"Someday man will harness the rise and fall of the tides, imprison the power of the sun, and *release atomic power*."

—Thomas Alva Edison

# 1.1 Introduction

There is no doubt that energy has been driving and will drive the technological progress of the human civilization. It is a very vital component for the economic development and growth, and thus our modern way of life. Energy has also been tied to the national security concerns. It has been projected that the world energy demand will almost double by the year 2040 (based on 2010 energy usage), which must be met by utilizing the energy sources other than the fossil fuels such as coal and oil. Fossil fuel power generation contributes to significant greenhouse gas emissions into the atmosphere and influences the climate change trend. Although several research and development programs (e.g., carbon sequestration and ultrasupercritical steam turbine programs) have been initiated to make the fossil power generation much cleaner, they alone will not be enough to fend off the bigger problem. Therefore, many countries worldwide have recognized the importance of clean (i.e., emission-free) nuclear energy, and there are proven technologies that are more than ready for deployment. The use of nuclear energy for the power generation varies widely in different parts of the world. The United States produces about 19% (2005 estimate) of its total energy from nuclear sources, whereas France produces  $\sim$ 79% and Brazil and India rely on the nuclear energy for only about 2.5% and 2.8% of their energy needs, respectively. Japan, South Korea, Switzerland, and Ukraine produce 30%, 35%, 48%, and 40%, respectively, of their energy requirements from the nuclear sources. It is important to note that the fast growing economies like China, India, and Brazil produce relatively less electricity from the nuclear sources. Hence, there are tremendous opportunities for nuclear energy growth in these emerging economies as well as many other countries. Nuclear reactors have been built for the primary purpose of electricity production, although they are used for desalination and radioisotope production.

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An Introduction to Nuclear Materials: Fundamentals and Applications, First Edition.

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There are now about 440 nuclear power reactors worldwide generating almost 16% of the world electricity needs; among them, 104 nuclear reactors are in the United States. Since the first radioactive chain reaction that was successfully initiated at the University of Chicago research reactor in the 1940s, the field has seen an impressive growth until Three Mile Island and Chernobyl accidents happened. Following these incidents, the public confidence in the nuclear power dwindled, and the nuclear power industry saw a long stagnation. However, the US government's decision to increase energy security and diversity by encouraging nuclear energy generation (as laid out in the US government's Advanced Energy Initiative in 2005) has rekindled much hope for the revival of the nuclear power industry in the United States, and as a matter of fact, the Nuclear Renaissance has already begun - the US Nuclear Regulatory Commission (NRC) approved an early site permit application for the Clinton Power Station in Illinois (Exelon Power Corporation) in March 2007. As the scope of the nuclear energy is expanded, the role of materials is at the front and center. Recent (2011) accidents in Japan due to earthquakes and tsunami are now pointing toward further safeguards and development of more resistant materials. Thus, this book is devoted to addressing various important fundamental and application aspects of materials that are used in nuclear reactors.

# 1.2 Types of Nuclear Energy

Nuclear energy can be derived from many forms such as nuclear fission energy, fusion energy, and radioisotopic energy.

## 1.2.1

## Nuclear Fission Energy

The essence of nuclear fission energy is that the heat produced by the splitting of heavy radioactive atoms (nuclear fission) during the chain reaction is used to generate steam (or other process fluid) that helps rotate the steam turbine generator, thus producing electricity. Nuclear fission energy is the most common mode of producing the bulk of the nuclear energy.

## 1.2.2

## Nuclear Fusion Energy

A huge amount of energy (much higher than fission) can be produced using the nuclear fusion reaction (deuterium-tritium reaction). There is currently no commercial fusion reactors and is not envisioned to be set up for many years. A prototype fusion reactor known as ITER (International Thermonuclear Experimental Reactor) is being built in France and scheduled to produce the first plasma by 2018.

# 1.2.3 Radioisotopic Energy

Either radioactive isotopes (e.g., <sup>238</sup>Pu, <sup>210</sup>Po) or radioactive fission products (e.g., <sup>85</sup>Kr, <sup>90</sup>Sr) can produce decay heat that can be utilized to produce electric power. These types of power sources are mainly used in remote space applications.

# 1.3 Neutron Classification

Chadwick discovered neutron in 1932. Generally, neutrons are generated during radioactive chain reactions in a power reactor. Neutron is subatomic particle present in almost all nuclides (except normal hydrogen isotope or protium) with a mass of  $1.67 \times 10^{-27}$  kg and has no electrical charge.

Neutrons are classified based on their kinetic energies. Although there is no clear boundary between the categories, the following limits can be used as a useful guideline:

Cold neutrons (<0.003 eV), slow (thermal) neutrons (0.003–0.4 eV), slow (epithermal) neutrons (0.4–100 eV), intermediate neutrons (100 eV–200 keV), fast neutrons (200 keV–10 MeV), high-energy (relativistic) neutrons (>10 MeV). *Note:*  $1 \text{ eV} = 1.6 \times 10^{-19} \text{ J}.$ 

Generally, thermal neutrons are associated with a kinetic energy of 0.025 eV that translates into a neutron speed of  $2200 \text{ m s}^{-1}$ !

# 1.4

### Neutron Sources

Various radiation types are produced in a nuclear environment. It could be alpha particles, beta particles, gamma rays, or neutrons. In this book, we are primarily concerned with the radiation damage and effects caused by neutrons. There could be several sources of neutrons, including alpha particle-induced fission, spontaneous fission, neutron-induced fission, accelerator-based sources, spallation neutron source, photoneutron source, and nuclear fusion.

# 1.5 Interactions of Neutrons with Matter

Collision of neutrons with atom nuclei may lead to different scenarios – scattering of the neutrons and recoil of nuclei with conservation of momentum (elastic scattering) or loss of kinetic energy of the neutron resulting in gamma radiation (inelastic scattering). The capture of neutrons may result in the formation of new

nuclei (transmutation), or may lead to the fragmentation of the nucleus (fission) or the emission of other nuclear particles from the nucleus. We shall discuss some of the effects in more detail in Chapter 3.

a) Elastic Scattering

Elastic scattering refers to a neutron–nucleus event in which the kinetic energy and momentum are conserved.

b) Inelastic Scattering

This interaction refers to neutron–nuclide interaction event when the kinetic energy is not conserved, while momentum is conserved.

c) Transmutation

When a nuclide captures neutrons, one result could be the start of a sequence of events that could lead to the formation of new nuclide. The true examples of this type of reaction are  $(n, \alpha)$ , (n, p),  $(n, \beta^+)$ ,  $(n, \beta^-)$ , and (n, f). Reactions like  $(n, \gamma)$  and (n, 2n) do not result in new elements, but only produce isotopes of the original nuclide.

### d) Fission

Fission is a case of (n, f) reaction, a special case of transmutation reaction. Uranium is the most important nuclear fuel. The natural uranium contains about  $0.7\% \text{ U}^{235}$ , 99.3%  $\text{U}^{238}$ , and a trace amount of  $\text{U}^{234}$ . Here, we discuss the neutron-induced nuclear fission, which is perhaps the most significant nuclear reaction. When a slow (thermal) neutron gets absorbed by a U<sup>235</sup> atom, it leads to the formation of an unstable radionuclide U<sup>236</sup>, which acts like an unstable oscillating droplet, immediately followed by the creation of two smaller atoms known as fission fragments (not necessarily of equal mass). About 2.5 neutrons on average are also released per fission reaction of U<sup>235</sup>. An average energy of 193.5 MeV is liberated. A bulk of the energy ( $\sim$ 160 MeV or  $\sim$ 83%) is carried out by the fission fragments, while the rest by the emitted neutrons, gamma rays, and eventual radioactive decay of fission products. Fission fragments rarely move more than 0.0127 mm from the fission point and most of the kinetic energy is transformed to heat in the process. As all of these newly formed particles (mostly fission fragments) collide with the atoms in the surroundings, the kinetic energy is converted to heat. The fission reaction of U<sup>235</sup> can occur in 30 different ways leading to the possibility of 60 different kinds of fission fragments. A generally accepted equation for a fission reaction is given below.

$$U_{92}^{235} + n_0^1 \to Kr_{36}^{92} + Ba_{56}^{142} + 2n_0^1 + \text{Energy}, \tag{1.1}$$

which represents the fission of one  $U^{235}$  atom by a thermal neutron resulting into the fission products (Kr and Ba) with an average release of two neutrons and an average amount of energy (see above). It is clear from the atomic masses of the reactant and products, that a small amount of mass is converted into an equivalent energy following Einstein's famous equation  $E = mc^2$ .

 $U^{235}$  is the one and only naturally occurring radioisotope (fissile atom) in which fission can be induced by thermal neutrons. There are two other fissile atoms (Pu<sup>239</sup> and U<sup>233</sup>) that are not naturally occurring. They are created during

the neutron absorption reactions of U<sup>238</sup> and Th<sup>232</sup>, respectively. Each event consists of  $(n, \gamma)$  reactions followed by beta decays. Examples are shown below:

$$U_{92}^{238} + n_0^1 \to U_{92}^{239} + \gamma \tag{1.2a}$$

$$U_{92}^{239} \to Np_{93}^{239} + \beta^{-}, \quad t_{1/2} = 23.5 \text{ min}$$
 (1.2b)

$$Np_{93}^{239} \rightarrow Pu_{94}^{239} + \beta^-, \quad t_{1/2} = 23.5 \text{ days}$$
 (1.2c)

$$Th_{90}^{232} + n_0^1 \to Th_{90}^{233} + \gamma \tag{1.3a}$$

$$Th_{90}^{233} \to Pa_{91}^{233} + \beta^-, \quad t_{1/2} = 22.4 \text{ min}$$
 (1.3b)

$$Pa_{91}^{233} \to U_{92}^{233} + \beta^-, \quad t_{1/2} = 27.0 \text{ days}$$
 (1.3c)

The concept of the "breeder" reactors is based on the preceding nuclear reactions, and U<sup>238</sup> and Th<sup>232</sup> are known as "fertile" atoms. Heavy radioisotopes such as Th<sup>232</sup>, U<sup>238</sup>, and Np<sup>237</sup> can also undergo neutron-induced fission, however, only by fast neutrons with energy in excess of 1 MeV. That is why these radionuclides are sometimes referred to as "fissionable."

### 1.5.1 **Fission Chain Reaction**

As the preceding section on fission emphasized, each fission reaction of an U<sup>235</sup> atom leads to the release of an average of 2.5 neutrons. Hence, to sustain a continuous fission reaction (i.e., a chain reaction), these neutrons should be able to initiate the fission of at least another fissile atom. Note the schematic of a chain reaction involving  $U^{235}$  atoms in Figure 1.1. A majority of the neutrons (~99.25%) produced due to the fission reaction of U<sup>235</sup>, known as prompt neutrons, are released instantaneously (within  $10^{-14}$  s). But there are about 0.75% neutrons that are released over

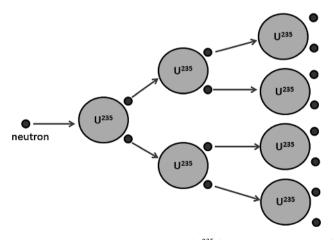


Figure 1.1 A schematic chain reaction of U<sup>235</sup> fissile atoms in progress (for the sake of simplicity, it is assumed that two neutrons are released due to the fission of one U<sup>235</sup> atom and fission fragments created in each fission are also not shown).

a longer period (over  ${\sim}20\,s)$  and these neutrons are called *delayed neutrons*. These delayed neutrons play a very important role in controlling the fission chain reaction.

There is always a competition for neutrons between various processes, namely, (i) fission reaction of fissile atom nuclei, (ii) nonfission capture of neutrons by uranium and other reactions, (iii) nonfission capture of neutrons by other components in the reactor core, and (iv) leakage of neutrons from the core. The reaction can be termed as a chain reaction when the number of neutrons consumed in the processes (ii)-(iv) is at least equal to or less than that consumed in the process (i). Thus, neutron economy plays a very important role in the design of a nuclear reactor. The need for a favorable neutron economy necessitates certain conditions to be met by a chain reacting system. For a given geometry, there is a certain minimum size of a chain reacting system, called the *critical size* (in terms of volume), for which the production of neutrons by fission just balances the loss of neutrons by leakage and so on, and the chain reaction can be sustained independently. The mass corresponding to the critical size is called critical mass. Dependent on the relative generation of fission neutrons and their loss, the reactor is said to be in different stages: subcritical (neutron loss more than the production), critical (balance between the neutron production and loss,  $\kappa = 1$ ), or supercritical (the neutron production is more than the loss,  $\kappa > 1$ ). The multiplication factor  $\kappa$  is often used to express the criticality condition of a reactor. This factor is basically the net number of neutrons per initial neutron.

#### 1.6

#### **Definition of Neutron Flux and Fluence**

The concept of neutron flux is very similar to heat flux or electromagnetic flux. Neutron flux ( $\phi$ ) is simply defined as the density of neutrons *n* (i.e., number of neutrons per unit volume) multiplied by the velocity of neutrons *v*. Hence, *nv* represents the *neutron flux*, which is the number of neutrons passing through a unit cross-sectional area per second perpendicular to the neutron beam direction. However, sometimes this is called *current* if we consider that neutrons moving in one direction only. Neutron flux, in general terms, should take into account all the neutrons moving in all directions and be defined as the number of neutrons crossing a sphere of unit projected area per second. Total neutron flux ( $\phi$ ) is expressed by the following integral:

$$\phi = \int_0^\infty \phi(E_i) \mathrm{d}E_i,\tag{1.4}$$

where  $\phi(E_i) dE_i$  is the flux of neutrons with energies between  $E_i$  and  $E_i + dE_i$ .

The term "*nvt*" represents the neutron fluence, that is, neutrons per unit crosssectional area over a specified period of time (here *t*). Thus, the units of neutron flux and fluence are n cm<sup>-2</sup> s<sup>-1</sup> and n cm<sup>-2</sup>, respectively.

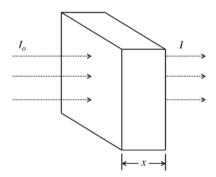
## 1.7 Neutron Cross Section

We have already discussed different ways in which a neutron can interact with nuclides (or specifically nuclei). This is indeed a probabilistic event that depends on the energy of the incident neutrons and the type of nuclei involved in the interaction. Therefore, one can define this probability of interactions in terms of *cross section* that is a measure of the degree to which a particular material will interact with neutrons of a particular energy. But remember that the neutron cross section for a particular element has nothing to do with the actual physical size of the atoms. The range of neutrons (the distance traveled by the neutron before being stopped) is a function of the neutron energy (recall the classification based on neutron energy) as well as the capture cross section of the medium/material through which the neutrons traverse.

To understand it easily, one may consider a simple case shown in Figure 1.2 with a beam of neutrons impinging on a material of unit area (in cm<sup>2</sup>) and thickness x (in cm). Thus, the intensity of neutrons traveling beyond the material will be diminished depending on the number of nuclei per unit volume of the material (n') and the "effective area of obstruction" (in cm<sup>2</sup>) presented by a single nucleus. This area of obstruction is generally called "microscopic cross section" ( $\sigma$ ) of the material. Like any other absorption equation, one can write

 $I = I_0 \exp\left(-n'\sigma x\right),\tag{1.5}$ 

where the quantity  $n'\sigma$  is called the macroscopic cross section or obstruction coefficient  $\sum$  (unit in cm<sup>-1</sup>). This represents the overall effect of nuclei (n') in the neuron beam path and the power of the nuclei to take part in the interaction. Equation (1.5) is specifically for only one type of reaction when the absorber material contains only one type of pure nuclide. But actually the material could consist of several types of nuclei, and in that case, we should add all the neutron cross sections for all possible reactions to obtain the total neutron cross section.



**Figure 1.2** Attenuation of an incident neutron beam of intensity  $I_0$  by an absorber material.

From Eq. (1.5), a half-value thickness ( $x_{1/2}$ ) for neutron beam attenuation can be derived using the following relation:

$$x_{1/2} = 0.693/(n' \cdot \sigma). \tag{1.6}$$

The values of neutron microscopic cross section ( $\sigma$ ) are typically between  $10^{-22}$  and  $10^{-26}$  cm<sup>2</sup>, leading to the development of a convenient unit called *barn* (1 barn =  $10^{-24}$  cm<sup>2</sup>).

*Note*: Macroscopic neutron cross section ( $\sum$ ) can be calculated with the knowledge of n' that can be calculated from the following relation:

$$n' = (\rho/M) \times (6.023 \times 10^{23}), \tag{1.7}$$

where  $\rho$  is the density (g cm<sup>-3</sup>) and *M* is the atomic weight of the element.

Thus, 
$$\sum = (\varrho/M) \times (6.023 \times 10^{23})\sigma.$$
 (1.8)

In order of increasing cross section for absorption of thermal neutrons, various metals can be classified as follows (normalized to Be):

We will see in later chapters that neutron capture cross section has a significant role to play in the selection of reactor materials coupled with other considerations. See Table 1.1 for some representative values of neutron capture cross sections for several important nuclides.

Most nuclides exhibit both the  $1/\nu$  ( $\nu$  = velocity of neutron) dependence of neutron cross section and the resonance effects over the entire possible neutron energy spectrum. We should not forget that the neutron cross sections heavily depend on the type of reactions they take part in, such as alpha particle producing reaction ( $\sigma_{\alpha}$ ), fission reactions ( $\sigma_{f}$ ), neutron capture cross sections ( $\sigma_{c}$ ), and so on. As discussed above, the total cross section ( $\sigma_{t}$ ) is a linear summation of all neutronic reactions possible at the specific neutron energy level.

 Table 1.1
 Neutron cross sections (in barn) for capture of thermal neutrons (i.e., of average kinetic energy 0.025 eV) of a few nuclides.

Nuclide	Neutron cross section (b)
1 <sub>1</sub> H	0.332
<sup>1</sup> H <sup>2</sup> H <sup>12</sup> C <sup>238</sup> 9U <sup>235</sup> 92U	0.00052
<sup>12</sup> <sub>6</sub> C	0.0035
<sup>238</sup> <sub>92</sub> U	2.7
<sup>235</sup> <sub>92</sub> U	586

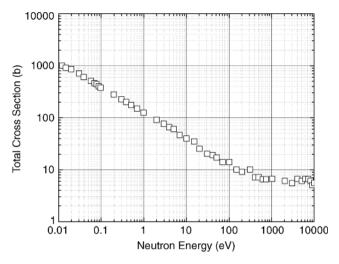
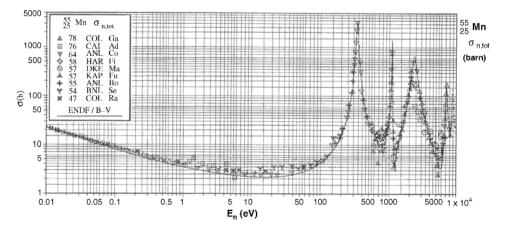


Figure 1.3 Variation of total cross section of elemental boron as a function of neutron energy.

The inverse proportionality of total neutron cross section in elemental boron as a function of neutron energy is shown in Figure 1.3. However, this is not always the case. Many nuclides show abrupt increases in neutron cross section at certain narrow energy ranges due to resonance effects, which happens when the energy of the incident neutron corresponds to the quantum state of the excited compound nucleus. An example of such a situation for Mn-55 is shown in Figure 1.4.

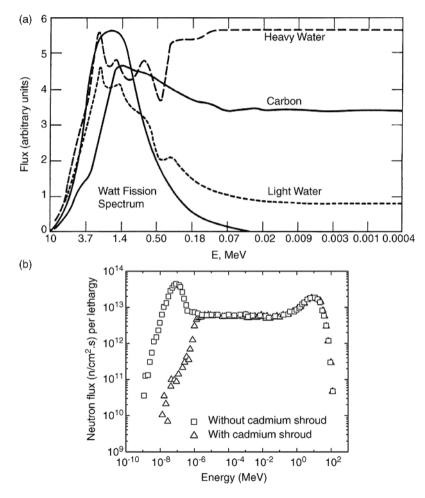


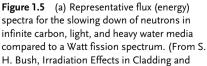
**Figure 1.4** Total cross section curve for Mn-55 over the neutron energy range of 0.01 eV–10 keV. (From M.F.L' Annunziata, Handbook of Radioactivity Analysis, Academic Press, New York, 1998; with permission.)

## 1.7.1

## **Reactor Flux Spectrum**

The neutron energy spectrum is affected by various factors, including reactor type, position in the reactor, and immediate surroundings, such as adjacent fuel, control rods, and empty surroundings. The overall shape of the neutron spectrum is influenced by the specific type of reactor. For reactors using moderators, such as heavy water, light water, or graphite, Figure 1.5a depicts idealized curves for a normalized flux of neutrons as a function of neutron energy.





Structural Materials, Rowman & Littlefield Inc., New York, 1965; with permission.) (b) The reactor flux spectra in the ATR with and without cadmium shroud. The neutron fission spectrum calculated by Watt is also superimposed on the graph for comparison. Convenient techniques such as assuming monoenergetic neutron flux and the arbitrary selection of neutron flux cutoff level (>1 MeV) are mostly general approximations. Remember that most neutron fluxes cited at irradiation damage studies are expressed in terms of >1 MeV. Figure 1.5b depicts the two flux spectra obtained from the Advanced Test Reactor (ATR). One spectrum is without the use of cadmium shroud and another one is with the cadmium shroud (of ~1.14 mm thickness). It is clear that fast (hard) spectrum is achieved with the use of cadmium shroud (i.e., irradiation jig wrapped into cadmium foil) due to its absorption of thermal neutrons, but not fast ones. Dosimetric experiments followed by calculations can generate the flux spectrum for a specific position in the reactor.

## 1.8 Types of Reactors

Even though a nuclear reactor can be defined in different ways, almost all reactors except fusion reactor (commercially nonexistent) can be defined as follows: "A nuclear reactor is a device, designed to produce and sustain a long term, controlled fission chain reaction, and made with carefully selected and strategically placed collection of various materials." The classification of reactors vary and are generally based on the following: type of fission reaction (thermal, epithermal, and fast reactors), purpose of the reactor (power reactors, research reactors, and test reactors), type of the coolant present (such as light/heavy water reactors, gas-cooled reactors, and liquid metal-cooled reactors), type of core construction (cubical, cylindrical, octagonal, and spherical reactors), and so forth.

## 1.8.1

## A Simple Reactor Design

Almost all the reactors in the United States and a majority in the world are thermal reactors wherein *thermal neutrons* cause the bulk of the fission reactions. If one starts to think about designing a prototype reactor, the several design elements need to be flawlessly integrated. Figure 1.6 shows such a schematic for a primitive thermal reactor. The tubes containing fuels are generally made of metallic alloys (also known as fuel cladding). The radioactive fuels (such as uranium) could be in metallic, alloy, or ceramic forms. The fuel cladding serves many purposes: it provides mechanical support to the fuel, keeps the fission products from leaving the fuel element, and protects the fuels from corrosion from the coolant. The fuel elements are arranged in a distinct regular pattern (square, hexagon, etc., dictated by neutronics and other factors) with the moderator. Moderator slows down the neutron to sustain the fission reaction with thermal neutrons. The fuel–moderator assembly is surrounded by a

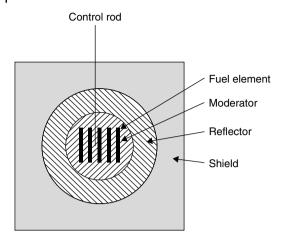


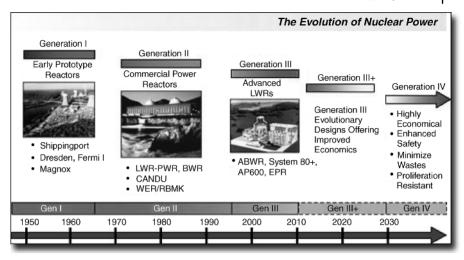
Figure 1.6 A schematic of a simple reactor design. (adapted from C.O. Smith, Nuclear Reactor Materials, Addison-Wesley, Reading, MA, 1967)

reflector. The purpose of a *reflector* is to direct all neutrons generated toward the core so that neutron leakage can be controlled, thus improving the neutron economy. On the outside, the reactor is lined by shielding materials that absorb neutrons and gamma rays that escape the core and reduce the radiation intensity to a tolerable level so that people near the reactor are not exposed to these radiations. The control rod (usually an assembly) helps control the chain reaction by absorbing neutrons, maintaining the steady state of operation. Hence, the control materials are neutron-absorbing materials (boron, hafnium, and so forth), and are generally fabricated in the form of rods (in some cases, plates). A reactor is typically equipped with two types of control rods - regulating rods for routine control reasons and safety rods (to permit shutdown in the case of emergency). Even though coolant is not shown in Figure 1.6, it is an important component of a reactor. As a huge amount of heat is generated in the fuel elements, the heat needs to be removed continuously in an efficient manner in order to maintain a safe, steady-state reactor operation. This means an efficient coolant is needed. The coolant can be a gas or liquid (such as light or heavy water, carbon dioxide, liquid metals, and molten salts). However, it is important to remember that the presence of any coolant tends to adversely affect the neutron economy. Hence, the balance between the reduction in the neutron economy and the efficiency of heat removal needs to be carefully considered.

### 1.8.2

#### **Examples of Nuclear Reactors**

A good point to start the discussion on various examples of nuclear reactors is to understand the evolution of nuclear power over the past six to seven



**Figure 1.7** Nuclear power evolution in the world. *Courtesy:* The US Department of Energy Gen-IV Initiative.

decades. As shown in Figure 1.7, there are different generations of reactors. Thus, a chronological approach has been taken to start an overview of various nuclear power reactors. However, it is, by no means, exhaustive account. The first nuclear reactor known as Chicago Pile 1 (CP-1) was built during the heydays of the World War II (criticality was achieved on December 2, 1942) at the University of Chicago, Chicago, IL. It was designed and built by a team led by Enrico Fermi. This reactor was a thermal reactor with graphite moderators and natural uranium dioxide fuel. No coolant or shielding was used. The reactor could produce only  $\sim$ 200 W of heat. However, the primary aim of the reactor was to demonstrate the occurrence of the fission chain reaction. It was dismantled in February 1943, and CP-2 reactor was installed at the Argonne National Laboratory based on the experience gained with CP-1.

### 1.8.2.1 Generation-I Reactors

Generation-I reactors were built in the initial period of nuclear power expansion and generally had primitive design features. Most of these reactors have either been shut down or will be soon done so. Examples of such reactors are Magnox reactor (Calder Hall reactor in the United Kingdom) and first commercial power reactor at Shippingport in 1957 (in the state of Pennsylvania in the United States).

### Magnox Reactor

This is a notable Generation-I gas-cooled reactor. Early breed of this reactor was used for the purpose of plutonium production (for atomic weapons) as well as electricity generation. Figure 1.8a shows a cross section of a typical Magnox reactor. The

Calder Hall station in the United Kingdom was a Magnox type of reactor starting successful operation in 1956. Following that, several of these reactors were built and operated in the United Kingdom and a few elsewhere (e.g., Italy, France, and Japan). Generally, Magnox reactors were graphite moderated, and used the natural uranium as fuel clad in thin cylindrical tubes of a magnesium alloy (Magnox comes from the name of the magnesium-based alloy with a small amount of aluminum and other minor elements, magnesium nonoxidizing, for example, Mg–0.8Al–0.005Be) and carbon dioxide (CO<sub>2</sub>) as coolant (heat transfer medium). Magnesium-based alloy was chosen since Mg has a very low thermal neutron capture cross section (0.059 b; lower than Zr or Al). The fuel elements were impact extruded with the integral cooling fins or machined from finned extrusions (Figure 1.8b). Also,

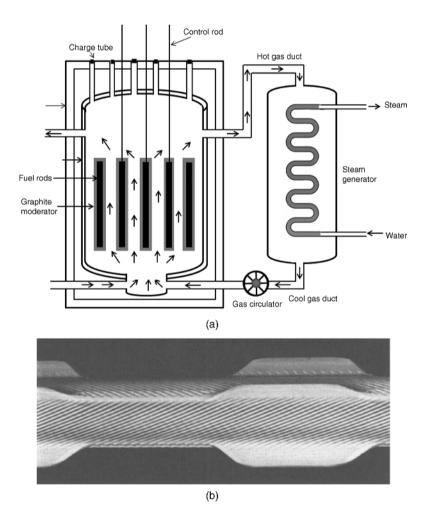


Figure 1.8 (a) A schematic of a Magnox reactor. (b) A part of the magnesium alloy fuel can of a British Magnox reactor. *Courtesy:* Light Alloys by Ian Polmear.

the alloy was resistant to creep and corrosion from CO<sub>2</sub> atmosphere in the operating temperature range, and contrary to Al, the alloy did not react with the uranium fuel. The addition of Al in the alloy provided solid solution strengthening, while the presence of minor amounts of Be helped improve the oxidation resistance. CO<sub>2</sub> was circulated under pressure through the reactor core and sent to the steam generator to produce steam that is then passed through a turbo generator system generating electricity. These reactors could sustain lower temperatures (maximum coolant temperature of 345 °C) and, thus, has a limited plant efficiency and power capacity. This was mainly out of the concern of the possible reaction of CO2 with graphite at higher temperatures and the lower melting point of uranium fuel (1132 °C). Another problem was that the spent fuel from these reactors could not be safely stored under water because of its chemical reactivity in the presence of water. Thus, the spent fuels needed to be reprocessed immediately after taking out of the reactor and expensive handling of equipment was required. Only two Magnox reactors still operating in the United Kingdom are scheduled to be decommissioned soon. Magnox reactors were followed by an improved version of gas-cooled reactor known as Advanced Gas-Cooled Reactor (AGR) operating at higher temperatures and thus improving the plant efficiency. The magnesium fuel element was replaced by stainless steels.

### 1.8.2.2 Generation-II Reactors

Most of the commercial nuclear power plants operating today are of Generation-II type. Also, the reactors employed in naval vessels (such as aircraft carriers and submarines) and many research/test reactors are of this type. The Generation-II reactors incorporated improved design and safety features and productivity over Generation-I reactors. In the Western Hemisphere, a majority of commercial nuclear power plants have light water reactor (LWR), both pressurized water reactor (PWR) and boiling water reactor (BWR). It is important to remember that LWRs were also built as Generation-I reactors (such as Shippingport facility with 60 MWe power capacity), however most of them are no longer in operation. Another variety is the CANDU (Canadian Deuterium Uranium) reactor, which is basically a pressurized heavy water reactor (PHWR). There are a few different versions of pressurized water reactors (e.g., RBMK type) in Russia and former Soviet-block countries, but discussion on those reactors is outside the scope of this book.

#### **Light Water Reactors**

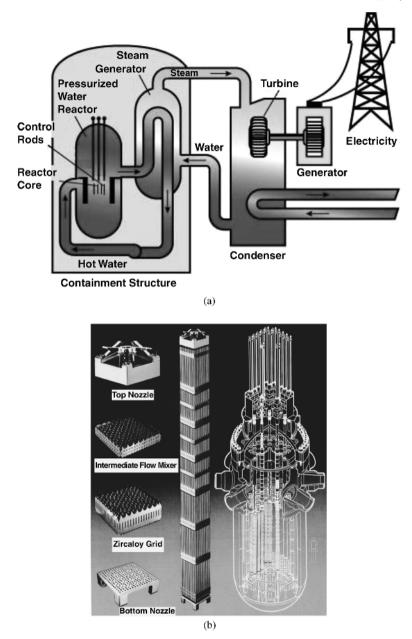
As the name implies, LWRs use light water as the coolant and the moderator, and in many cases as the reflector material. These are typical thermal reactors as they utilize thermalized neutrons to cause nuclear fission reaction of the  $U^{235}$  atoms. The thermal efficiency of these reactors hover around 30%. Two main types of LWR are PWR and BWR. These two types are created mainly because of the difference in approaches of the steam generating process (good quality steam should not contain more than 0.2% of condensed water). LWRs have routinely been designed with 1000 MWe capacity.

**Pressurized Water Reactor** PWRs were designed and implemented commercially much sooner than the BWRs due to the earlier notion that the pressurized liquid water would somehow be much safer to handle than the steam in the reactor core and would add to the stability of the core during the operation. That is why the first commercial reactor in Shippingport was a PWR. PWRs are designed and installed by companies such as Westinghouse and Areva.

A schematic design of a typical PWR plant is shown in Figure 1.9a. A PWR plant consists of two separate light water (coolant) loops, primary and secondary. The PWR core is located inside a reactor pressure vessel (RPV) made of a low-alloy ferritic steel (SA533 Gr. B) shell (typical dimensions: outside diameter  $\sim$ 5 m, height  $\sim$ 12 m, and wall thickness 30 cm), which is internally lined by a reactor cladding of 308-type stainless steel or Inconel 617 to provide adequate corrosion resistance against coolant in contact with the RPV. The PWR primary loop works at an average pressure of 15–16 MPa with the help of a set of pressurizers so that the water does not boil even at temperatures of 320-350 °C. The PWR core contains an array of fuel elements with stacks of a slightly enriched (2.5-4%) UO<sub>2</sub> fuel pellets clad in Zircaloy-4 alloy (new alloy is Zirlo or M5). Individual cladding tubes are generally about  $\sim 10 \,\mathrm{mm}$  in outer diameter and  $\sim 0.7 \,\mathrm{mm}$  in thickness. The fuel cladding tube stacked with fuel pellets inside and sealed from outside is called a fuel rod or fuel pin. About 200 of such fuel rods are bundled together to form a fuel element. Then, about 180 of such fuel elements are grouped together to form an array to create the reactor core of various shapes - square, cylindrical, hexagonal, and so on (Figure 1.9b). The reactor core is mounted on a core-support structure inside the RPV. Depending on the specific design, the above-mentioned dimensions of the various reactors may vary.

The control rod used is typically an Ag–In–Cd alloy or a  $B_4C$  compound, which is used for rapid control (start-up or shutdown). Boric acid is also added to the primary loop water to control both the water chemistry acting as "poison" and the long-term reactivity changes. This primary loop water is transported to the steam generator where the heat is transferred to the secondary loop system forming steam. The steam generator is basically a heat exchanger containing thousands of tubes made from a nickel-bearing alloy (Incoloy 800) or nickel-based superalloy (e.g., Inconel 600) supported by carbon steel plates (SA515 Gr.60).

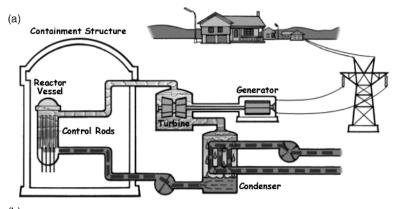
**Boiling Water Reactor** BWR design embodies a direct cycle system of cooling, that is, only one water loop, and hence no steam generator (Figure 1.10a). Early boiling water experiments (BORAX I, II, III, etc.) and development of experimental boiling water reactor (EBWR) at the Argonne National Laboratory were the basis of the future commercial BWR power plants. Dresden Power Station (200 MWe), located at the south of Chicago, IL, was a BWR power plant that started operating in 1960. It is of note that this was a Generation-I BWR reactor. However, most BWRs operating today are of Generation-II type and most significant features are discussed below. The reactors (Fukushima Daiichi) that underwent core melting following the unprecedented earthquake and tsunami in Japan during 2011 were all of BWR type.



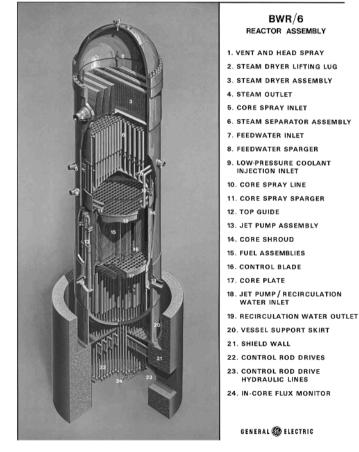
**Figure 1.9** (a) A view of a typical PWR plant. Courtesy: US Nuclear Regulatory Commission (b) A view of various PWR components. Courtesy: Westinghouse Electric Corporation.

The BWR reactor core is located near the bottom end of the reactor pressure vessel. Details of various components in a typical BWR are shown in Figure 1.10b. The BWR RPV is more or less similar to the PWR one. The BWR core is made of fuel assembly consisting of slightly enriched  $UO_2$  fuels clad with recrystallized Zircaloy-

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(b)



**Figure 1.10** (a) A view of a BWR plant. (b) A view of the cut section of a typical BWR. *Courtesy:* The US Nuclear Regulatory Commission and GE.

PWR	BWR
Principle of steam generation	
RPV pressure $\sim 15$ MPa	RPV pressure ${\sim}7{ m MPa}$
RPV temperature $\sim$ 326 °C	RPV temperature ~290 °C
Steam generated in steam generator (via	Steam generated in RPV (with
secondary loop)	separator and dryer)
No bulk boiling in RPV	Bulk boiling allowed in RPV
Major components	U U
RPV	RPV with separator and dryer
Two–four steam generators	No steam generator
One pressurizer	No pressurizer
Top entry control rod clusters	Bottom entry control rod drives
Zircaloy-4 fuel cladding tubes	Zircaloy-2 fuel cladding tubes

Table 1.2 PWR versus BWR.

2 cladding tubes (about 12.5 mm in outer diameter). For a BWR core of 8 × 8 type, each fuel assembly contains about 62 fuel rods and 2 water rods, which are sealed in a Zircaloy-2 *channel box*. The control material is in the shape of blades arranged through the fuel assembly in the form of a cruciform and is generally made of B<sub>4</sub>C dispersed in 304-type stainless steel matrix or hafnium, or a combination of both. Water is passed through the reactor core producing high-quality steam and dried at the top of the reactor vessel. The BWR operates at a pressure of about 7 MPa and the normal steam temperature is 290–330 °C.

#### Note

Tables 1.2 and 1.3 contain relevant information on PWRs and various types of BWRs.

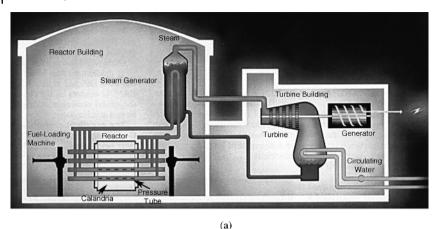
Parameter/feature	BWR (Browns Ferry 3)	ABWR	ESBWR <sup>a)</sup>
Power (MWt/MWe)	3293/1098	3926/1350	4500/1590
Vessel height/diameter (m)	21.9/6.4	21.7/7.1	27.6/7.1
Fuel bundles (number)	764	872	1132
Active fuel height (m)	3.7	3.7	3.0
Recirculation pumps (number)	2 (Large)	10	Zero
Number of control drive rods	185	205	269
Safety diesel generator	2	3	Zero
Safety system pumps	9	18	Zero
Safety building volume	120	180	135

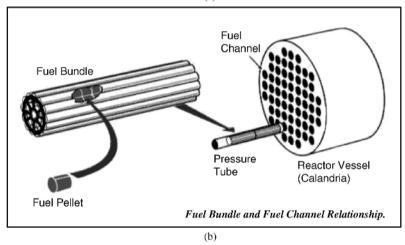
Table 1.3 Operating parameters and design features of BWRs.

Courtesy: GE Global Research.

a) ESBWR – economic simplified boiling water reactor – of Generation-III+ category, developed by the GE.

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**Figure 1.11** (a) A simplified schematic view of a CANDU reactor. *Courtesy:* Canadian Nuclear Association. (b) The configuration of fuel bundles in the fuel channel. *Courtesy:* www.cameco.com.

**Pressurized Heavy Water Reactor** The PHWR reactors were mainly developed by a partnership between the Atomic Energy of Canada Limited (AECL) and Hydro-Electric Power Commission of Ontario in 1960s. The reactors were of Generation-II type. Notably, these reactors are also called CANDU reactors (Figure 1.11a). They are so named because they use heavy water (deuterium oxide) as the moderator and natural uranium as the fuel. These reactors are located mainly in Canada, India, China, and few other countries. The CANDU reactor design does not require a reactor pressure vessel as in LWRs, and hence not a single CANDU reactor operates in the United States since the nuclear safety regulations of the US Nuclear Regulatory Commission specifically call for an RPV in a compliant reactor design.

Unlike LWRs, the natural uranium (0.7% U<sup>235</sup>) oxide fuel clad in zirconium alloy tubes (known as pressure tubes, made of Zr-2.5Nb) is used in this reactor. These hundreds of pressure tubes are kept inside a calandria shell made of an austenitic stainless steel and reinforced by outer stiffening rings. The shell also keeps channels for the pressurized coolant (hot heavy water or light water) and moderator (heavy water). If light water is used to moderate the neutrons, it would adversely affect the neutron economy due to the absorption of neutrons. That is why cold heavy water is used as the moderator. The pressure tubes along with moderator and cooling tubes are arranged in a horizontal fashion (not vertical as in LWRs) and the fuels can be replaced and reloaded without shutting down the whole reactor. Note the fuel and associated structural configuration in Figure 1.11b. The pressurized coolant stays typically at about 290 °C. This reactor system requires a steam generator to produce steam as does a conventional PWR. The control rods are dropped vertically into the fuel zones in case total shutdown or controlling of reactivity is necessary. Gadolinium nitrate is added in the moderator system that acts as a secondary shutdown system.

Liquid Metal Fast Breeder Reactor Liquid metal (generally sodium) is used in liquid metal fast breeder reactors (LMFBRs) to transport the heat generated in the core. These reactors are called "breeder" because more new fuels are produced than consumed during its operation. The reactor can convert fertile material (containing U<sup>238</sup> and Th<sup>232</sup>) into fissile materials (Pu<sup>239</sup> and U<sup>233</sup>), respectively. The concept of this reactor type is very practical from the fuel utilization viewpoint. The natural uranium contains only about 0.7% of fissile U<sup>235</sup>. The majority of the natural uranium contains U<sup>238</sup> isotope. These reactors are characterized by high power density (i.e., power per unit volume) due to the lack of a moderator (i.e., much more improved neutron economy). The reactor cores are typically small because of the high power density requirements. The temperature attained in this type of reactors is higher and thus leads to higher efficiency of electric power generation (~42% in LMFBRs versus  $\sim$ 30% in LWRs). The use of Th<sup>232</sup> in LMFBRs is particularly advantageous for countries like India that do not have a large deposit of uranium, but has plenty of thorium. It should be noted that sodium used in this type of reactor transfers heat to the steam generators. The system containing sodium should be leak-proof since sodium reacts with oxygen and water vapor very fast. Furthermore, it becomes radioactive as the coolant passes through the reactor core. The whole primary coolant system should be put in the shielded protection to keep the operating personnel safe.

The first prototype LMFBR reactor named EBR-I (Experimental Breeder Reactor) was built at the present-day site of the Idaho National Laboratory near Idaho Falls, ID. This was the first reactor to demonstrate that the electricity can be generated using the nuclear energy. Also, it was the first "fast breeder" reactor. It used sodium–potassium (NaK) as coolant. It started its operation in 1951, and was decommissioned by 1964. By this definition, it was a Generation-I fast reactor

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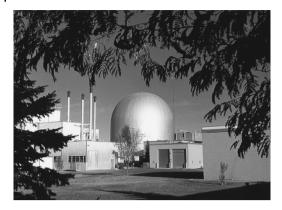


Figure 1.12 A view of EBR-II reactor.

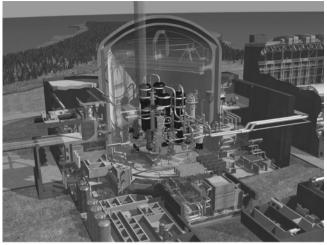
design. Following the decommissioning of EBR-I, another fast breeder reactor (EBR-II) was installed and started operation in 1963. EBR-II (Figure 1.12) was operated very successfully before it was shut down in 1994. The fuel used was a mixture of 48–51% of  $U^{235}$ , 45–48%  $U^{238}$ , and the rest a mixture of fissium metals (Mo, Zr, Ru, etc.) and plutonium.

#### 1.8.2.3 Generation-III and III+ Reactors

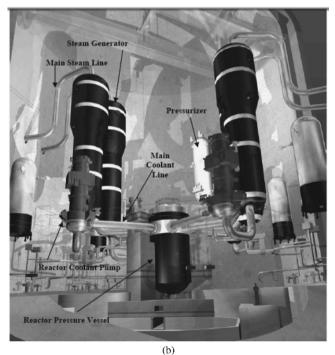
The new reactors that are being built or will be built within a few years are of Generation-III category. These are mainly advanced LWRs. Examples include advanced boiling water reactor (ABWR) and evolutionary or European power reactor (EPR). In the same line, Generation III+ category aims to provide reactor systems that have much improved designs and safety features, and much greater capacities. Notably, all these reactors are thermal in nature. No fast reactor is in the pipeline in these categories. An improved version of the US EPR<sup>®</sup> is being designed and developed as a Generation III+ reactor by AREVA NP. It is a four-loop plant with a rated thermal power of 4590 MW<sub>th</sub>. Figure 1.13a and b shows the details of a EPR power plant.

The primary system design and main components are similar to those of PWRs currently operating in the United States. However, the US EPR incorporates several new advancements in materials technology as well as new uses for existing materials. Some notable examples include M5<sup>(R)</sup> fuel cladding and the stainless steel heavy neutron reflector. Some details of these are discussed below:

a) Zircaloy-4 (Zr-4) has been used extensively for many years in PWR fuel cladding applications. Zr-4 is a cold worked stress-relieved (CWSR) alloy with a zirconium base containing tin, iron, chromium, and oxygen. This offers good corrosion resistance and mechanical properties. AREVA NP has developed an advanced zirconium alloy for PWR fuel rod cladding and fuel assembly structural components, known as M5<sup>®</sup> (Zr-0.8–0.12Nb–0.09–0.13O). M5<sup>®</sup> makes high-burnup fuel cycles possible in the increasingly higher duty operating environments of PWRs. Introduced commercially in the 1990s after a rigorous development program, M5<sup>®</sup> was a breakthrough in zirconium alloy development. This fully recrystallized, ternary Zr–Nb–O alloy produces improved in-reactor corrosion,



(a)



**Figure 1.13** (a) A layout of a US  $EPR^{IB}$  power plant. (b) Main coolant line components in the US  $EPR^{IB}$ . *Courtesy:* Areva NP.

hydrogen, growth, and creep behaviors. The remarkably stable microstructure responsible for these performance improvements is the result of carefully controlled ingot chemistry and product manufacturing parameters. M5<sup>®</sup> helps utilities achieve significant fuel cycle cost savings and enhanced operating margins

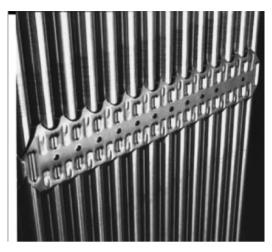


Figure 1.14 A 14  $\times$  14 matrix fuel assembly made of M5<sup>®</sup> alloy. *Courtesy:* Areva NP.

by allowing higher burnups and higher duty cycles. Excellent corrosion resistance and low irradiation growth allow higher burnups, extended fuel cycle operation, and fuel assembly design modifications that enhance operating flexibility. The corrosion resistance of M5<sup>®</sup> allows operation in high pH environments, eliminating the risk of oxide spalling and helping to minimize dose rates. M5<sup>®</sup> has been proven to withstand severe operating conditions: high neutron flux, heat flux, and high operating temperatures. As of January 2007, over 1576000 fuel rods in designs from  $14 \times 14$  (Figure 1.14) to  $18 \times 18$  matrices have been irradiated in commercial PWRs to burnups exceeding 80 000 MWd per mtU. In fuel rod cladding with hydrogen concentrations above the solubility limit, excess hydrogen precipitates as brittle hydrides, reducing the ability of the cladding to cope with pellet-to-clad mechanical interactions during reactivity insertion accidents. During a LOCA, ( Loss Of Coolant Accident) high hydrogen levels increase the transport and solubility of oxygen at high temperatures, leading to substantial embrittlement of the cladding as it cools and the oxygen precipitates. M5<sup>®</sup> cladding absorbs approximately 85% less hydrogen than other Zr-4-based alloys. As a result, M5<sup>®</sup> will not reach hydrogen levels sufficient enough to precipitate hydrides and will not lead to excess oxygen absorption during a LOCA.

b) The space between the multicornered radial periphery of the reactor core and the cylindrical core barrel is filled with an austenitic stainless steel structure, called the *heavy reflector*. The primary purpose of the heavy reflector is to reduce fast neutron leakage and flatten the power distribution of the core, thus improving the neutron utilization. It also serves to reduce the reactor vessel fluence  $(\sim 1 \times 10^{19} \text{ n cm}^{-2}, E > 1 \text{ MeV}$  after 60 years). The reflector is inside the core barrel above the lower core support plate. The reflector consists of stacked forged slabs (rings) positioned one above the other with keys, and axially restrained by tie rods bolted to the lower core.

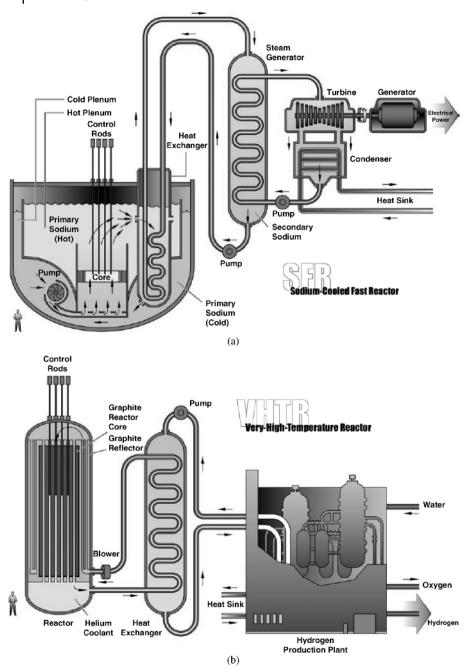
#### 1.8.2.4 Generation-IV Reactors

Gen-IV reactors are the futuristic reactors for which research and development efforts are currently in progress. These reactors will be more efficient, safer, longer lasting (60 years and beyond), proliferation-resistant, and economically viable compared to the present nuclear reactors. Six reactor designs were selected at the outset. They are summarized in Table 1.4 including information on the type (fast or thermal spectrum) of the reactor, coolants, and approximate core outlet temperatures. Two reactor concepts, *sodium fast reactor* (SFR) – along with the *advanced burner reactor* (ABR) concept under the erstwhile Advanced Fuel Cycle Initiative Program) – and *very high-temperature reactor* (VHTR) under the Next Generation Nuclear Plant (NGNP) are of the highest priority in the United States (Figure 1.15a and b, respectively). VHTR is the reactor concept employed for the Next Generation Nuclear Plant program where the heat generated will cogenerate electricity and hydrogen.

The demanding service conditions (higher neutron doses, exposure to higher temperatures, and corrosive environments) that the structural components will experience in these reactors would pose a significant challenge for the structural materials selection and qualification efforts. Because of the stringent requirements noted above, the materials employed in today's commercial reactors are not suitable for use in Gen-IV reactors. For example, zirconium alloys (Zircaloy-2, Zircaloy-4, Zr-2.5Nb, M5) have been used routinely as fuel cladding and other reactor internals in both light and heavy water reactors because of their low neutron capture cross section and acceptable mechanical and corrosion resistance in high temperature

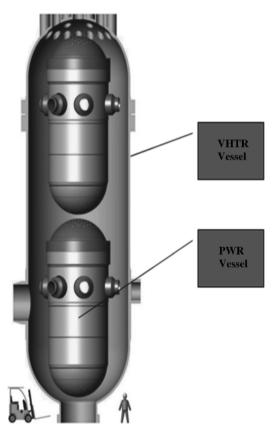
Reactor system	Priority/timeline	Coolant	Neutron spectrum	Core outlet temperature (°C)
Gas-cooled fast reactor (GFR)	Medium/long term	Gas (e.g., He)	Fast	~850
Lead-cooled reactor (LFR)	Medium/long term	Liquid metal (e.g., Pb, Pb–Bi)	Fast	550-800
Molten salt reactor (MSR)	Low/long term	Molten salt (e.g., fluoride salts)	Thermal	700–800
Sodium-cooled fast reactor (SFR)	High/mid term	Liquid metal (Na)	Fast	~550
Very high- temperature reactor (VHTR)	High/mid term	Gas (e.g., He), CO <sub>2</sub>	Thermal	> 900
Supercritical water-cooled reactor (SCWR)	Medium/long term	Water	Thermal/fast	280–620

 Table 1.4
 A summary of Gen-IV reactor system designs.



**Figure 1.15** Schematics of (a) a sodium fast reactor and (b) a very high-temperature reactor. (Courtesy: US Department of Energy Gen-IV Initiative)

(probably never exceeding 350–380 °C under normal service conditions) aqueous environment. However, higher temperatures envisioned in Gen-IV reactors would limit the use of zirconium alloys because of increased susceptibility to hydrogen embrittlement due to severe hydride formation, allotropic phase changes at higher temperatures ( $\alpha$ – $\beta$  phase), poor creep properties, and oxidation. It is instructive to note that some high-performance zirconium alloys may be of possible use in relatively low-temperature Gen-IV reactor design (such as SCWR). Furthermore, the out-of-core components (pressure vessel, piping, etc.) may need to be made from materials other than the low-alloy ferritic steels (e.g., A533B) currently employed primarily because similar components in Gen-IV reactors are expected to withstand much higher temperatures and neutron doses. Some of the fabrication difficulty involved in the VHTR construction demands mention here. For example, the pressure vessel for VHTR reactor as shown in Figure 1.16 is about double the size of the currently operating PWRs. Heavy component forgings will be needed in the construction of these huge pressure vessels.



**Figure 1.16** A schematic size comparison between the reactor pressure vessels in a typical PWR and a conceptual VHTR. *Courtesy:* Nuclear News.

#### **Test Reactors**

So far we have discussed the past, current, and future nuclear reactors in detail. Now let us discuss about a test reactor. One of the well-known test reactors in the United States is the Advanced Test Reactor located at the Idaho National Laboratory near Idaho Falls, ID. ATR is a flagship reactor that serves the US Department of Energy, Naval Nuclear Propulsion Program, and different other governmental and commercial entities. It also acts as a user facility that the university-led teams can use to irradiate materials and perform postirradiation examination (PIE) upon going through a competitive proposal process. ATR started its operation in 1967 and is being operated continuously since then with 250+ days of operation in an average year. A view section of the ATR is shown in Figure 1.17.

ATR is a pressurized, light water cooled and moderated, 250 MW<sub>th</sub> reactor with beryllium reflectors and hafnium control drums. The metallic fuel (U or U–Mo) is in the plate morphology clad in an aluminum alloy. There are 40 fuel assemblies in the reactor core; each core contains 19 fuel plates. At 250 MW, maximum thermal neutron flux is  $\sim 10^{15}$  n cm<sup>-2</sup> s<sup>-1</sup>, and the maximum fast neutron flux could reach  $5 \times 10^{14}$  n cm<sup>-2</sup> s<sup>-1</sup>. Thus, ATR can be used to study the radiation damage under the fast neutron spectrum. The ATR has 77 irradiation positions (4 flux traps, 5 in-pile tubes, and 68 in-reflector) (for details, see Figure 1.18). The reactor pressure vessel is made of stainless steel, and is 3.65 m in diameter and  $\sim 10.67$  m in height. Table 1.5 lists some of the differences between ATR and a typical PWR.

ATR even though used for neutron irradiation experiments is not a fast reactor facility. Note that at present the United States does not have any operating/underconstruction fast reactor test facility (EBR-II and FFTF facilities were shut down during 1990s) as opposed to countries like France (Phenix), India (prototype fast breeder reactor (PFBR)), Japan (Joyo and Monju), Russia (BOR-60), and China (China experimental fast reactor (CEFR). The lack of a fast reactor facility is a challenge for the US nuclear R&D community. The proposed Advanced Burner Test Reactor (ABTR), a sodium-cooled fast reactor, is still under the planning stage, and there is no further confirmation of its installation yet.

# 1.9

## Materials Selection Criteria

"We physicists can dream up and work out all the details of power reactors based on dozens of combinations of the essentials, but it's only a paper reactor until the metallurgist tells us whether it can be built and from what.

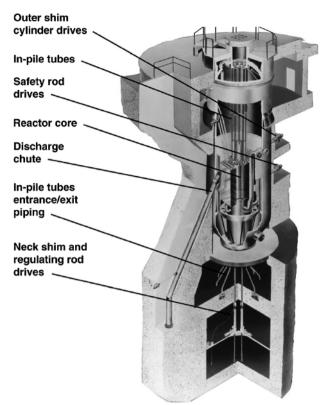


Figure 1.17 A view section of the Advanced Test Reactor. Courtesy: Idaho National Laboratory.

Reactor Features	ATR	PWR
Power (MW <sub>th</sub> )	250 (maximum design)	~3800
Operating pressure (MPa per psig)	~2.5/~355	~15.5/~2235
Inlet temperature (°C)	~52	$\sim 288$
Outlet temperature (°C)	~71	$\sim$ 327
Power density (kW per ft <sup>3</sup> )	$\sim 28\ 300$	$\sim \! 2800$
Fuel element shape	Plate	Tubular
Fuel	Enriched U <sup>235</sup>	3–4% Enriched U <sup>235</sup>
Fuel temperature (°C)	~462	>538

 Table 1.5
 Comparison between ATR and a typical PWR.

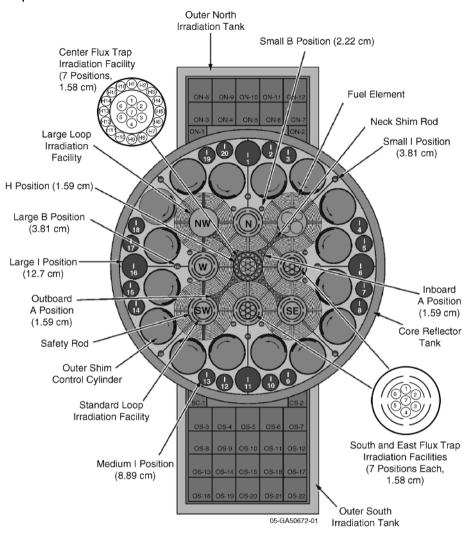


Figure 1.18 Various irradiation positions. Courtesy: Idaho National Laboratory.

Then only can we figure whether there is any hope that they can produce power at a price."

—Dr. Norman Hillberry (former director of Argonne National Laboratory, 1957–1961) (excerpt from the book by C.O. Smith)

The above statement by Dr. Hillberry (a physicist by training) says it all! Erstwhile metallurgical engineering field has now largely morphed into the field of materials science and engineering (MSE). MSE as a field of study is based on a common theme of finding out the interlinkage between processing, structure, and properties in various types of materials. If the interlinkage is clearly understood and established, the performance of these materials under service conditions could be ensured. Hence, the materials selection process in any structure design is a very important step. It is very common to encounter various *tradeoffs* during the materials selection process for a given application, and most times compromise is called for. Moreover, a nuclear reactor design entails complex procedures in itself given the multiple challenges. Different components of a reactor may require different types of materials. This is mostly done with the help of engineering expertise (experience and judgment).

There could be two broader types of materials selection considerations - general and special. General considerations involve factors such as mechanical strength, ductility, toughness, dimensional stability, fabricability, cost and availability, heat transfer properties, and so on. General properties come from the general engineering considerations as they would be applicable in most engineering designs. On the other hand, special properties are considered solely because the materials are to be used in a nuclear reactor. These include properties like the neutron absorbing characteristics, susceptibility to induced radioactivity, radiation damage resistance, and ease of reprocessing of materials. Each of the material characteristics is evaluated following standard (sometimes nonconventional) testing procedures. The knowledge of service conditions and broader goals of the future reactor is a must for a successful materials selection process. This information may come from utilizing predictive capabilities (modeling and simulation tools) and/or known data/experiences from previous reactor design and operations, if available. Brief discussions on these properties have been made in the following sections. Some of these properties will be again elaborated in the subsequent chapters.

### 1.9.1

#### General Considerations

#### 1.9.1.1 General Mechanical Properties

Important general mechanical properties include tensile strength, ductility, and toughness. The material should be strong enough to bear the loads of the structure and also sustain any internal or external stresses generated during service. Also, the material should have enough ductility (a measure of percentage elongation or reduction in area in standard tensile specimens) to avoid any catastrophic failure. Usually, as a rule of thumb, a percentage elongation of 5% is considered a minimum requirement for a load-bearing engineering structure. But one must admit that this often changes with the type of application at hand. In some cases, the materials should have sufficient ductility in order to be formed into different components. Toughness is defined as the ability of a material to absorb energy without failure, and that dictates how tough a

material is for use. Generally, tensile strength and ductility combined is referred to as toughness. However, generally impact tests and fracture toughness tests are conducted to evaluate toughness properties of materials. All these affect the mechanical integrity of the reactor components.

# 1.9.1.2 Fabricability

Fabricability includes a host of characteristics such as formability, weldability, machinability, and so on. If fabricability issues are not dealt with during the first stage, it may cause problems at the later stages. In many cases, some parts of the nuclear power plant are to be built at the site (also called *field fabrication*) from smaller parts. If the materials do not have the requisite fabricability, it would not be possible to use the material no matter what fantastic properties it may have!

# 1.9.1.3 Dimensional Stability

The material should have adequate stability in properties. For example, many nuclear components would work at higher temperatures for extended period of time. So, the creep deformation (i.e., time-dependent plastic deformation) may cause dimensional stability problems.

One should also recognize that the microstructure of a material changes as a function of temperature, time, and stresses. So, the effects of these factors on microstructure and the consequent effects on the properties need to be taken into account carefully.

# 1.9.1.4 Corrosion Resistance

Corrosion is an electrochemical process that causes the surface of the metals/alloys degrade over time in the presence of a chemical environment. Corrosion resistance of materials used in nuclear components is important in many applications to ensure that they serve as desired. The "cost of corrosion" can result in immediate property and life endangerment and increased downtime, leading to substantial losses. Many nuclear components inside the reactor stay in close contact with reactor fluids (e.g., coolant in the form of liquid or gas). These effects get exacerbated due to the presence of radiation fields.

# 1.9.1.5 Design

Although design does not generally fall under the purview of a materials engineer, he/she is in a unique position to figure out early whether the faulty design would pose a problem. Designs leaving stress concentration sites (sharp recesses, keyholes, and the like) are typically unwarranted in load-bearing applications since it may interfere with the capability of the component to serve properly. For example, fatigue properties are especially prone to the presence of stress concentration sites.

## 1.9.1.6 Heat Transfer Properties

As we know from our fundamental physics classes, heat transfer modes are of three types – conduction, convection, and radiation. The first two processes are of

importance in nuclear reactor materials selection. Most important example in nuclear reactor is the choice of fuel and cladding materials. The safety and efficiency of the reactor depends on how efficiently the heat generated inside the fuel can be removed. Hence, thermal conductivity is an important property. Otherwise, the fuel will melt such as in a "loss of coolant accident" (LOCA) scenario. Similarly, heat transfer properties are also important for various balance-of-plant features, such as heat exchangers, condensers, and other ancillary equipment (such as steam generator in a PWR system).

## 1.9.1.7 Availability and Cost

This is the last but not the least general consideration in the materials selection process. Availability and cost are the economic consideration that may trump technical considerations. If a material is not available in the form or at an allowable price, the prudent engineering decision would be to find an alternative material with similar properties or a different form of material or make changes in design to allow different characteristics. Cost-benefit analysis must be at the heart of that process. In this regard, the fair question becomes, "If it were your money, would you refuse to buy the item because it costs too much?"

### 1.9.2

#### **Special Considerations**

### 1.9.2.1 Neutronic Properties

Neutronic properties are of significant consideration in the design and development of nuclear reactors. As discussed earlier, the fission chain reaction requires continued supply of neutrons for it to proceed and that is why neutron economy plays an important role. Fuel cladding materials need to have a lower neutron absorption cross section and that is why zirconium alloys are used in LWRs (see Appendix 1.A). On the other hand, to control the chain reaction, the control materials should have high neutron absorption cross section. The same consideration would also apply to shielding materials.

## 1.9.2.2 Susceptibility to Induced Radioactivity

The materials in the reactor can absorb fast/thermal neutrons and undergo reactions that may lead to the production of different radioactive isotopes of the constituent elements of the materials. These reactions can induce radioactivity as these isotopes would decay by emitting gamma rays, beta rays, and alpha rays of different energy levels. While selecting an alloy, we should be concerned about the following factors: (a) quantity of the impurities/alloying elements, (b) abundance of the isotopes and corresponding cross section, (c) half-life of the product nuclide, and (d) the nature of the radiation produced. If the produced isotope has a short half-life and emit radiation of low energy, it should not be a cause for great concern. However, if the isotope is long-lived and produces radiation of high energy, all precautions must be taken. For

example, the main isotope of iron ( $Fe^{56}$ ) that accounts for almost 92% of the natural iron forms a stable isotope ( $Fe^{57}$ ) upon absorbing neutrons. The absorption of neutrons in  $Fe^{54}$  and  $Fe^{58}$  yielding  $Fe^{55}$  (half-life: 2.9 years) and  $Fe^{59}$  (half-life: 47 days) results in activation. However, the impurities or alloying elements cause more induced radioactivity than iron itself. Generally, the test samples irradiated in a reactor are not examined immediately after taking out from the reactor because they remain literally hot and continue to be hot due to the decay heat produced by various reactions even if the fission chain reaction no longer occurs. The Fukushima Daiichi accident in Japan did show the severity of the heat produced due to these decay reactions even after the emergency shutdown of the reactor, leading to very high temperatures (in the absence of proper coolant) and eventually resulting in the cladding breach and perhaps some form of core melting.

## Note

The development of reduced activation steels comes from the consideration of the induced radioactivity. In the mid-1980s, the international fusion reactor program initiated the development of these steels first in Europe and Japan and later in the United States. The rationale behind developing these materials stems from the easier hands-on maintenance and improved safety of operation requirements that the materials used to build the fusion reactor would not activate when irradiated by neutrons or even if it gets activated may develop only low level of activation and would decay fast. Thus, the program to produce reduced activation steels that would require only shallow burial as opposed to putting them in deep geologic repository was pursued. Researchers found out that replacing or minimizing the amount of molybdenum, niobium, nickel, copper, and nitrogen in the alloy steels would help in developing reduced activation steels. Tungsten, vanadium, and/or tantalum (low activating) have been added to these steels. Table 1.6 shows the nominal compositions of three reduced activation steels. Although the approach has originated in the fusion reactor program, it can be equally applicable to fission reactor systems.

Steel	Region	с	Si	Mn	Cr	W	v	Та	N	В
JLF-1	Japan	0.1	0.08	0.45	9.0	2.0	0.2	0.07	0.05	_
Eurofer	Europe	0.11	0.05	0.5	8.5	1.0	0.25	0.08	0.03	0.005
9Cr–2WVTa	USA	0.10	0.30	0.40	9.0	2.0	0.25	0.07	—	—

Table 1.6	Nominal compositions	of reduced activation steels	(in wt%, balance Fe).
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#### 1.9.2.3 Radiation Stability

In the subsequent chapters, we will see more detailed accounts of how energetic radiation plays a significant role in modifying the microstructure of the materials involved. Radiation damage under the fast neutron flux involves atomic displacements (i.e., displacement damage) leading to the creation of a host of defects in the material. The effects of radiation can be diverse, including radiation hardening, radiation embrittlement, void swelling, irradiation creep, and so forth, with all having significant effects on the performance of the reactor components. Another interesting effect of radiation is the radiolytic decomposition of coolant (e.g., water molecule is radiolyzed into more active radicals) that may definitely affect the corrosion behavior of the reactor components. Fission fragments also cause damage, but they are mostly limited to the fuel. So, for selecting materials for a nuclear reactor, we must know the concomitant radiation effects on these materials. That is why millions of dollars are spent to wage materials irradiation campaigns in test reactors followed by careful postirradiation examination to ascertain fitness-for-service quality of the materials to be used in nuclear reactors.

#### 1.9.3

## Application of Materials Selection Criteria to Reactor Components

Here, we summarize the criteria for materials selection for different nuclear components. Let us take the example of fuel cladding material for the LWR. As noted before, cladding materials are used to encapsulate the fuel and separate it from the coolant. The requirements for fuel cladding material are as follows: (a) low cross section for absorption of thermal neutrons, (b) higher melting point, adequate strength and ductility, (c) adequate thermal conductivity, (d) compatibility with fuel, and (e) corrosion resistance to water. Following the first factor, we have discussed in Section 1.7 how different metals have different cross sections for absorption of thermal neutrons. Although Be, Mg, and Al all have lower cross sections for absorption of thermal neutrons, other nonnuclear factors become the impediment for their use in commercial power reactors. Even though Be has a high melting point (1278 °C), it is scarce, expensive, difficult to fabricate, and toxic. Mg has a low melting point (650 °C), is not strong at higher temperatures, and has poor resistance to hot water corrosion. Al has a low melting point (660 °C) and poor high-temperature strength. Even though an Albased alloy has been used as fuel cladding materials in reactors like ATR, and in the past a magnesium-based alloy was used in Magnox reactors, their use remains very limited. This leaves zirconium-based materials as the mainstay of fuel cladding materials for LWRs. Zirconium has various favorable features: (a) relatively abundant, (b) not prohibitively expensive, (c) good corrosion resistance, (d) reasonable high-temperature strength, and (e) good fabricability. Some of the properties could be further improved through appropriate alloying. More detailed discussion on the development of zirconium alloys is included in Appendix 1.A at the end of the chapter.

# 1.9.3.1 Structural/Fuel Cladding Materials

Major requirements	Possible materials
Low neutron absorption Stability under heat and radiation Mechanical strength Corrosion resistance Good heat transfer properties	Al, Be, Mg, and Zr Stainless steels Superalloys (Ni-based) Refractory metals (Mo, Nb, Ti, W, etc.)

# 1.9.3.2 Moderators and Reflectors

Major requirements	Possible materials
Low neutron absorption	Water (H <sub>2</sub> O, D <sub>2</sub> O)
Large energy loss by neutron per collision	Beryllium (BeO)
High neutron scattering	Graphite (C)

# 1.9.3.3 Control Materials

Major requirements	Possible materials
High neutron absorption	Boron
Adequate strength	Cadmium
Low mass (for rapid movement)	Hafnium
Corrosion resistance	Hafnium
Stability under heat and radiation	Rare earths (Gadolinium, Gd; Europium, Eu)

# 1.9.3.4 Coolants

Major requirements	Possible materials
Low neutron absorption	Gases (air, hydrogen, helium, carbon dioxide, and water)
Good heat transfer properties	Water ( $H_2O$ and $D_2O$ )
Low pumping power (i.e., low melting point)	Liquid metals (Na, Na—K, Bi)
Stability under heat and radiation	Molten salts (—Cl, —OH, —F)
Low induced radioactivity	Organic liquids
Noncorrosiveness	-

(Example:

The world's first nuclear power plant was EBR-1.

It carried a coolant, an alloy of sodium (Na) and potassium (K), called Na-K ("nack").

The following are the coolant characteristics:

- Stays liquid over a wide range of temperatures without boiling away.
- Transfers heat very efficiently taking heat away from the reactor core and keeping it cool.
- Allows neutrons from the reactor core to collide with U-238 in the breeding blanket and produce more fuels.)

## 1.9.3.5 Shielding Materials

Major requirements	Possible materials
Capacity to slow down neutrons Absorption of gamma radiation	Light water (H <sub>2</sub> O) Concrete, most control materials, and metals (Fe, Pb, Bi, Ta, W, and Broal – a B and Al alloy)
Absorb neutrons	

# 1.10 Summary

In this introductory chapter, we first introduced nuclear energy and discussed its significance in the modern civilization. We also discussed some nuclear physics fundamentals such as half-value thickness for neutron beam attenuation, nuclear cross sections, neutron flux and fluence, and other concepts. A detailed overview of different reactors is presented. The material selection criteria for nuclear components are also discussed.

# Appendix 1.A

Zirconium-based alloys are commonly used in water reactors for cladding UO<sub>2</sub>, while Zircaloy-2 and Zircaloy-4 are used in BWRs and PWRs, respectively. The following are the reasoning and historical development of these cladding materials:

The fuel  $(UO_2)$  is inserted in canning tubes that separate the radioactive fuel from the coolant water. The requirements for cladding materials thus are as follows:

- Low cross section for absorption of *thermal* neutrons
- Adequate strength and ductility
- Compatibility with fuel

- 38 1 Overview of Nuclear Reactor Systems and Fundamentals
  - Adequate thermal conductivity
  - Corrosion resistance to water

In order of increasing cross section for absorption  $\left\{\sum_{a}^{th}\right\}$  of thermal neutrons, the various metals can be classified [normalized to Be] as follows:

Relative to Be	1	7	20	24	122	278	281	322	410	512
							Fe			
Melting point $\{^{\circ}C\}$	1283	650	1845	660	2415	2625	1539	1890	1083	1455

Be: scarce, expensive, difficult to fabricate, and toxic

Mg: not strong at high temperatures and poor resistance to hot water corrosion Al: low melting point and poor high temperature strength

Zirconium is relatively abundant, is not prohibitively expensive, has good corrosion resistance, has *reasonable* high-temperature strength, good fabricability, and can be further improved by *proper alloying*. Processing of Zr metal from ore requires removal of hafnium [Hf], which is always associated with Zr. Hf has relatively high absorption of thermal neutrons. This Kroll process was relatively more expensive  $\Rightarrow$  Mg treated. The elements used in alloying for increasing strength are O, Sn, Fe, and [Cr, Ni] and for improving corrosion are Cr, Ni, and Fe.

Thus, the Zircaloys were developed (mainly from the US Navy in 1950s). For a nice description of the history of Zry development, refer to the following:

- Krishnan, R. and Asundi, M.K. (1981) Zirconium alloys in nuclear technology, in *Alloy Design* (eds. S. Ranganathan *et al.*), Indian Academy of Sciences.
- Rickover, H.G. (1975) History of Development of Zirconium Alloys for Use in Nuclear Power Reactors, US ERDA, NR&D.

Element [w/o]	Sn	Fe	Cr	Ni	0	Zr
Zircaloy-2	1.5	0.12	0.10	0.05	0.01	Balance – BWRs
Zircaloy-4	1.5	0.21	0.10	_*	0.01	Balance – PWRs

\*Ni enhances hydrogen pickup and thus was removed for PWR applications. To compensate for the corrosion and strength improvements realized by adding Ni, it was replaced by Fe.

Element [w/o]	Sn	Fe	Zr	
Zircaloy-1	2.5	-	Balance	Break-away transition <i>not improved</i>
Zircaloy-3	0.25	0.25	Balance	Poor mechanical strength

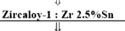
Recent Developments

*Barrier Cladding* \_ Zircaloy-2 with Zr liner (ID) for PCI resistance Zirlo@ alloys \_ Zircaloy-4 + 0.5 to 1% Nb \_ good long-term corrosion *Duplex alloy* \_ low Sn on the surface (OD) but still in ASTM spec

• Zr + Fe liner in lieu of Zr • Zry-2/Zr/Zry-2 (Tri-clad) • M5 (Zr-1Nb



Sn, Ta, and Nb (in decreasing order of effectiveness) circumvent damaging effect of impurities such as nitrogen and improve corrosion resistance Sn selected since neutron economy is little affected and 2.5%Sn improved corrosion, strength, and fabricability



Breakway transition in corrosion (wt gain versus time) was not improved Zircaloy-2 was developed as an accidental finding at Bettis labscorrosion improvement noted by contamination from SS  $\Rightarrow$  new alloy developed with Fe and Cr similar to impurity levels in Zr + 0.05% Ni Sn reduced to 1.5%-enough to counteract the nitrogen effect

Zircalov-2 : Zr 1.5%Sn 0.15%Fe 0.05%Ni 0.1Cr (BWRs) • has as good a strength as Zry-1 but improved corrosion •

Sn is known to be bad for long-time corrosion under PWR conditions Ø reduced Sn to 0.25% and simultaneously increased Fe to 0.25% lead to the most corrosion resistant alloy in Zircaloy family

Zircaloy-3 : Zr 0.25%Sn 0.25%Fe

• the mechanical strength was not adequate and thus Zry-3 was abandoned •

Hydrogen effects on mechanical properties were just emerging and zirconium hydrides resulted in reduced impact energy Ø again, accidentally the effect of Ni on hydrogen absorption was noted (during work on eutectic diffusion bonded plates)  $\emptyset$ Ni-free Zircaloy-2 was developed but with poor corrosion resistance increased Fe to 0.24% with no Ni  $\rightarrow$  almost as good steam corrosion resistance as Zry-2 but with reduced (by 50%) hydrogen absorption Zircaloy-4 : Zr 1.5%Sn 0.24%Fe 0.1%Cr (PWRs)

Recent Trends Barrier cladding  $\rightarrow$  Zry-2 with Zr liner for PCI resistance  $\oslash$  Tri-Clad *Zirlo* alloys  $\rightarrow$  Zry + 0.5 to 1% Nb  $\rightarrow$  good for long-term corrosion Duplex alloy  $\rightarrow$  low Sn on the surface (but still in ASTM spec) versus bulk

## Problems

- 1.1 a) What is the percentage of  $U^{235}$  in naturally occurring uranium and what is the rest made of?
  - b) A nuclear fission reaction of an  $U^{235}$  atom caused by a neutron produces one barium atom, one Krypton atom, and three more neutrons. Evaluate approximately how much energy is liberated by this reaction.

Approximately how much percentage of energy is carried by the fission fragments (no calculation necessary for the last part of the question)?

- 1.2 What is the difference between fissile and fertile isotopes? Give two examples of each. What is the role of fertile isotopes in a breeder reactor?
- **1.3** Define a nuclear reactor? What is the basic difference between an atomic bomb and a power-producing reactor?
- 1.4 What are the prime differences between LWR and CANDU reactors (comment mostly on materials aspects)?
- 1.5 Describe the importance of control materials with respect to reactor safety and control. What are the primary requirements for a control material? Give at least four examples of control materials.
- 1.6 Categorize neutrons based on their kinetic energy. What is the major difference between a thermal reactor and a fast reactor?
- 1.7 a) Zirconium and hafnium both have crystal structures (HCP) in the general operating regimes of LWRs. Naturally occurring Zr always has some Hf (1–3 wt%) in it. Hf-containing Zr alloys are very common in chemical industries but not in nuclear industries. Why?
  - b) What is the main application of Zr alloys in LWRs? What are the various functions of this reactor component? What are the reasons that make Zr alloys suitable for such use?
- 1.8 What are the two main zirconium alloys used in light water reactors? Give their compositions. Name two recently developed zirconium alloys with their compositions.
- 1.9 What is neutron economy? What significance does it have? How much influence does it exert in the selection of materials used in nuclear reactors?
- 1.10 a) Define neutron flux and neutron fluence. What are their units?
  - b) Define neutron cross section? Briefly comment on the importance of neutron cross section from a reactor perspective.
  - c) Neutrons of 10 keV energy are incident on a light water barrier. The neutron cross section for hydrogen (protium) at 10 keV is about 20 b and that of oxygen is only 3.7 b. Determine the half-value thickness of neutron attenuation for the water barrier (assume that neutron interaction with oxygen in water molecule is negligible). Find out the half-thickness value for 1 MeV neutrons traveling through the water barrier (neutron cross section for protium is 4.1 b for 1 MeV neutrons). Comment on the significance of the results.

## **Additional Reading Materials**

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- 2 Murray, R.L. (1955) Introduction to Nuclear Engineering, Prentice Hall, New York.
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- 5 Klueh, R.L. (2005) Elevated temperature ferritic and martensitic steels and their application to future nuclear reactors. *International Materials Review*, 50 (5), 287–310.
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- 7 Hinds, D. and Maslak, C. (January 2006) Next Generation Nuclear Energy: The ESBWR, Nuclear News, pp. 35–40.
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