-life of about 2.6 years. Even <sup>235</sup>U and <sup>239</sup>U fission spontaneously, but with half-lives of roughly 10<sup>17</sup> years and 10<sup>16</sup> years, respectively; these values are at least 10<sup>7</sup> times greater than the  $\propto$ -decay half-lives.

Charged particles, gamma rays, and neutrons are all capable of inducing fission. The first two are of essentially no significance in the present context. As indicated in the previous chapter, neutron-induced fission chain reactions are the basis for commercial nuclear power.

# Neutron-Induced Fission

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When a neutron enters a nucleus, its mere presence is equivalent to an addition of energy because of the binding energy considerations discussed earlier. The binding energy change may or may not be sufficient by itself to cause fission.

A fissile nuclide is one for which fission is possible with neutrons of any energy. Especially significant is the ability of these nuclides to be fissioned by thermal neutrons, which bring essentially no kinetic energy to the reaction. The important fissile nuclides are the uranium isotopes  ${}^{235}_{92}U$  and  ${}^{233}_{92}U$  and the plutonium isotopes  ${}^{239}_{92}Pu$  and  ${}^{241}_{92}Pu$ . It has been noted that  ${}^{235}U$  is the only naturally occurring member of the group.

A nuclide is *fissionable* if it can be fissioned by neutrons. All fissile nuclides, of course, must fall in this category. However, nuclides that can be fissioned only by high-energy, "above-threshold" neutrons are also included. This latter category includes  $^{212}_{92}$ Th,  $^{218}_{92}$ U, and  $^{240}_{94}$ Pu, all of which require neutron energies in excess of 1 MeV.

The fissile nuclides that do not exist in nature can only be produced by nuclear reactions. The target nuclei for such reactions are said to be *fertile*. Figure 2-5 traces the mechanisms by which the three major fertile nuclides,  ${}^{2}_{90}$ Th,  ${}^{2}_{90}$ U, and  ${}^{2}_{94}$ Pu, produce  ${}^{2}_{92}$ U,  ${}^{2}_{94}$ Pu, and  ${}^{2}_{94}$ Pu, respectively. The first two are each based on radiative

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Chains for conversion of fertile nuclides to fissile nuclides: (a)  $^{11}$  Th to  $^{11}$  (b)  $^{11}$  (b)  $^{11}$  (c)  $^{11}$  Pu; (c)  $^{11}$  Pu to  $^{11}$  Pu.

capture followed by two successive beta decays. The last process is much simpler in being complete following the capture reaction.

It may be noted that the fertile nuclides are also fissionable (by fast neutrons). The neutrons below the threshold energy cannot cause fission, but can produce more fissile material. However, from the standpoint of neutron economy, it must be recognized that the latter process requires the equivalent of two neutrons for each fission.

As introduced in the previous chapter, a reactor will be classified as a converter or a breeder based on its production of new fissile from fertile nuclides. These concepts are considered in more detail in Chap. 6.

When a nucleus fissions, the major products are fission fragments, gamma rays, and neutrons. The fission fragments then undergo radioactive decay to yield substantial numbers of beta particles and gamma rays, plus a small quantity of neutrons.

#### Fission Fragments

A fissioning nucleus usually splits into two fragments. Because of some complex effects related to nuclear stability, the split does not usually produce equal masses. The asymmetric distribution, or characteristic "double-humped" distribution, for thermal-neutron fission of <sup>235</sup>U is shown in Fig. 2-6. It may be noted that equal-mass fragments ( $A \approx 117$ ) are produced by only about 0.01 percent of the fissions. Fragment nuclides in the ranges of roughly 90-100 and 135-145 occur in as many as 7 percent of the fissions.

In about 1 case out of 400, 3 fragments are produced by a *ternary fission*. One such fragment may be tritium [{T], the beta-active isotope of hydrogen. As discussed in Chap. 19, the presence of tritium can be a significant problem in waste management.



#### FIGURE 2-6

Fission yield as a function of mass number for thermal-neutron fission of <sup>335</sup>U. (Adapted from Nuclear Chemical Engineering by M. Benedict and T. H. Pigford, © 1957 by McGraw-Hill Book Company, Inc. Used by permission of McGraw-Hill Book Company.)

The fission fragments tend to be neutron-rich with respect to stable nuclides of the same mass number. The related energy imbalance is generally rectified by successive beta emissions, each of which converts a neutron to a proton. Two beta-decay "chains" (from *different* fissions) are shown in Fig. 2-7. The gamma rays are emitted whenever a beta decay leaves the nucleus in an excited state. The antineutrinos that accompany the beta decays are not shown because they have no direct effect on nuclear energy systems.

The chain in Fig. 2-7, which contains strontium-90 [ ${}_{38}^{90}$ Sr], is especially troublesome both in reactor accidents (Chaps. 13–15) and waste management (Chap. 19) because strontium is relatively volatile and this isotope has a long 29-year half-life coupled to the high fission yield shown in Fig. 2-6. Similar considerations apply to cesium-137 [ ${}_{33}^{133}$ Cs], which has a 30-year half-life.

Another problem associated with the fission fragments is the presence within the decay chains of nuclides, which capture neutrons that would otherwise be available to sustain the chain reaction or to convert fertile material. Two especially important neutron "poisons" are xenon-135 [ $^{135}_{54}$ Xe] and samarium-149 [ $^{169}_{62}$ Sm]. Each poses a slightly different problem for reactor operation (as discussed in Chap. 6).

#### Neutron Production

Thermal-neutron fission of <sup>235</sup>U produces (an average of) 2.5 neutrons per reaction. The majority of these are *prompt neutrons* emitted at the time of fission. A small fraction are *delayed neutrons*, which appear from seconds to minutes later.

The number of neutrons from fission depends on both the identity of the fis-





Two representative fission-product decay chains (from *different* fissions), (Adapted courtesy of U.S. Department of Energy.)

sionable nuclide and the energy of the incident neutron. The parameter  $\nu$  (referred to simply as "nu") is the average number of neutrons emitted per fission.

The energy distribution, or spectrum, for neutrons emitted by fission  $\chi(E)$  is relatively independent of the energy of the neutron causing the fission. For many purposes, the expression

$$\chi(E) = 0.453 e^{-1.036E} \sinh \sqrt{2.29E}$$
(2-9)

provides an adequate approximation to the neutron spectrum for <sup>235</sup>U shown by Fig. 2-8. The most likely neutron energy of about 0.7 MeV occurs where  $\chi(E)$  is a maximum.

The neutron spectrum  $\chi(E)$  is actually defined as a probability density, or the probability per unit energy that a neutron will be emitted within increment dE about energy E. Thus, neutron fractions can only be obtained by integration over finite energy intervals. As a probability density, it is required that

$$\int_0^\infty dE \chi(E) = 1$$

(where for actual data the infinite upper limit would be replaced by the maximum observed energy). The average or mean energy  $\langle E \rangle$  of the distribution is then

$$\langle E \rangle = \int_0^{\infty} dE \ E \ \chi(E)$$

which for Eq. 2-9 evaluates to very nearly 2.0 MeV.







Almost all fission neutrons have energies between 0.1 MeV and 10 MeV. Thus, reactor concepts that are based on using the very-low-energy thermal neutrons (E < 1 eV) must moderate or slow down the fission neutrons by a factor of one million or more. (Chapter 4 considers this situation in more detail.)

Delayed neutrons are emitted by certain fission fragments immediately following the first beta decay. They enter the system on time scales characterized by the halflives of the decay processes. Although delayed neutrons constitute a small fraction (less than 1 percent) of the neutron production attributed to the fission process, they play a dominant role in reactor control (as described in Chap. 5).

# Energy Production

A typical fission produces nearly 200 MeV of energy. By comparison, 2-3 eV of energy is released for each carbon atom that is burned with oxygen. The per-reaction difference by a factor of 70-100 million is a major advantage for nuclear fission.

A representative distribution of energy among the various products of the fission process is provided in Table 2-2. Essentially, all of the kinetic energy from the fragments, neutrons, and gammas is deposited within the reactor vessel in the form of heat. Of the energy associated with fragment beta decays, however, only the fraction carried by the beta particles produces heat; the remainder leaves the system with the antineutrinos.

Radiative capture reactions occur in all reactor systems. Thus, while they are not fissions, they do absorb fission neutrons and add gamma energy. Their actual contribution depends on the composition of the reactor in terms of fissile, fertile, and other species.

The energy from the prompt radiations is deposited at the time of fission. That from the delayed radiations appears in a time-dependent manner characterized by the half-lives of the various fission-product species. In both cases, the energy appears as heat.

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#### TABLE 2-2 Representative Distribution of Fission Energy

	Fission	Heat produced		
linergy source	(MeV)	MeV	% of total	
Fission fragments	168	168	84	
Neutrons	5	Ś	2 4	
Prompt gamma rays	7	7	1.5	
Delayed radiations		,	2.5	
Beta particles <sup>†</sup>	20	*	4	
Gamma rays	7	7	14	
Rudiative capture gammas <sup>‡</sup>	_	5	2.5	
foral	207	200	100	

<sup>1</sup>Includes energy carried by beta particles and antineutrinos; the latter do not produce heat in reactor systems.

<sup>4</sup> Nonfission capture reactions contribute heat energy in all real systems; design-specific considerations may change this number by about a factor of two in either direction.

The rate of energy generation—i.e., the power—from fission product decay can be described by the equation

$$\frac{P(t)}{P_0} = 6.6 \times 10^{-2} \left[ t^{-0.2} - (t + t_0)^{-0.2} \right]$$
(2.10)

for decay power P(t) at t seconds after reactor shutdown and where  $t_0$  is the time (in seconds) that the reactor had operated at a steady thermal power level of  $P_0$ . The heat generation rate falls off only as the one-fifth power of time, resulting in a sizeable heat load at relatively long times after shutdown. This decay-heat source has important impacts on spent fuel handling, reprocessing, waste management, and reactor safety (all of which are considered in later chapters).

With 200 MeV available from each fission, heat is produced such that

 $3.1 \times 10^{10}$  fission/s = 1 W

A gram of any fissile nuclide contains about 2.5  $\times$  10<sup>21</sup> nuclei. Thus, the energy production per unit mass may be expressed roughly as

Fission energy production = 1 MW d/g

ባለል ከዚሉ ማስታ የአስት አስት የመጠቀት በሚያለበዋል። የማስፈለም በምሳሌዎቹ የሚያለበዋል የሚያለዋ የአስት ማስቀ በአስት ማስቀም የአስት የሚያለዋ የአስት የአስት የአስት የ

In real systems where radiative capture reactions convert some nuclei before they can tission, somewhat larger amounts of fissile material are expended for a given energy production.

# **REACTION RATES**

The power output of a nuclear reactor is directly proportional to the rate at which fissions occur. However, the rates for all reactions that produce or remove neutrons

determine the overall efficiency with which fissile and fertile materials are employed. The ability to calculate such reaction rates is extremely important to the design and operation of nuclear systems.

When a single compound nucleus has more than one mode for deexcitation, it is not possible (with current knowledge) to predict with absolute certainty which reaction will occur. (This characteristic is analogous to the uncertainty in time of emission for radioactive decay.) The relative probability for each outcome, however, can be determined.

Because of the extreme complexity of the interactions that occur within nuclei, theoretical considerations alone are seldom adequate for predicting nuclear reaction rates. Instead, experimental measurements provide the bulk of available information.

Reaction rates are generally quantified in terms of two parameters—a macroscopic cross section related to the characteristics of the material in bulk and a flux characterizing the neutron population. The form is based, at least in part, on the historical development of the fields of nuclear physics and reactor theory.

# **Nuclear Cross Sections**

The concept of a nuclear cross section  $\sigma$  was first introduced with the idea that the effective size of a nucleus should be proportional to the probability that an incident particle would react with it. The following steps are representative of the conceptual development:

- 1. Each spherical nucleus is pictured as representing a cross-sectional, or target, area to a parallel beam of neutrons traveling through the reference medium.
- 2. A "small" cylinder of area dA and thickness dx is constructed of a single-nuclide material with an *atom density* of n per cm<sup>3</sup> (as shown in Fig. 2-9).
- 3. A "point" neutron traveling perpendicular to the face of the cylinder sees each nucleus in the sample (i.e., the sample is "thin" enough that the nuclei do not "shadow" each other).
- 4. A neutron entering the disk at a random location on surface dA has a probability for a "hit," which is equal to the total area presented by the targets divided by the area of the disk.
- 5. If each nucleus is assumed to have a cross-sectional area  $\sigma$ , the total area of the targets is the number of nuclei in the disk (i.e., atom density times volume, n dV = n dA dx) multiplied by the area per nucleus.

Total target area =  $\sigma n dA dx$ 



# FIGURE 2-9

Basic geometry for developing the concept of a nuclear cross section: thickness = dx(cm); area = dA (cm<sup>3</sup>); atom density = n(at/cm<sup>3</sup>).

P.M. C. A. CLARKER, DRIVE STREET, SPECIFICATION, CONTRACT, MARKED AND ADDRESS A ADDRESS AND ADDRESS 6. Thus, the probability that a neutron will interact in traveling a distance dr through the material is

Interaction probability = 
$$\frac{\text{total target area}}{\text{disk surface area}} = \frac{\sigma n \, dA \, dx}{dA}$$
  
Interaction probability =  $n \, \sigma \, dx$  (2-11)

According to the original definition in Eq. 2-11, the interaction probability is the product of the atom density n, the cross-sectional area  $\sigma$  of the nucleus, and the distance of travel dx (the latter only for "short" distances where nuclei do not shadow each other). Measurements of interaction probabilities in materials of known density and thickness were employed to determine values for  $\sigma$  for various nuclei.

Later development showed, however, that there are many practical cases where the apparent area changes dramatically with neutron energy. These seemingly unphysical phenomena were eventually traced to the complex interactions of the nuclear forces and particles. Nuclei can have an especially great affinity for those neutrons that are capable of exciting their discrete energy levels.

This situation suggested that the connotation of cross-sectional area be dropped. Thus,  $\sigma$  is merely a *cross section* defined such that its product with the atom density n and distance dx is equal to the interaction probability. (It should be noted that although Eq. 2-11 is still valid, the emphasis has been shifted from defining the probability to defining the cross section.) In the present context, Eq. 2-11 may be rearranged to

$$\sigma = \frac{\text{interaction probability}}{n \, dx} \tag{2-12}$$

which defines the cross section  $\sigma$  as the interaction probability per unit atom density per unit distance of neutron travel.

For reasons that become more apparent later,  $\sigma$  is more formally called the *microscopic cross section* (where the added word signifies application to describing the behavior on a nucleus-by-nucleus, or actually *submicroscopic basis*). Every nuclide can be assigned a cross section for each possible type of reaction and each incident neutron energy. The form  $\sigma'_{\ell}(E)$  may be employed to emphasize dependence on the nuclide *j*, reaction type *r*, and neutron energy *E*.

Many typical cross sections have been found to have values on the order of  $10^{-24}$  cm<sup>2</sup>. The tongue-in-check comment that this area is "as big as a barn door" led to defining the unit

 $1 \text{ barn } [b] = 10^{-24} \text{ cm}^2$ 

A long-standing compilation of cross-section data is contained in the various editions and parts of BNL-325 (Hughes/BNL-325, 1955), collectively referred to as the "barn book" (having a sketch of a barn on the covers). BNL-325 and the more recent computer-based Evaluated Nuclear Data File [ENDF] (Honeck/ENDF, 1966) contain substantial cross-section data classified by nuclide, interaction type, and energy. (2.13)

# Interaction Types

One possible classification system for neutron cross sections is shown in Fig. 2-10. The total cross section  $\sigma_i$  represents the probability that any reaction will occur for the given nuclide and neutron energy. It consists of scattering ( $\sigma_s$ ) and absorption ( $\sigma_o$ ) components. The scattering, in turn, is split between the elastic ( $\sigma_c$ ) and inelastic ( $\sigma_i$ ) processes described earlier.

The absorption cross section  $\sigma_u$  includes contributions from all reactions except scattering. Each absorption, then, produces one or more new nuclei. Fission is treated as an absorption event based largely on useage in the various calculational methods. The neutrons produced by fission are treated separately from the initial absorption. (Chapter 4 discusses the role of the cross sections in reactor calculations.)

The nonfission absorption processes are referred to as *capture* [nonfission-capture] events. Radiative capture  $[(n, \gamma)]$ , charged-particle  $[(n, p), (n, d), (n, \alpha), (n, 2\alpha), \ldots$ ], multiple-neutron  $[(n, 2n), (n, 3n), \ldots]$ , and charged-particle/neutron  $[(n, pn), (n, dn), \ldots]$  processes are the important constituents.

The detailed interaction spectrum—i.e., the far-right portion of Fig. 2-10—is generally used only to construct a "condensed" *library* of cross sections. The library is then employed to generate the total, scattering, absorption, and fission cross sections used in most reactor calculations. (The basic models and their use of cross sections are described in Chap. 4.)

Fissile nuclei cease to exist whenever they experience an absorption reaction. In fission the nucleus is split, while in capture some new species is produced. The *capture-to-fission ratio*  $\alpha$  is defined as



FIGURE 2-10 The hierarchy of microscopic cross sections used to calculate neutron interaction rates.

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It has been found to provide a convenient measure of the probabilities of the two processes with respect to each other. By itself, a small value of  $\alpha$  would be favored since it implies more fission (and, thus, more energy production) per unit mass of fissionable material.

The prospects for converting and breeding in fertile material depend on the number of neutrons produced per fission. However, a more important measure is the number of neutrons produced per nucleus destroyed  $\eta$  (referred to simply as *eta*),<sup>†</sup> defined by

 $\eta = \nu \frac{\sigma_f}{\sigma_a}$ 

where  $\nu$  is the average number of neutrons per fission and the ratio of cross sections is the fraction of absorptions that produce fission. With one fission neutron always required to sustain the chain reaction, the remainder may be available for fertile conversion. The possibility for *breeding* (i.e., producing an amount of new fissile material that is greater than that expended) exists whenever  $\eta$  exceeds two.

Both the capture-to-fission ratio and eta vary with nuclide and neutron energy since they are based on cross sections with similar dependencies. The forms  $\alpha^{j}(E)$  and  $\eta^{j}(E)$  for nuclide j and energy E are appropriate for more detailed representation.

Cross sections, capture-to-fission ratios, and neutron production factors for the important fissile and fertile nuclides are compared in Table 2-3. All parameters have been evaluated at an energy of 0.025 eV (a standard reference that corresponds to the most probable neutron speed of a population in thermal equilibrium at room temperature).

Some observations based on the data in Table 2-3 (the significance of which will be described later) are:

• <sup>233</sup>U has the smallest fission cross section and the second lowest  $\nu$  yet has the largest  $\eta$  and, thus, the best prospect for breeding.

<sup>2</sup> η for a mixture is defined in Chap. 4 for the "four-factor formula."

#### TABLE 2-3

Thermal-Neutron Cross Sections and Parameters for Important Fissile and Fertile Nuclides

Cross section (b) <sup>†</sup>					
σ.	σ,	σι	a	¥۲	η
7.4	7.4	_	_		
577	46	531	0.087	2.50	2.30
584	99	585	0.169	2.44	2.09
2.68	2.68	_	-	_	_
1021	271	750	0.361	2.90	2.13
290	290	0.05	_	_	_
1371	361	1010	0.357	3.00	2.21
	7.4 577 584 2.68 1021 290 1371	$\begin{tabular}{ c c c c c c } \hline Cross section (b) \\ \hline $\sigma_{a}$ & $\sigma_{r}$ \\ \hline $7.4$ & $7.4$ \\ $577$ & $46$ \\ $584$ & $99$ \\ $2.68$ & $2.68$ \\ $1021$ & $271$ \\ $250$ & $290$ \\ $1371$ & $361$ \\ \hline \end{tabular}$	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	$\begin{array}{c c c c c c c c c c c c c c c c c c c $

<sup>†</sup>Cross sections from Chart of the Nuclides (GE/Chart, 1989) for 0.025 eV "thermal" neutrons;  $\sigma_i$  has been assumed to be equal to the radiative-capture cross section  $\sigma_{y}$ .

<sup>4</sup> μ values at 0.025 eV from Hughes/BNL-325 (1958)

- Although the fissile plutonium isotopes each produce about three neutrons per fission, their large capture-to-fission ratios result in fairly low values of η.
- The fertile nuclides <sup>232</sup>Th and <sup>238</sup>U have absorption cross sections that are on the order of only 1 percent of those of their respective conversion products <sup>233</sup>U and <sup>239</sup>Pu.
- Fertile <sup>240</sup>Pu has a large capture cross section for production of fissile <sup>241</sup>Pu.

It must be emphasized that the comments above apply strictly to 0.025 eV neutrons and may at most extend to thermal neutrons. In fact, as shown later in Chap. 6, several completely opposite conclusions must be drawn when fission-neutron energies are considered.

The fissionable nuclides may be identified by a shorthand notation (which actually dates back to the Manhattan Project and the need for code names for the weaponsuseable materials). For example,

 $^{2}y_{2}^{3}U \rightarrow 25$ 

based on the last digits of the atomic number and the atomic mass number, respectively. The plutonium isotopes  $^{239}_{94}$ Pu,  $^{240}_{94}$ Pu, and  $^{241}_{94}$ Pu, which become 49, 40, and 41, respectively, may be noted to have identifiers that are not consecutive. Overall, the format can be quite useful, especially as a compact superscript for cross sections or other parameters (e.g., as nuclide identification j in  $\sigma_j^{j}$  or  $\eta^{j}$ ).

# Energy Dependence

The change in cross sections with incident neutron energy can be fairly simple or extremely complex, depending on the interaction and the nuclide considered. Only some of the more important general features are considered here.

The total cross section for <sup>238</sup>U plotted as a function of energy is shown in Fig. 2-11. Significant features (on the doubly logarithmic scale) are the "linear" region at low energies and the region of high, narrow peaks at intermediate energies.

The sloped, linear portion of the cross section curve in Fig. 2-11 is characteristic of a process referred to as l/v-absorption (or "one-over-v" absorption) for neutron speed v. Among neutrons passing close to a given nucleus, the slower ones spend



#### FIGURE 2-11



more time near the nucleus and experience the nuclear forces for a longer time. The absorption probability, then, tends to vary inversely as the neutron speed and, thus, give rise to the 1/v-behavior. When an absorption cross section  $\sigma_0$  is known for energy  $E_0$ , values for other energies in the 1/v-region may be readily calculated from

$$\sigma_{\nu}(E) = \sigma_{0} \frac{v_{0}}{v}$$
or 
$$\sigma_{\mu}(E) = \sigma_{0} \frac{\sqrt{E_{0}}}{\sqrt{E}}$$
(2-14)

because the kinetic energy in terms of speed is just  $E = \frac{1}{4}mv^2$ . The "thermal" energy E = 0.025 eV (or v = 2200 m/s) is generally used as the reference whenever it lies within the range.

Almost all fissionable and other nuclides have regions that are roughly 1/v, many to higher energies than shown for <sup>238</sup>U. In the important neutron poison <sup>10</sup>B, this regular behavior actually continues rather precisely up to energies in the keV range.

• The very high, narrow resonance peaks in the center region of Fig. 2-11 are a result of the nucleus's affinity for neutrons whose energies closely match its discrete, quantum energy levels. It may be noted for the lowest-energy resonance that the total cross section changes by a factor of nearly 1000 from the peak energy to those just slightly lower. All fissionable materials exhibit similar resonance behavior.

Scattering, the other contributor to the total cross section, tends to take two forms. In *potential scattering*, the cross section is essentially constant (i.e., independent of energy) at a value somewhat near the effective cross-sectional area of the nucleus. *Resonance scattering*, like its absorption counterpart, is based on the energy-level structure of the nucleus.

The distinction between fissile and nonfissile isotopes is readily observed from the plot of the fission cross sections for <sup>235</sup>U and <sup>238</sup>U in Fig. 2-12. As neutron energy



FIGURE 2-12

Microscopic fission cross section for fissile <sup>335</sup>U and fissionable <sup>238</sup>U. (Data from Hughes/BNL-325, 1955.)

increases, the <sup>235</sup>U curve is characterized by large (near-1/ $\nu$ ) values, resonance behavior, and slightly irregular low values. The nonfissile <sup>238</sup>U shows a fission threshold near 1 MeV followed by a maximum cross section of one barn. The fission cross section in <sup>235</sup>U clearly exceeds that for <sup>238</sup>U at all neutron energies of interest in reactor systems.

Figure 2-12 also shows that for <sup>235</sup>U thermal-neutron fission is more probable than fast-neutron ( $E \ge 0.1$  MeV) fission by over three orders of magnitude. This is one of the important factors that has favored thermal reactors over fast reactors.

#### Interaction Rates

The cross section quantifies the relative probability that a nucleus will experience a neutron reaction. The overall rate of reaction in a system, however, also depends on the characteristics of the material and of the neutron population. The following simplified derivation identifies the important features of interaction-rate calculations.

Considering first a parallel beam of monoenergetic neutrons with a speed v and a density of N per unit volume, it is necessary to determine the rate at which they pass through a sample like that of Fig. 2-9. If, as before, the disk has an area dA and a thickness dx, then:

- 1. When the beam is perpendicular to dA, only neutrons directly in front of and within a certain distance l can reach the surface in a given time dt.
- 2. The distance l traveled by the neutrons is equal to their speed v multiplied by the time dt, or l = v dt.
- 3. As shown by Fig. 2-13, only the neutrons within the cylinder of length l and area dA can enter the sample in time dt (all others will either not reach the target or will pass outside of its boundaries).
- 4. The number of neutrons in the cylinder is the density N times the volume dV = l dA, or v dt dA from (2)],

Number of neutrons passing 
$$= \frac{N dV}{dt} = \frac{N v dt dA}{dt} = Nv dA$$
 (2-15)

The reaction rate must be equal to the product of the rate of entry of neutrons and the probability that a neutron will interact with a nucleus. Thus, combining the results of Eqs. 2-15 and 2-11,

Reaction rate = 
$$(Nv \, dA)(n\sigma \, dx)$$





Basic geometry for developing the concept of nuclear reaction rate in terms of macroscopic cross section and neutron flux: I = v dt; V = I dA = v dt dA.

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for neutron density N, neutron speed v, nuclide density n, cross section  $\sigma$ , and disk area and thickness dA and dx, respectively. Rearranging terms and noting that dA dx is just the disk volume dV,

Reaction rate =  $(n\sigma)(N\nu) dV$  (2-16)

This result separates the effects of nuclide characteristics, neutron population, and sample volume.

The macroscopic cross section  $\Sigma$  is defined as

$$\Sigma = n\sigma \tag{2.17}$$

With *n* the number of atoms per unit volume and  $\sigma$  the area per atom, the macroscopic cross section is "per unit distance" or generally cm<sup>-1</sup>. It is the probability per unit distance of travel that a neutron will interact in a sample characterized by atom density *n* and microscopic cross section  $\sigma$ .

It is, perhaps, unfortunate that both  $\sigma$  and  $\Sigma$  bear the title "cross section" because they have different units. The microscopic cross section is an "effective area" used to characterize a single nucleus. The macroscopic cross section is the probability that a neutron will interact in traveling a unit distance through a (macroscopic) sample of material. Use of the common nicknames "micro" and "macro" without attaching the words "cross section" may provide a partial solution to the problem.

The neutron flux  $\Phi$  is defined by

$$\Phi = N\nu \tag{2-18}$$

for neutron density N and speed v. The density N is in terms of neutrons per unit volume, and the speed v is distance per unit time, thus, neutron flux  $\Phi$  is neutrons per unit area per unit time, or generally neutrons/cm<sup>2</sup>·s. Although the earlier derivation was based on a monodirectional, monoenergetic beam, the definition itself is completely general.

The reaction rate in Eq. 2-16 may be rewritten as

Reaction rate =  $\Sigma \Phi dV$  (2-(9)

for macroscopic cross section  $\Sigma$  and neutron flux  $\Phi$  as defined by Eqs. 2-17 and 2-18, respectively, and for sample volume dV. An alternative form is

Reaction rate per unit volume =  $\Sigma \Phi$  (2-20)

where now the volume dV is unspecified.

The macroscopic cross section  $\Sigma$  is constructed from microscopic cross sections, and thus varies with nuclide, interaction type, and neutron energy. In the form  $n^{j}\sigma_{i}^{j}(E) = \Sigma_{i}^{j}(E)$ , it represents the probability per unit distance of travel that a neutron of energy E will interact by mechanism r with nuclide j. These interaction probabilities for a given reaction and given nucleus are independent of other reactions and the other nuclei present, so they may be summed to describe any desired combination. The total macroscopic cross section for any given mixture is

$$\sum_{i}^{\min} = \sum_{\text{all } j} \sum_{\text{all } j} \sum_{r} \sum_{r}^{j} = \sum_{\text{all } j} \sum_{\text{all } r} n^{j} \sigma_{r}^{j}$$
(2-21)

where the summations are over all nuclides j in the mixture and all reactions r. In summing the reactions, care is advised in assuring that each is independent (e.g.,  $\Sigma_a$  may not be added to  $\Sigma_f$  or  $\Sigma_c$  because  $\Sigma_a = \Sigma_f + \Sigma_c$ , as shown by Fig. 2-10).

Based on the inherent limitations of the earlier development of the concept of a microscopic cross section, it must be recognized that Eq. 2-11 and others based on it are valid only for "small" samples where the nuclei do not shadow or obscure each other from the neutron beam. "Thick" samples have the effect of removing neutrons and, therefore, changing the neutron flux seen by the more internal nuclei. If a beam of neutrons passes into a material sample, it is said to experience attenuation or a decrease of intensity.

If a beam of parallel monoenergetic neutrons crosses the surface of a sample, one neutron will be removed each time a reaction occurs. Thus, the rate of *decrease* of neutron flux  $\Phi(x)$  with distance x will be equal to the reaction rate,

$$-\frac{d\Phi(x)}{dx} = \Sigma_t \Phi(x)$$

where use of the macroscopic total cross section implies that all reactions remove neutrons from the beam. (Note that even scattering does this by changing the energy and direction of the neutron.) Rearrangement of terms yields

$$\Sigma_{t} = -\frac{d\Phi(x)/\Phi(x)}{dx}$$
(2-22a)

or 
$$\frac{d\Phi(x)}{\Phi(x)} = -\Sigma_t dx$$
 (2-22b)

where Eq. 2-22a shows that  $\Sigma_i$  is the fractional loss of flux (or neutrons, since  $\Phi = Nv$ ) per unit distance of travel. Viewed another way, it restates the definition of  $\Sigma_i$  as the total interaction probability per unit distance of travel.

The solution to Eq. 2-22b is

$$\Phi(x) = \Phi(0)e^{-\Sigma_{x}x}$$
(2-23)

for entering flux  $\Phi(0)$  at position x = 0. This exponential decay may be noted to parallel the decay of radionuclides in Eq. 2-6. Figure 2-14 depicts the effects of attenuation (and emphasizes again that  $\Sigma$  serves the role of an attenuation coefficient rather than a true cross section).

The neutron mean free path  $\lambda$ , or the average distance of travel before an interaction, may be shown to be

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Usponential attenuation of a neutron beam in a material with total macroscopic cross section  $\Sigma_i$ .

For any given nuclide j and reaction r (or combination thereof), the mean free path  $\lambda_i^r = 1/\Sigma_i^j$  is the average distance of neutron travel between reactions of the type considered.

Because of the exponential nature of the attenuation described by Eq. 2-23, the neutron flux never becomes identically zero, not even at arbitrarily large distances. There is, then, always some probability for very deep penetration.

# Nuclear Data

Four of the more important sources of nuclear data are:

- 1. Chart of the Nuclides [GE/Chart, 1989]
- 2. Table of Isotopes [Lederer & Shirley, 1978]
- 3. Neutron Cross Sections [Hughes/BNL-325, 1955]
- 4. Evaluated Nuclear Data File (ENDF) [Honeck/ENDF, 1966]

The first of these is a *must* for anyone doing work related to nuclear energy. The others, which are of somewhat more specialized interest, are described first.

The Table of Isotopes is essentially a compilation of experimental data on halflives, radiations, and other parameters for almost all radionuclides. It contains detailed energy-level diagrams and identifies the energy and emission probability for each radiation. The data itself plus the extensive referencing make the Table of Isotopes virtually indispensable to anyone doing activation-analysis or other radionuclide measurements.

As mentioned previously, BNL-325 and ENDF contain cross-section data. BNL-325, the "barn book," presents data in tabular and graphical form, which is of most use for overview purposes dealing with a limited range of nuclides, reactions, and/or energies.

The computer-based ENDF is the workhorse for nearly all large-scale calculations. The ENDF/B set contains complete, evaluated sets of nuclear data for about 80 nuclides and for all significant reactions over the full energy range of interest. Revised versions are developed, tested, and issued periodically.

# Chart of the Nuclides

The Chart of the Nuclides is very useful for performing preliminary or scoping calculations related to reactors. A portion containing important fissile and fertile nuclides is sketched in Fig. 2-15. Because of the vast amount of information, the figure shows the basic structure plus detail for only the <sup>235</sup>U, <sup>238</sup>U, and <sup>239</sup>Pu nuclides. The basic grid has the elements in horizontal rows with vertical columns representing neutron numbers. The block at the left end of each row gives the chemical symbol and, as appropriate, the thermal [0.025 eV] absorption and fission cross sections for the naturally occurring isotopic composition.

Basic data for individual nuclides includes (as appropriate):

- nuclide ID
- natural isotopic composition in atom percent [at %]
- half-life

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- type of radiation(s) emitted and energies in decreasing order ("..." implies additional lower energy values not included)
- .. thermal neutron cross sections and resonance integrals, both in barns
- isotopic (atomic) mass

Because the isotopic compositions are given in atom percent [at %], while fuel cycle mass flows are usually in weight percent [wt %], there is occasional confusion (e.g., the enrichment of natural uranium is 0.72 at % or 0.711 wt % <sup>235</sup>U). The resonance integral mentioned above is intended as a measure of an "effective" cross section that fission neutrons would see in slowing down through the resonance-energy region (see also Chap. 4).

The masses used in the Chart of the Nuclides are for the entire atom rather than for the nucleus. Thus, they include the mass of the electrons less the contribution of electron binding energy. The electron binding energies are almost always too small to be significant in comparison with nuclear binding energies. Further, because nuclear mass differences (as opposed to the masses themselves) are of most interest, atomic masses can be used consistently. (It may be noted that charge conservation guarantees that electrons are handled appropriately. In beta decay, for example, the new electron can enter an orbit to balance the extra proton in the nucleus.)

The Chart of the Nuclides also contains a wealth of useful information not shown in Fig. 2-15. One interesting feature is the use of color codes for both half-life and cross-section ranges. A recent addition has been fission-product production data for typical power reactors. Fortunately, the introduction and guide to use of the Chart of the Nuclides are well written and easily understood. Currently, it is available in both wall-chart and booklet form.

 $<sup>\</sup>lambda = \frac{1}{\Sigma}$ 



# Summary

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Most of the important nuclides and many of the concepts of this chapter are directly or indirectly summarized in Fig. 2-16. Isotopes are in rows with arrows representing radiative capture or  $(n, \gamma)$  reactions. Beta decays are shown by downward-pointing arrows. Thermal-neutron fission is represented by diagonal dashed arrows. Decay halflives and thermal cross sections are from the Chart of the Nuclides.

As a review, the reader should identify on Fig. 2-16:

- the four major fissile nuclides
- three chains for converting fertile nuclides to fissile nuclides
- an important (n, 2n) reaction
- nonfission capture events in fissile nuclides

Overall, the information on this figure should be of substantial value in a number of the later chapters.



FIGURE 2-16 Neutron irradiation chains for heavy elements of interest for nuclear reactors.

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# Sample Calculations

The data in Figs. 2-15 and 2-16 plus the equations developed in this chapter can be used to determine a number of important reactor and fuel cycle characteristics. The following are representative examples:

1. The reaction and decay equations and total mass change for conversion of fertile  $^{238}_{234}$ U to fissile  $^{239}_{234}$ Pu:

$$\begin{cases} 2392 U + \frac{1}{9}n \rightarrow (239 U)^* \rightarrow 239 U + \frac{0}{9}\gamma \\ 2392 U \rightarrow 239 Np + -\frac{0}{1}\beta \\ 2393 Np \rightarrow 239 Pu + -\frac{0}{1}\beta \end{cases}$$
(Fig. 2-5)

Noting that charge and mass number are conserved,

$$\begin{array}{cccc}
 & 238 \\
 & 92 \\
 & 92 \\
 & 92 \\
 & 10 \\
 & 10 \\
 & 10 \\
 & 1008665 \\
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In terms of kinetic energy,

0.007293 amu × 931.5 MeV/amu = 6.79 MeV

2. The nuclide density  $n^{28}$  of  $^{238}$ U. The definition of SI units (see App. 11) provides that one mole [mol] of any element contains the same number of atoms as 0.012 kg [12 g] of  $^{12}$ C. In the same manner that a mole of  $^{12}$ C has a mass equivalent to its mass number (A = 12.000, as used to define the atomic mass unit [amu]) in grams, a mole of each other element consists of *its own* A-value in grams. A mole of substance, in turn, contains Avogadro's number A<sub>0</sub> of atoms ( $\sim 6.02 \times 10^{23}$ at/mol as noted in App. 11). Thus, nuclide density  $n^{j}$  may be expressed as

$$n^{j} = \frac{A_{0} (at/mol)}{A (g/mol)} \times \rho (g/cm^{3}) = \frac{A_{0}}{A} \rho (at/cm^{3})$$

for density  $\rho$ . Uranium-238 with nominal density  $\rho^{28} \approx 19.1$  g/cm<sup>3</sup> would then have

$$n^{24} = \frac{6.022 \times 10^{23} \text{ at/mol}}{238.05 \text{ g/mol}} \times 19.1 \text{ g/cm}^3 = \boxed{4.83 \times 10^{22} \text{ at/cm}^3}$$

(Note that use of the integer value 238 compared to the actual atomic mass of 238.05 (Fig. 2-15) produces the same result.) [Nuclide density calculations for

mixtures are somewhat more complex because compositions are typically expressed as percentages or fractions by molar composition (or, equivalently, atom number), weight, or volume. Accordingly, the corresponding A- and p-values must be weighted averages of those for the constituent species.]

3. The activity of 1 g of <sup>238</sup>U:

Activity = 
$$\lambda^{28} n^{28}(t)$$
 (Eq. 2-4)  
 $T_{1/2}^{28} = 4.47 \times 10^9 \text{ y}$  (Fig. 2-15)  
 $\lambda^{28} = \frac{\ln 2}{T_{1/2}} = \frac{0.693}{4.47 \times 10^9 \text{ y}} = 1.55 \times 10^{-10} \text{ years}^{-1}$   
 $n^{28} = \frac{A_0}{A_{28}} = \frac{6.022 \times 10^{23} \text{ at/mol}}{238.05 \text{ g/mol}} = 2.53 \times 10^{21} \text{ at/g}$   
Activity =  $1.55 \times 10^{-10} \text{ y}^{-1} \times 2.53 \times 10^{21}$   
Activity =  $3.92 \times 10^{11} \text{ y}^{-1} \times \frac{1 \text{ y}}{3.15 \times 10^7 \text{ s}} \times \frac{1 \text{ Bq}}{1 \text{ s}^{-1}}$   
Activity =  $1.24 \times 10^4 \text{ s}^{-1} \times \frac{1 \text{ Ci}}{3.7 \times 10^{10} \text{ s}^{-1}} = 3.37 \times 10^{-7} \text{ Ci}$   
 $= 0.335 \,\mu\text{Ci}$ 

4. Time required for <sup>238</sup>U to decay by 1 percent:

$$n(t) = n(0)e^{-\lambda^{24}t} \quad (Eq. 2-6)$$
  

$$n(t) = 0.99n(0)$$
  

$$\lambda^{28} = 1.55 \times 10^{-10} \text{ y}^{-1}$$
  

$$\frac{n(t)}{n(0)} = 0.99 = e^{-(1.55 \times 10^{-10} \text{ y}^{-1})t}$$
  

$$\ln(0.99) = -(1.55 \times 10^{-10} \text{ y}^{-1})t$$
  

$$t = 6.48 \times 10^7 \text{ y} = \overline{(64,800,000 \text{ y})}$$

5. Average neutron density corresponding to a typical LWR thermal flux  $\Phi = 5 \times 10^{13} \text{ cm}^{-2} \text{s}^{-1}$ , assuming an effective speed  $v = 2200 \text{ m/s} - 2.2 \times 10^3 \text{ cm/s}$ :

$$\Phi = Nv \quad \text{(Eq. 2-18)}$$

$$N = \frac{\Phi}{v} = \frac{5 \times 10^{13} \text{ cm}^{-2} \text{s}^{-1}}{2.2 \times 10^5 \text{ cm} \text{ s}^{-1}} = \boxed{2.27 \times 10^8 \text{ cm}^{-3}}$$

6. Power produced by 1 g of <sup>235</sup>U fission in the LWR thermal flux in (5):

Fission rate = 
$$\sum_{i=1}^{25} \Phi dV = n^{25} \sigma_{i}^{25} \Phi dV$$
 (Eq. 2-19)

 $n^{25} dV = n' =$  number of atoms in the l-g sample

$$n' = \frac{6.022 \times 10^{23} \text{ at}}{235.04 \text{ g}} = 2.56 \times 10^{21} \text{ at/g}$$
  
$$\sigma_f^{25} = 585 \text{ b} \times \frac{10^{-24} \text{ cm}^2}{1 \text{ b}} = 585 \times 10^{-24} \text{ cm}^2$$

Fission rate =  $7.49 \times 10^{13}$  fissions/s

Power =  $7.49 \times 10^{13}$  fissions/s  $\times \frac{1 \text{ W}}{3.1 \times 10^{10} \text{ fissions/s}}$ =  $2.42 \times 10^3 \text{ W}$ Power =  $2.42 \times 10^3 \text{ W}$ 

For equal nuclide densities of <sup>235</sup>U and <sup>239</sup>Pu in a given reactor, find (a) fraction of fissions for each and (b) absorption mean free path for each nuclide and for the mixture. Assume each has an atom density of 10<sup>21</sup> aUcm<sup>3</sup>.

a. Fission fraction

$$F^{j} = \frac{\sum_{f}^{j} \Phi}{\sum_{f}^{\min} \Phi} = \frac{n^{j} \sigma_{f}^{j}}{n^{25} \sigma_{f}^{25} + n^{49} \sigma_{f}^{49}} \xrightarrow{\sigma_{f}^{13} = n^{44}} \frac{\sigma_{f}^{j}}{\sigma_{f}^{25} + \sigma_{f}^{49}}$$

$$F^{25} = \frac{585 \text{ b}}{585 \text{ b} + 750 \text{ b}} = \boxed{0.44} \text{ in } {}^{235}\text{U}$$

$$F^{49} = \frac{750 \text{ b}}{585 \text{ b} + 750 \text{ b}} = \boxed{0.56} \text{ in } {}^{239}\text{Pu}$$

b. Mean free paths

$$\Sigma_{u}^{25} = n^{25}(\sigma_{\gamma}^{25} + \sigma_{f}^{25})$$

$$n^{25} = 10^{21} \text{ at/cm}^{3} \times \frac{10^{-24} \text{ cm}^{2}}{1 \text{ b}} = 10^{-3} \text{ at/b·cm}^{\dagger}$$

$$\Sigma_{u}^{25} = 10^{-3} \text{ at/b·cm} (99 \text{ b} + 585 \text{ b}) = 0.684 \text{ cm}^{-1}$$

$$\Sigma_{u}^{49} = 10^{-3} \text{ at/b·cm} (271 \text{ b} + 750 \text{ b}) = 1.021 \text{ cm}^{-1}$$

$$\Sigma_{u}^{min} = \Sigma_{u}^{25} + \Sigma_{u}^{49} = 1.705 \text{ cm}^{-1}$$

$$\lambda_{u}^{25} = (\Sigma_{u}^{25})^{-1} = 1.46 \text{ cm} \text{ for } ^{235} \text{U}$$

<sup>†</sup>The unit at/b-cm is convenient and often used for atom densities employed in construction of macroscopic cross sections.

$$\lambda_a^{49} = (\Sigma_a^{49})^{-1} = 0.979 \text{ cm}$$
 for <sup>2,19</sup>Pu  
 $\lambda_a^{\text{mia}} = (\Sigma_a^{\text{mia}})^{-1} = 0.587 \text{ cm}$  for both

# **EXERCISES**

#### Questions

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- 2-1. Identify the constituent parts and describe the basic structure of an atom.
- 2-2. Define the following terms: isotope, half-life, and mean free path.
- 2-3. Define fissile, fissionable, and fertile. Identify the major nuclides in each of these three categories.
- 2-4. Describe the distribution of energy among the product particles and radiations associated with fission. Explain the basis for decay heat.

2-5. Sketch the relationship among the following reactor cross sections:

- a. total interaction
- b. scattering
- c. absorption
- d. fission
- e. capture
- 2-6. Differentiate between microscopic and macroscopic cross sections.

# Numerical Problems

- 2-7. Consider a thermal-neutron fission reaction in <sup>235</sup>U that produces two neutrons.
  - a. Write balanced reaction equations for the two fission events corresponding to the fragments in Fig. 2-7.
  - b. Using the binding-energy-per-nucleon curve (Fig. 2-1), estimate the total binding energy for <sup>235</sup>U and each of the fragments considered in (a).
  - c. Estimate the energy released by each of the two fissions and compare the results to the accepted average value.
- 2-8. The best candidate for controlled nuclear fusion is the reaction between deuterium and tritium. The reaction is also used to produce high energy neutrons.a. Write the reaction equation for this D-T reaction.
  - b. Using nuclear mass values in Table 2-4, calculate the energy release for the reaction.
  - c. Calculate the reaction rate required to produce a power of I W, based on the result in (b).
  - d. Compare the D-T energy release to that for fission on per-reaction and perreactant-mass bases.
- 2-9. Gamma rays interacting with <sup>2</sup>Be or <sup>2</sup>H produce "photoneutrons." Write the reaction equation for each and calculate the threshold gamma energy. (Use mass data from Table 2-4).
- 2-10. The nuclide <sup>2</sup>/<sub>1</sub>Po emits either an alpha particle or a beta particle with a halflife of 3.10 min.
  - a. Write equations for each reaction.
  - b. Calculate the decay constant  $\lambda$  and mean lifetime  $\tau$ .
  - c. Determine the number of atoms in a sample that has an activity of 100  $\mu$ Ci.
  - d. Calculate the activity after 1, 2, and 2.5 half-lives.

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#### of Basic Theory

#### TABLE 2-4 Nuclear Mass Values for Selected Nuclides

Nuclide	Nuclear mass (amu)		
<u></u>	1.008665		
(11	1.007825		
н	2.014102		
н	3.016047		
He	3.016029		
lle	4.002603		
Be	8.005305		
Be	9.012182		

- 2-11. Natural boron has a density of 0.128  $\times$  10<sup>24</sup> at/cm<sup>3</sup> and cross section  $\sigma_a =$  764 b and  $\sigma_r =$  4 b at an energy of E = 0.025 eV.
  - a. Calculate the macroscopic cross sections at 0.025 eV for absorption, scattering, and total interaction.
  - b. What fractional attenuation will a 0.025-eV neutron *beam* experience when traveling through 1 mm of the boron? 1 cm?
  - c. Assuming the absorption cross section is "one-over-o" in energy, calculate the macroscopic cross sections for boron for neutrons of 0.0025-eV and 100eV energies.
  - d. What thickness of boron is required to absorb 50 percent of a 100-eV neutron beam?
- 2-12. Estimate the fraction of thermal neutron absorptions in natural uranium that causes fission. Estimate the fraction of fast-neutron (2-MeV) fissions in natural uranium that occurs in <sup>238</sup>U. (Use data from tables and figures in this chapter.)
- 2-13. Recent measurements of particle fluxes from supernova place an upper limit on the mass of a neutrino (and antineutrino) as 11 eV. What fraction is this of the  $\beta$  particle or electron mass?

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<sup>†</sup> Full citations are contained in the General Bibliography at the back of the book.

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# 18 FUEL FABRICATION AND

HANDLING

# Objectives

After studying this chapter, the reader should be able to:

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- 1. Trace the major steps in LWR fuel fabrication from  $UF_6$  receipt to fuelassembly completion.
- 2. Identify the major differences between fabrication for  $UO_2$  and mixed oxide fuel assemblies.
- 3. Describe the major differences between LWR and HTGR fuel fabrication.
- 4. Explain the difference between self-generated and open-market recycle strategies for reactor use of plutonium.

5. Sketch the basic features of the "full-recycle" mode for a reference HTGR.

- 6. Describe the basic features of spent-fuel storage using: pool reracking; fuelassembly consolidation; dry-storage with vault, metal-cask, and concrete-cask systems; and away-from-reactor facilities.
- 7. Describe the basic features of the design and required testing of an LWR spentfuel cask. Identify two major purposes of the full-scale shipping cask crash tests conducted in the United States and the United Kingdom.
- Perform calculations of estimated fuel recycle mass flows, costs, and enrichment requirements based on data contained in this and the previous chapter.

# Prerequisite Concepts

Nuclear Fuel Cycle Fissile Plutonium Isotopes Chapter 1 Chapters 2 and 6

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Reactor Fuel AssembliesChapReactor SystemsChapUranium Processing and EnrichmentChap

Chapter 9 Chapters 10-12 Chapter 17

The fuel assemblies for six reference reactors were described briefly in Chap. 1 and in more detail in Chaps. 9-12. Each represents the results of a process that considers economy, reliability, safety, and other attributes, including fabrication, one of the subjects of this chapter.

Following fuel loading, reactor operation, and defueling (e.g., as summarized in Chaps. 10-12), the spent fuel assemblies are no longer directly useful for energy production. Their residual fissile content, however, can be recovered and recycled for future use. The large inventory of radioactivity in the spent fuel assemblies, however, leads to stringent requirements for storage and transportation.

# FABRICATION

The pellet-with-cladding-tube fuels used by the water-cooled reactors and LMFBR, as well as the microsphere fuels for the HTGR, were designed to include ease of fabrication. In each of the reference reactors (and others as well), the fuel assemblies are characterized by standardization, quality control, and acceptable cost.

#### **Oxide Fuels**

The reference reactors which use  $UO_2$  or mixed-oxide [sometimes called  $MO_2$  or MOX] pellets employ similar fuel fabrication techniques. The most substantial difference occurs when plutonium is present and many operations must be conducted with containment to protect personnel from its chemical and radiological toxicity. (Because plutonium and its daughter products do not emit highly penetrating radiations, heavy shielding and/or remote operations are generally *not* required.)

The first step of the fabrication process for most oxide fuels is production of  $UO_2$  powder. The natural uranium for CANDU fuel assemblies is converted to  $UO_2$  during the milling-purification step of the fuel cycle.

Slightly enriched uranium for LWR and PTGR systems, as well as depleted uranium for fast reactors, generally begins in the form of  $UF_6$  from gaseous-diffusion or gas-centrifuge enrichment plants. Conversion of  $UF_6$  to  $UO_2$  may be performed at a separate facility or at the fabrication plant. The solid  $UF_6$  is sublimed to its gaseous form and, in one specific process, is reacted with superheated steam through the "integrated dry route" using a rotary kiln for conversion to  $UO_2$  powder as shown in Fig. 18-1. Alternatively, the  $UF_6$  can be subjected to multiple step "wet chemistry" processing and then dried.

Succeeding steps in LWR fuel fabrication (generally following Fig. 18-1) include:

- blend UO<sub>2</sub> powder batches for uniformity
- add pore-former and binder materials to control the final porosity and other physical properties of the pellets
- precompact the powder to low-density solid form then granulate it to powder of uniform consistency

- dry, condition, screen, and add lubricant to the powder
- dry press powder into "green pellet" form (named for their color and also reflecting their "uncured" state)
- heat at moderate temperature to drive off the binder additives
- heat at high temperature to sinter the pellets to final density
- grind pellets to final diameter (typically ±0.01 mm)
- wash and dry pellets
- inspect pellets by visual, dimensional, chemical, and radiographic techniques and remove those that are unacceptable

The assembly of fuel pins then proceeds with steps to:

- form the pellets into stacks, weigh and record their content, and vacuum outgas them at elevated temperatures to remove moisture and organic contaminants
- load the dry pellet stack plus internal hardware (e.g., as shown in Figs. 1-7 and 9-7) into a clean, dry cladding tube which has one end cap welded in place
- inspect and decontaminate the loaded tubes
- weld the second end cap after evacuation and addition of inert-gas backfill (at atmospheric or higher pressure, as desired)
- leak test, radiograph, nondestructively assay the fissile loading, and inspect the welds

Following some combination of dimensional inspections, cleaning, etching, and autoclaving (to form a corrosion-resistant oxide film by treatment with high-pressure steam) the fuel pins and other associated hardware are formed into fuel assemblies. For the BWR this may include arranging pins of several enrichments with water rods in retention or spacer grids ["egg crates"] and surrounding the array with the exterior fuel channel (Figs. 1-6 and 10-4). When a BWR or other reactor uses axial pellet-topellet variations in enrichment or integral-burnable-poison (e.g., gadolinia) loading within a given fuel pin, quality control measures are increased to prevent misloading (and the potential for later in-reactor damage). PWR pins are loaded into fuel assemblies (Figs. 1-7 and 10-11) with retention grids, control-rod guide tubes, instrument tubes, and, if appropriate, burnable poison rods.

Small interelement spacer pads are welded to each of the short CANDU fuel elements [rods]. The rods are then welded to end support plates in a tightly packed circular configuration (Fig. 1-9). The double-section fuel bundles for the PTGR (Fig. 1-8) are assembled around the central coolant tube in stainless steel spacer grids.

Mixed-oxide fuel fabrication follows similar steps with a separate input stream for PuO<sub>2</sub>. Differences include steps that:

- calcine ["bake"] plutonium nitrate solution from the reprocessing plant to form PuO<sub>2</sub> powder
- blend PuO<sub>2</sub> and depleted UO<sub>2</sub> batches to a homogeneous mixture and mill it to uniform consistency

These and subsequent steps take place in a contained environment (e.g., glove boxes described below) until the pellet stacks are loaded into cladding tubes, the last end cap is welded into place, and the tubes are verified to be leak tight and free of plutonium



contamination. From then on the mixed oxide pins are handled in the same manner as  $UO_2$  pins with similar fuel assembly configurations (e.g., Figs. 1-6 and 1-7 for BWR and PWR, respectively).

As described in Chaps. 9 and 12, LMFBR fuel pins may be fabricated with either core fuel, blanket fuel, or both, plus a variety of hardware (Fig. 9-8). A wire wrap welded to each pin allows assembly in a tightly packed becagonal array that is then encased in an exterior flow channel (Figs. 1-11 and 12-7).

# **HTGR Fuels**

As would be expected, the fabrication of HTGR fuel assemblies is quite different from that considered above for the conventional oxide fuels. Figure 18-2 shows the basic features of one manufacturing process. A number of steps depend on the use of *fluidized bed* technology, where droplets or particles are suspended for relatively long periods of time by carefully adjusted upflow of gases. The droplets can be dried or coated while suspended.

The first step of the manufacturing process for the fissile TRISO particles (Figs. 1-10 and 9-9) is loading uranium nitrate solution into weak-acid resin spheres, as



FIGURE 18-2 Fuel fabrication process for high-temperature gas-cooled reactor [IITGR] fuel. (Courtesy of GA Technologies.)

shown by Fig. 18-2. The resulting microspheres are then dried and sintered to produce high-density  $UC_2$  or UOC kernels. The various carbon and silicon carbide coatings are applied by controlled fluidized-bed treatment with hydrocarbon and silane gases, respectively.

The fertile BISO particles are produced by preparing a thorium hydrosol [sol] broth from  $ThO_2$  powder and dilute nitric acid. Sol droplets from a vibrating nozzle are converted to gelatin [gel] spheres by extracting the water content in a column containing ammonia gas. After a screening process selects spheres of the proper size, drying and sintering in a high-temperature furnace produces the final  $ThO_2$  kernels. The two graphite coatings are applied by a fluidized-bed process.

Finished BISO and TRISO particles are metered at the desired ratio and blended with graphite. The mixture is then formed into a packed bed for injection molding with a carbonaceous binder, as shown in Fig. 18-2. The rods are loaded into machined hexagonal graphite blocks of the type depicted by Figs. 1-10 and 11-8. The fuel assembly is heated in a high-temperature furnace to remove volatile components of the fuel rod binder and to convert the binder residue to a carbon char matrix. The cured fuel element is complete after it has been fitted with graphite positioning dowels.

#### **General Considerations**

The high economic value of fuel assemblies and their constituent parts dictates very careful handling during all processing steps. Finished assemblies are cleaned and packaged with protective covers for temporary on-site storage. They are shipped in rugged, shock-mounted casks. The casks, individually or in small groups, are transported by truck or rail to their reactor-site destinations.

Unirradiated uranium fuel constitutes minimal radiological and chemical hazards. Plutonium, however, must be isolated from contact with operating personnel.

Plutonium is usually handled in *glove boxes*—enclosed, sealed work spaces that may range from less than a cubic meter to the size of a room. These boxes typically have: (1) flexible gloves that are a portion of the physical containment boundary and with which work can be performed; (2) air-lock devices and detailed procedures for insertion and removal of fuel and other materials; and (3) controlled atmosphere (e.g., with negative-pressure, filtered discharge, and continuous monitoring). Plutonium, mixed-oxide, and other designated fuel materials are handled in glove boxes, or equivalent containments, until they are sealed in cladding tubes, as noted previously. Automated processing for glove-box operations is under development and in limited use in the United States, Japan, and elsewhere.

If the fuel contains radionuclides that emit penetrating radiations, glove boxes are inadequate. Remote operations behind heavy shielding may be necessary. Although procedures and equipment are well developed for such operations, their cost and inconvenience make remote activities a last resort. The HTGR fuel fabrication processes have been developed to be ammenable to remote operations at such time as <sup>233</sup>U recycle (with the attendant <sup>232</sup>U contamination noted in Chap. 6) becomes desirable. Use of <sup>233</sup>U or fission-product-contaminated plutonium in any of the other reference reactor designs would require substantial modification of existing processes.

All facilities that handle plutonium or other highly radioactive materials must take steps to limit radiation exposures (external and internal) and radionuclide releases

to as low as reasonably achievable [ALARA] levels. Ventilation and filtration systems are designed for this purpose. Support is provided by monitoring with remote sensors and routine surveys by radiation-safety personnel.

Criticality safety measures are applied to fabrication operations (except for those solely with natural or depleted uranium). With slightly enriched uranium and equivalent mixed-oxide compositions, wet chemistry operations are performed in limited-geometry vessels (e.g., slender cylindrical tanks) while dry powder, pellets, and fuel pins are processed, handled, and stored with provisions for combinations of mass and geometry limits, moderator exclusion, and spacing. More highly enriched uranium and plutonium, which can be critical even when dry (i.e., without any moderator), are subject to even more stringent limits, e.g., smaller diameters, lower masses and volumes, and wider spacing.

# Status

Many of the countries with major nuclear programs have fuel fabrication capability to match their reactor types (e.g., as summarized in Table 17-2). In the case of mixedoxides, capacities are tied to national plans for LWR plutonium recycle and for advanced-reactor and LMFBR development as described in Chap. 12.

A recent trend has been toward cooperative and joint fuel fabrication ventures with a distinctly international flavor. In the United States, for example, this has included four of the five LWR fuel fabricators (with General Electric the exception). Advanced Nuclear Fuels (formerly owned by Exxon) is now a Siemens-KWU company. The B&W Fuel Company is owned by Babcock & Wileox and the French companies Framatome, Cogema, and Uranium Pechiney. Combustion Engineering is now owned by ABB Brown Boveri. Finally, Westinghouse and ABB-Atom in Sweden (itself the result of earlier acquisition of ASEA-Atom by ABB Brown Boveri)—PWR and BWR manufacturers, respectively—have had subcontract agreements for marketing the other's fuel designs.

# FUEL RECYCLE

All discharged reactor fuel has residual fissile inventory that could be recycled to reduce overall uranium-resource and enrichment requirements in future cycles. It has been estimated that full uranium and plutonium recycle in an LWR, for example, could cut yellow cake and enrichment demands by about one-third and one-quarter, respectively (e.g., as calculated by comparing Fig. 17-1*a* and 17-1*b*).

As described in the previous chapter, the economy of recycle depends on cost comparisons between once-through fuel cycle operations and those for reprocessing and recycle fabrication. The high costs of MOX fabrication, including glove-box operations and other safety- and safeguards-related systems and reprocessing, make recycle expensive. This, coupled with falling costs for yellow cake and enrichment, has made recycle economically unfavorable (e.g., as shown by Table 17-1).

From another vantage point, sizeable quantities of plutonium already exist in the countries in which spent fuel is reprocessed. A study by the OECD (1989b) concluded that, if this particular material is considered to be essentially "free," its use could produce meaningful cost savings compared to contemporary uranium prices. Where reprocessing must "pay its own way," the economics remain unfavorable.

# LWR Considerations

Fissile mass flows for PWR once-through and self-generated uranium and plutonium recycle modes are shown by Figs. 17-1a and 17-1b. At 0.83 wt%, the <sup>235</sup>U content exceeds the assay of natural uranium. The plutonium content, equivalent to 0.5-0.7 wt% of the heavy metal, is an isotopic mixture (e.g., as shown by Fig. 6-2). Similar trends exist for BWR systems,

Uranium recycle is complicated by buildup of <sup>236</sup>U from nonfission neutron capture in <sup>235</sup>U (as described in Chap. 6). When gaseous diffusion was the only available enrichment technology, there was great concern about contamination of the massive quantities of material in these continuous-process systems. "Fissile equivalent" algorithms were developed to assign value to feed material of such off-normal compositions. A potentially better option, however, was use of a separate enrichment plant dedicated to uranium recycle. Development of gas-centrifuge enrichment in the interim has improved the prospects. Urenco's marketing strategy, for example, has stressed the ease with which its centrifuge cascades can be cleared between reprocesseduranium and natural uranium runs. Laser processes, when available, may provide the ideal, "zero-holdup" solution to uranium recycle.

Plutonium may be employed in LWR's in two general ways. In the self-generated recycle [SGR] mode, a reactor uses only the mass of plutonium it produces. The openmarket recycle [OMR] mode considers any amount of plutonium from a small quantity to a full core. The SGR option allows reactor fuel management based on a fixedfraction plutonium inventory. For OMR operations, the system must be designed with enough flexibility to accommodate anywhere from zero to 100 percent plutonium as the fissile content of the core.

The fissile content of recycle fuel batches is most likely to include both plutonium and slightly enriched uranium. One fabrication option is to mix the two homogeneously so that each fuel pin has the same composition. This has an advantage in terms of power peaking, but would, of course, mean that glove-box operations would be necessary for each pin. The other option-to produce separate uranium and mixed-oxide [PuO<sub>2</sub> plus depleted UO<sub>2</sub>] pins—reduces the need for glove-box operations. However, it also leads to some burnup-dependent mismatches between the pin types that can. result in high power peaking problems. Such problems are potentially greatest for separate uranium and plutonium assemblies, but also exist in the "small-island" concept where each assembly contains some plutonium pins. Overall economic considerations---mainly fabrication costs versus power capability----ultimately serve as the basis for selecting among the procedures.

The characteristics of plutonium with successive recycle are tracked in Table 18-1 for a PWR using only the self-generated plutonium. The initial loading is slightly enriched  $UO_2$  similar to that of Fig. 17-1*a*. The first reload is this same fuel because the discharge batch must be reprocessed before its plutonium is available for recycle. In subsequent reloads, the plutonium recovered from all discharged fuel is recycled in separate MOX fuel pins. Table 18-1 shows that the proportion of MOX in each reload batch rises from 18 percent at the first recycle to just under 30 percent in the sixth cycle, where equilibrium is said to occur because as much plutonium is recovered from spent MOX and UO, fuel pins as had originally been loaded into the MOX fuel rods. The percentage of plutonium in MOX rises also, from 4.7 percent plutonium in

	lnital cvcle	First reload	Second reload (first recycle)	Third reload (second recycle)	Fourth reload (third recycle)	Fifth reload (fourth recycle)	Sixth reload (fifth recycle)
						07	3.0
<sup>35</sup> U in UO <sub>2</sub> , w1%	2.14	3.0	3.0	3.0	5.0 6.89	12.5	8.05
∿in MOX, wt% 40X, ≸ of fue!			4.12 18.4	23.4	26.5	27.8	28.8
Discharged <sup>233</sup> U.							
wife	0.83						
Discharged Pu							
composition wr95			1	7 T T	47.1	40.9	40.0
		56.8	49.7	9. F	104	29.6	29.8
Ĩd <sub>or</sub>		23.8	27.0	58.7	r. c.	17.4	17.3
Pd 17.		14.3	16.2	2.11	1.11	12.1	12.9
Nd Sk		5.I	١.٢	C.4			

Sixth reload

TABLE 18-1 Plutonium Utilization in a PWR Recycling Only Self-Generated Plutonium

2. Clarendon Press, Oxford, England, 1982. With permission of Oxford University Technology Power Source: Adapted from W. Marshall (Ed.), Nuclear Press.

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natural uranium in the first recycle to 8 percent plutonium at equilibrium. Table 18-1 also shows that the fissile content of the plutonium (i.e.,  $^{239}$ Pu plus  $^{241}$ Pu) worsens with recycle from a little more than 70 percent in the discharged UO<sub>2</sub> fuel to 57 percent in the MOX fuel at equilibrium. During this time, the fraction of MOX in-reactor increases from 6-7 percent at the start of the first recycle to the equilibrium value of 30 percent.

Recycle operations depend, of course, on spent fuel reprocessing. (Reprocessing status is shown in Table 17-2 and considered further in the next chapter). In the United States, for example, a generally uneconomic outlook for reprocessing and MOX fuels has limited LWR recycle strictly to experimental activities.

Belgium, Germany, Switzerland, Japan, and especially France have developed plans to use MOX for the long-term economies they expect it to provide. The French, for example, loaded MOX fuel into the St. Laurent B-1 reactor in late 1987 signaling the start of routine plutonium recycle scheduled to extend to 16 units by 1995. Japan, similarly, started with four MOX assemblies in the small Mihama PWR with plans for quarter-core, three-cycle tests in large PWR and BWR units by the end of the century.

#### HTGR Recycle

The reference HTGR has many recycle options for uranium and thorium, some of which are represented on Fig. 18-3. Four possible (arbitrarily-named) cycles are:

- Nonrecycle—Highly enriched uranium is used in TRISO fuel for each cycle with discharged <sup>235</sup>U and <sup>233</sup>U stored or sold.
- Full recycle—All uranium is recycled in the BISO particle along with the thorium, and makeup <sup>235</sup>U is fabricated into TRISO particles.
- 3. Type I segregation—Bred <sup>233</sup>U is recycled with makeup <sup>235</sup>U in TRISO particles, but discharged <sup>235</sup>U is not recycled.
- 4. Type II segregation—Makeup <sup>235</sup>U is in TRISO particles with thorium and recycled <sup>233</sup>U in BISO (or other separate) particles.

The material flows for the HTGR full recycle mode are also shown by Fig. 17-1d.

The highly enriched uranium is recycled once or not at all, based on considerations shown by Table 18-2. The initial <sup>235</sup>U content is depleted by 92 percent and 99 percent of initial mass during the first and second cycles, respectively. The very large <sup>236</sup>U content (with its reactivity penalty) dictates against reenrichment or use past the second cycle.

Thorium is not likely to be recycled directly because it contains the short halflived  $^{278}$ Th isotope from the  $^{232}$ U decay chain (of Fig. 6-4). Instead, it may be stored for later use as indicated on Fig. 18-3.

There is very little plutonium produced from the highly enriched uranium feed in the reference HTGR design (Table 18-2). However, the next generation of HTGR, or MHTGR (Chap. 14), calls for  $\sim 20$  wt% <sup>235</sup>U fuel that would generate significantly more plutonium (from the 80 wt% <sup>238</sup>U). This, of course, would complicate fuel cycle operation (and Fig. 18-3) by introducing a new material stream.



#### FIGURE 18-3

HTGR recycle options. (Courtesy of Oak Ridge National Laboratory, operated by the Union Carbide Corporation for the U.S. Department of Energy.)

# Other Reactor Systems

The plutonium inventory in spent natural-uranium fuel in the CANDU-PHW fuel bundles is relatively low. This, coupled with low uranium prices and high reprocessing and MOX fabrication costs, make recycle in Canada highly unlikely in the foreseeable future. However, with the great flexibility afforded by on-line refueling, use of plutonium or <sup>233</sup>U fuel or a thorium fuel cycle is possible at any time (e.g., as described in Chap. 11). Tandem fuel cycles could also be viable, e.g., use of LWR slightly enriched or recovered uranium (because the latter has an assay above that of natural uranium as shown by Fig. 17-16 and Table 18-1).

The Soviet PTGR was designed to produce both electricity and plutonium (e.g., as described in Chap. 11). Although fuel-cycle strategies have not been reported, the system's capability for on-line refueling could provide some of the same flexibility the CANDU systems have.

The advanced converter reactors described in Chap. 12 also are tied to recycle. Framatome's RCVS spectral-shift converter reactor, for example, was designed for increased uranium utilization primarily by producing more plutonium and burning it

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# TABLE 18-2 Composition of Discharged HTGR Uranium Feed Material<sup>†</sup>

	Origi compo	Original composition		Composition after one 4-year cycle		Composition after two 4-year cycles	
Isotope	g/kg U	%	g/kg U	9%	g/kg U	%	
U-234	10	0.1	5	1.9	2	1.2	
U-235	930	93.0	75	28.6	7	4.3	
U-236	2	0.2	133	\$0.8	118	72.4	
U-238	58	5.8	49	18.7	36	22.1	

<sup>†</sup>Adapted from R. C. Dahlberg et al., "HTGR Fuel and Fuel Cycle Summary Description," GA-A12801 (Rev.), January 1974.

in place. Its unique fuel design, however, also allows optimized operation with recycled plutonium fuel.

The conceptual spectral-shift-control reactor (SSCR) also promises improved resource utilization. The moderate savings from an LWR-like once-through cycle could .- be increased with plutonium recycle and increased even more with a <sup>233</sup>U-thorium cycle.

The overall design concept of the breeder reactors (e.g., the light-water, moltensalt, and liquid-metal fast breeder reactors described in Chap. 12) is geared to fuel reprocessing and recycle. The LWBR and MSBR operate on thermal neutrons in <sup>233</sup>Uthorium fuel cycles which, once established, would require only thorium as fresh feed.

The LMFBR is based on a plutonium fuel cycle, generally with mixed-oxide as shown by Fig. 17-2e. Core and blanket fuel management (e.g., as introduced in Chaps. 9 and 12) may produce separate plutonium recycle streams. Depleted or natural uranium provides the fresh feed material for MOX fuel fabrication.

Initial LMFBR operation is generally with plutonium from LWR sources (whose isotopic composition happens to be well-suited to this purpose as discussed in Chap. 6). France especially, but also Japan, the United Kingdom, and others, recycle plutonium fuels for use in prototype fast reactors.

#### SPENT FUEL

Storage and transportation of spent fuel assemblies are important components of the nuclear fuel cycle, whether or not reprocessing and recycle occur. Interim storage at the reactor site allows the assemblies to cool prior to reprocessing or disposal. As existing on-site storage space is filled, capacity must be added or the spent fuel must be shipped to away-from-reactor-storage or disposal sites. Reprocessing and recycle would also require spent-fuel shipment.

# Storage

Most of the world's spent fuel is currently stored in water basins at LWR or CANDU sites. Water is a particularly convenient medium because it is inexpensive, can cool by natural convection, and provides shielding and visibility at the same time. However,

water is also a neutron moderator (with inherent criticality implications) and potential contributor to corrosion.

CANDU fuel is placed horizontally in trays that reside in the high-capacity storage bay shown in Fig. 11-4. Spent CANDU fuel bundles have relatively low burnup (and corresponding heat load) and are subcritical in any configuration in ordinary water. Thus, they can be stored readily at high density in the water pool. Lifetime fuel storage capacity has been built into many of these reactors because reprocessing and recycle have not been planned. Some CANDU fuel is also in storage in concrete casks and silos.

LWR and PTGR fuel assemblies are stored vertically in a pool with a fixedlattice structure. The LWRs traditionally have used aluminum racks with generous center-to-center spacing that provides both adequate natural-convection cooling and a subcritical configuration.

Dry storage wells with external water cooling have been designed for the graphite HTGR fuel blocks. LMFBR fuel assemblies are stored in liquid-sodium-filled basins, in principle, for a brief period of time prior to recycle of their plutonium content.

Many on-site storage facilities for LWR fuel were sized for a limited number of discharge batches in anticipation of fuel reprocessing and recycle. In the absence of reprocessing, many of the operating reactors, especially in the United States, have faced or will face both extended storage times and inadequate capacity. Fortunately, the zirconium-alloy clad has proven to resist degradation, e.g., as demonstrated by up to three decades of experience with LWR and CANDU fuels. Capacity enhancements include pool-storage alternatives and addition of dry storage. (Long-term options related to national waste-management strategies are covered in the next chapter.)

#### Pool Storage

Meeting the capacity challenge with construction of new pools or physical expansion of existing ones generally is too slow and too costly (i.e., has large, up-front capital costs). Therefore consideration has been focused on a combination of methods including:

- 1. Reracking-conversion of pools for high-density storage
- Burnup credit—reduction of fuel-assembly separation by taking credit for fuel burnup effects
- Rod consolidation—reduction of fuel-assembly volume through disassembly and compaction
- 4. Extended burnup-generation of less spent fuel per unit of power production
- 5. Transshipment-move fuel from one pool to another

High-density fuel-assembly storage is available with stainless-steel rack structures having integral neutron poisons such as boron carbide. Burnup credit may allow close spacing with less stainless steel and poison between assemblies. Reracking has been done in nearly all U.S. reactors whose pools can support the weight and still meet seismic and other applicable requirements. Another reracking alternative, which also depends on seismic capability, is adding a second tier to the pool as has been done at two small U.S. plants. In both reracking alternatives, forced cooling may be required to compensate for the high decay-heat load associated with freshly discharged fuel assemblies.

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Rod consolidation methods have been developed to remove end fittings, pull or push intact fuel rods from the assembly structure (e.g., Fig. 1-6 for BWR and Figs. 1-7 and 10-11 for PWR), place the rods in a canister in a close-packed configuration, and compact the remaining structural parts in the same or a different canister. The rods themselves can be consolidated in a 2:1 ratio and the structure from 6:1 to 10:1. Thus, a net reduction in volume of from 1.5:1 to 1.7:1 can be achieved. If the structure can be removed from the pool and stored elsewhere or disposed (e.g., as intermediatelevel waste as described in the next chapter), the net consolidation can be the 2:1 ratio of the fuel rods. The resulting increase in mass loading, of course, requires that seismic requirements be met, more so if in concert with reracking. More than a dozen demonstration projects have been completed or are underway in the United States with large-scale use planned by several utilities. The United Kingdom has applied consolidation to AGR fuel.

Extended burnup is an increasingly popular fuel-management option (described in Chap. 9), in part for reducing spent-fuel volumes. Transshipment from one pool to another is generally done within the same utility and, of course, applies only if space is available. Conceptually, this is no different than using away-from-reactor storage or an interim spent-fuel repository.

#### Dry Storage

The alternative to pool or "wet" storage is dry storage in vaults, metal casks, and concrete modules. The viability of spent-fuel storage in a variety of configurations backfilled with helium or nitrogen has been demonstrated in Canada, Germany, Switzerland, the United States, and the Soviet Union. Temperature-versus-time data also have been established. On-going studies eventually may determine conditions under which air can be used as the backfill medium (in addition to its traditional role in forced- or natural-convection cooling of the sealed containers).

One type of dry-storage vault consists of sealed concrete containment structures in modular units. Individual spent-fuel assemblies are stored inside, each in a separate vertical metal tube. Decay-heat removal is by natural-convection air cooling. Vault storage has been used primarily in the United Kingdom for gas-cooled reactor fuels and in France for fuels from early research and power reactors.

The metal cask generally consists of a coated metal body with fins for decayheat removal by convective cooling (e.g., as in one of the configurations in Fig. 18-4). One series of casks holds from 21 to 36 PWR or 52 to 76 BWR assemblies in an upright position on a concrete slab. Many metal casks are solely for temporary storage and, thus, non-transportable. Others are multipurpose with a common module for intermediate storage, transport, and long-term silo storage, e.g., as shown in Fig. 8-4 with a module holding 12 PWR assemblies.

Metal cask demonstrations have been performed cooperatively by U.S.DOE, EPRI, and U.S. utilities. A variety of casks (e.g., GNSI CASTOR V/21 and X/33, Transnuclear TN-24P, Westinghouse MC-10, NAC-28, and NAC-31), some of which already are seeing use in the United States and overseas, have been or will be included in the tests.

Concrete casks are similar to the metal storage casks except that the fuel is



#### FIGURE 18-4

Multipurpose cask for temporary on-site storage, shipment, and long-term storage. [Adapted courtesy of Nuclear Engineering International with permission of the editors.]

sealed in a stainless-steel canister and inserted into a hollow-concrete cask body (similar to the long-term-storage configuration of Fig. 8-4). Two examples are the:

- NUTECH horizontal modular storage [NUHOMS] system with stainless-steel canisters each holding seven PWR assemblies and stored horizontally in concrete modules
- NUPAC system with one or two assemblies each in smaller canisters stored vertically in concrete modules

These and other systems with capacities up to 28 PWR assemblies have been subject to demonstration in a U.S.DOE project in cooperation with U.S. commercial nuclear plants. Full-scale use of concrete casks has been scheduled by several U.S. utilities.

#### Away-From-Reactor Storage

An alternative or adjunct to increasing on-site spent-fuel storage capacity is the use of away-from-reactor [AFR] facilities (Table 17-2). AFR facilities are generally operated commercially or by a government. When intended strictly for interim spentfuel storage, AFR operations have many similarities to at-reactor pools and dry-storage configurations, including the ability to handle consolidated fuel. Long-term storage or waste-disposal applications (subjects of the next chapter) differ primarily in spent-fuel packaging.

Independent spent fuel storage installations [ISFSI] and AFRs often are considered synonymous. However, according to U.S.NRC regulations, the ISFSI classification also includes facilities that accept transshipment of fuel from another reactor operated by the same utility. A new storage facility, e.g., based on dry storage methods

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described above, used only for fuel from the same site also may be considered an ISFSI when it is separate from the original reactor complex.

A small U.S. away-from-reactor storage facility for LWR-spent fuel is operated in the spent-fuel pool of an inoperable reprocessing plant in Morris, Illinois. It has a capacity of about 750 t or 20–25 discharge batches. All fuel assemblies are received in rail-mounted shipping casks (e.g., like that described below). The cask is moved from the rail car into an air-locked room where it is decontaminated as necessary. The "clean" cask is transferred to an underwater storage area where the fuel assemblies are removed and placed into poisoned, stainless-steel *baskets* (one holding a three-bythree array of BWR assemblies, another a two-by-two array of PWR assemblies). The baskets are transferred to the pool where they are latched in place in a vertical position.

The Swedish CLAB is a storage pool that is located underground and receives spent LWR fuel in a packaged form intended for later disposal (as described in the next chapter). Finland has an ISFSI-type pool for excess spent fuel from the Olkiluoto site. Fuel from Finland's Loviisa station was to be returned to the Soviet Union.

Although the former German Democratic Republic also returns spent fuel, it operates an AFR facility at the site of the 8-unit Nord station. The four pools with high-density racks extended the site's storage capacity from three years to eight years at the request of the Soviet Union. These pools are covered by a floor of steel plates which, in concert with IAEA seals, prevent unauthorized spent-fuel movement (a material safeguards measure considered in Chap. 20).

Virginia Power's Surry Station has installed an ISFSI that began with four concrete casks that were loaded immediately on delivery to alleviate a pressing need for storage capacity. Dry-cask AFR capacity for LWR fuel is also in operation in Germany with more under construction, including a yet-to-be licensed facility at Gorleben (which is also the proposed site of a high-level-waste repository as described in the next chapter). The dry cask and vault facilities in France and the United Kingdom that were mentioned earlier are also away-from-reactor facilities.

# Transportation

Until they have been irradiated in a reactor, fuel cycle materials do not pose serious transportation hazards. This is not true for the spent fuel, which may contain sizeable quantities of plutonium and is "hot" both thermally and radiologically. Shipments of spent fuel are, thus, subject to very stringent domestic and international regulations.

#### Regulations

Responsibility for safe shipment of commercial nuclear materials in the United States is shared by the Nuclear Regulatory Commission and the Department of Transportation [DOT]. DOT establishes general regulations for packaging, loading, and operation of vehicles, as well as for inspection, shipping documents, and accident reporting. The NRC sets packaging standards for highly-radioactive materials. It also recommends performance standards related to the potential for adverse effects on operating personnel and the general public due to radiation or release of radioactive material. The U.S. regulations for transporting radioactive materials have been developed to be consistent with those of the International Atomic Energy Agency [IAEA]. Packages are generally divided into two categories from a radiological standpoint:

- 1: Class A—"small" quantities which if released directly to the environment would have minimal consequences (i.e., be within prescribed limits)
- 2. Class B-non-class A package which must be designed for containment under (defined) normal and accident conditions

Class B packages, including spent-fuel transport containers, must be able to withstand all normally anticipated transport contingencies with:

- no leakage of material
- radiation levels <200 mr/h at the surface and 10 mr/h at 1 m</li>
- no criticality
- surface temperature <80°C</li>

Each container type must also be shown to be capable of withstanding the following accident sequence with most of the shielding still intact and with only limited escape of coolant and/or inert radioactive gases:

- free fall of 10 m on a flat, unyielding horizontal surface (an approximation to a 95 km/h [60 mi/h] vehicle crash)
- puncture test with a 100-cm drop onto a 15-cm-diameter bar located to maximize damage
- thermal test with a temperature of ~800°C for 30 min (an approximation to a gasoline or kerosene fire)
- water immersion at a depth of >1 m for 8 h

# Spent-Fuel Casks

Casks for transporting spent-fuel assemblies must be designed to accommodate both high radiation levels and large decay-heat loads while meeting all necessary regulatory requirements such as those presented above. One container for LWR fuel is the General Electric IF-300, shown separately and rail-car mounted in Fig. 18-5. Its important components are:

- a fuel basket holding seven PWR assemblies or 18 BWR assemblies (similar to the inner storage module in Fig. 18-4)
- a stainless-steel cylinder sealed with a closure head
- depleted-uranium or lead shielding material
- an outer stainless-steel shell
- a corrugated stainless-steel outer jacket holding neutron shielding (usually water)
- · fins to aid forced cooling (if required) and minimize impact damage

With slight modifications of the fuel basket, the cask in Fig. 18-5 could accommodate most types of reactor fuel (e.g., from CANDU to LMFBR), TMI-2 fuel-debris canisters, or solidified waste. Reactor-specific casks, however, have the advantage of more optimized capacity. An LMFBR cask, for example, accommodates smaller but highly active (e.g., Table 6-3), high-power-density assemblies (because the conceptual LMFBR is designed for very high burnup and short at-reactor storage prior to reprocessing).

HTGR fuel can be shipped in a simpler cask because the assemblies have the

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FIGURE 18-5





General Electric IF-300 spent-fuel shipping cask (a) separately, and (b) in normal rail-transport configuration. (Courtesy General Electric Company.)

high inherent heat capacity of graphite and will have been cooled longer. The same cask also can be used for transporting fresh fuel because the external shielding, roughly a 3.5-in cylindrical-shell of depleted uranium for a 6-assembly stack, is not excessive.

# Crash Tests

In 1977-1978 Sandia Laboratories prepared and conducted a series of full-scale crash tests for spent-fuel casks of a type similar to that in Fig. 18-5. The tests were not intended to replace the series required for regulatory certification. Instead, one purpose was to evaluate current capabilities for predicting crash results. The other major purpose was to demonstrate for the general public the overall safety of the transport method even under highly unlikely accident conditions.

The series consisted of:

- 1. 97 and 130 km/h crashes of tractor-trailer rigs with a spent-fuel cask into a massive. stationary concrete barrier
- 2. a 130 km/h locomotive crash on a stationary, cask-loaded tractor-trailer rig at a simulated grade crossing
- 3. a 130 km/h impact of a special railcar-mounted cask into the concrete barrier with a subsequent 125-min burn in JP-4 fuel at 980-1150°C

The results of the tests demonstrated that the predictive methods are very accurate. Each cask survived with minimal damage and without leakage of (simulated) radioactive material. The photographs in Fig. 18-6 show selected features of the test series.

In 1986, the United Kingdom conducted similar public demonstration tests for their roughly cube-shaped spent-fuel "flask." Following a 9-m drop on a solid unyielding anvil, the flask was placed in position on railroad tracks so that an oncoming full-sized diesel locomotive traveling at 160 km/h struck its lid (where there was the best chance the lid would be ripped off). Although the flask was thrown 60 m, it survived intact, validating model tests and computer predictions, as well as providing spectacular (if expensive) public relations.

# EXERCISES

#### Questions

- 18-1. Trace the major steps in LWR fuel fabrication from UF<sub>6</sub> receipt to fuelassembly completion.
- 18-2. Identify the major differences between fabrication for UO<sub>2</sub> and mixed-oxide fuel assemblies.
- 18-3. Describe the major differences between LWR and HTGR fuel fabrication.
- 18-4. Explain the difference between self-generated and open-market recycle strategies for reactor use of plutonium.
- 18-5. Sketch the basic features of the "full-recycle" mode for a reference HTGR.
- 18-6. Describe the basic features of spent-fuel storage using: pool reracking; fuelassembly consolidation; dry-storage with vault, metal-cask, and concrete-cask systems; and away-from-reactor facilities.



(c) Spem-fuel stripping eask full-scale accident test sequences: (d) and (d) 1.30 km/h tractor-itailer crash; (c) FIGURE 18-6



(P)

(g) EFGURE 18-6 Continued): (c) and (c) post-test sequences (Continued): (c) and (f) 1.30 km/b taileat impact and burn: and (c) post-test scatus of cask. (Photographs Courtesy of Sandia National impact and burn: and (c) post-test scatus of cask. (Photographs Courtesy of Sandia National impact and burn: and (c) post-test scatus of cask. (Photographs Courtesy of Sandia National impact and burn: and (c) post-test scatus of cask. (Photographs Courtesy of Sandia National impact and burn: and (c) post-test scatus of cask. (Photographs Courtesy of Sandia National impact and burn: and (c) post-test scatus of cask.

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