Topics in Mining, Metallurgy and Materials Engineering

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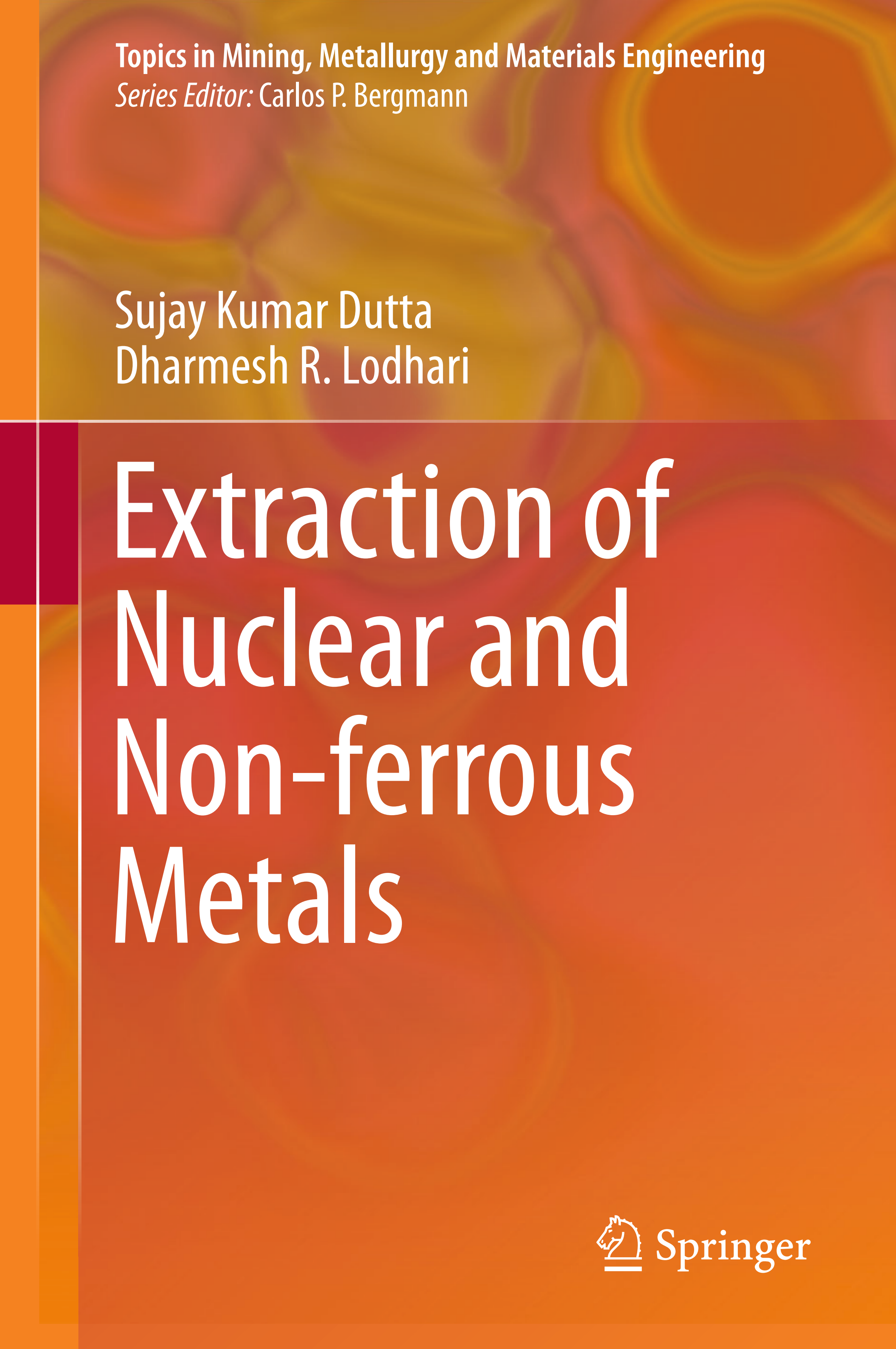
Dharmesh R. Lodhari

Extraction of Nuclear and Non-ferrous Metals



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Extraction of Nuclear and Non-ferrous Metals

Springer

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Preface

Extraction of Nuclear and Non-Ferrous Metals is a basic science and has a wide applicability in extraction and refining of metals. This book covers the undergraduate curriculum in **Nuclear Metallurgy and Extraction of Non-Ferrous Metals** as prescribed for degree courses and allied professional courses in Metallurgical and Materials Engineering.

Chapter 1 of this textbook, *Fundamentals of Nuclear Metallurgy*, covers almost all the important basic concepts of **Nuclear Metallurgy** for graduate engineering students. Even postgraduate students, engineers and researchers can brush up their understanding. Chapters 2–6 briefly cover **Nuclear Metals**, their extraction, properties and applications.

Chapters 7–13 discuss **Extractive Metallurgy of Common Metals**. There are detailed discussions about production of common metals (like copper, aluminium, zinc, lead etc.), their properties and applications.

Chapters 14–20 describe the **Extractive Metallurgy of Less Common/Ferro-Alloying Metals.** Their production, properties and applications are discussed in detail. Chapter 21 discusses **Ultra-High-Purity Metals Production.**

Needless to say that although few books are available, none discusses the subject matter of **Nuclear Extractive Metallurgy** in easily understood terms for students. Based on our experience of teaching this subject at undergraduate level for more than three decades, we have made a sincere attempt to cover the topics of **Nuclear Metallurgy** in an easy way.

In spite of taking all possible care, there may be some errors or mistakes left unnoticed. If so, please feel free to interact with us. We poured our long experience into this project, and also collected the information from several sources. We are indebted to one and all, from whose valuable knowledge we have been benefited. Thanks all of them.

We are confident that this textbook will make the subject matter of *Extraction of Nuclear and Non-Ferrous Metals* more simple and easy to understand. We wish to thank the publisher Springer for their continuous support in preparation and publication of this textbook within a short period.

Vadodara, India

Sujay Kumar Dutta
Dharmesh R. Lodhari

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About the Authors

Dr. Sujay Kumar Dutta is a former Professor and Head of the Department of Metallurgical and Materials Engineering, Maharaja Sayajirao University of Baroda, India. He received his Bachelor of Engineering (Metallurgy) from Calcutta University in 1975 and Master of Engineering (Industrial Metallurgy) from Maharaja Sayajirao University of Baroda in 1980. He was awarded Ph.D degree by the Indian Institute of Technology Kanpur, India in 1992. He joined the M. S. University of Baroda as Lecturer in 1981, subsequently being appointed a Reader in 1993 and a Professor in 2001.

Professor Dutta has received several awards, including the Essar Gold Medal (2006) from the Indian Institute of Metals, a Fellowship (2014) from the Indian Institute of Metals, and the IIM Distinguished Educator Award (2015) from the Indian Institute of Metals, Kolkata in recognition of his distinguished service to Metallurgical Education and to the Indian Institute of Metals.

Professor Dutta has published three books along with other authors: *Metallurgical Thermodynamics, Kinetics and Numericals* (2011); *Alternate Methods of Ironmaking (Direct Reduction and Smelting Reduction Processes)* (2012); *Iron Ore–Coal/Coke Composite Pellets* (2013), and *Extractive Metallurgy (Processes and Applications)* is currently in publication. He has also published 120 papers in national and international journals and conference proceedings.

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Part I
Extractive Metallurgy of Nuclear Metals

Chapter 1
Fundamentals of Nuclear Metallurgy

1.1 Atomic Structure

The operation of a nuclear reactor depends upon various interactions of neutrons with atomic nuclei. In order to understand the nature and characteristics of these reactions, it is desirable to consider briefly some of the fundamentals of atomic and nuclear physics. An atom consists of a positively charged *nucleus* surrounded by a number of negatively charged *electrons*; so the atom as a whole is electrically neutral. Atomic nuclei are combination of two primary particles, namely, *protons* and *neutrons*. Protons and neutrons are jointly named nucleus. The protons carry a single unit positive charge, equal in magnitude to the electronic charge. The neutron is very slightly heavier than the proton and it is an electrically neutral particle carrying no charge. The neutrons are the heart of nuclear energy and they play an important role in the release of atomic energy. All atomic nuclei, with the exception of hydrogen, contain one or more neutrons in addition to protons.

Atomic energy is derived by virtue of the transformation of matter into energy. Calculations show that the conversion of small amount as 1 g of matter yields energy equivalent of 25 million kWh. By contrast, the combustion of the same amount of coal gives the insignificant amount of 0.0085 kWh. The difference in these figures shows the distinction between atomic and chemical energy.

For a given element, the number of protons present in the atomic nucleus is called the *atomic number* and is denoted by $\boldsymbol{Z}$. It is similar to the number of the element in the *Periodic Table* (Fig. 1.1).

e.g. H → 1, He → 2, Li → 3, C → 6, O → 8, Fe → 26, U → 92 etc.

The total number of protons and neutrons in an atomic nucleus is called the *mass number* of the element and is denoted by $\boldsymbol{A}$. Hence, number of neutrons in the atomic nucleus is $(A - Z)$. In general the element (M) can be represented by $_ZM^A$.

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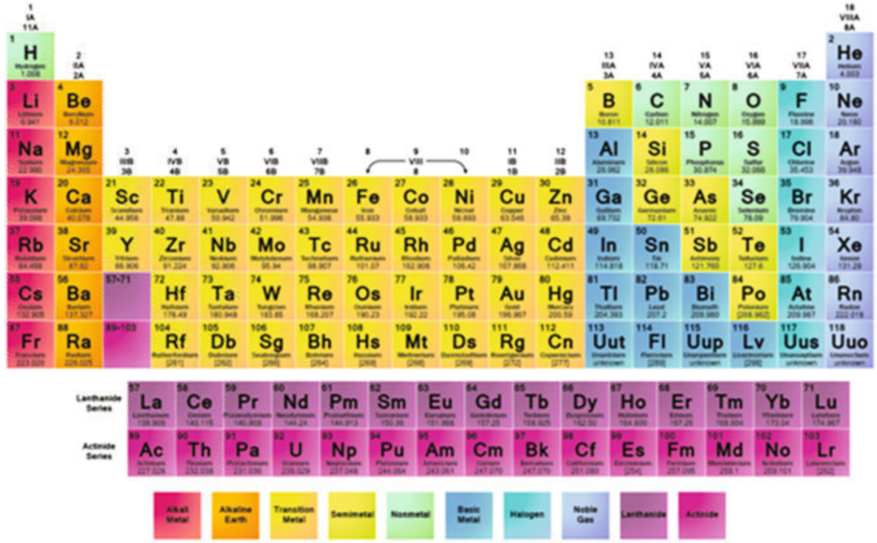

Fig. 1.1 Periodic Table of the elements

1.2 Isotopes

It is the atomic number, i.e. the number of protons in the nucleus, which determines the chemical nature of an element. This is so because the chemical properties depend on the orbital electrons which are surrounding the nucleus; and their number must be equal to the number of protons, since the atom as a whole is electrically neutral.

Atoms with nuclei containing the same numbers of protons (i.e. with the same atomic number, but with different mass numbers) are essentially identical chemically, although they frequently exhibit marked differences in their nuclear characteristics. Such species, having the same atomic number but different mass numbers, are called *isotopes*. Uranium-232 ($_{92}U^{232}$) has three isotopic forms in nature, with mass numbers 234, 235, and 238 ($_{92}U^{234}$, $_{92}U^{235}$ and $_{92}U^{238}$).

1.3 Nuclear Binding Energy

By means of the mass spectrograph, it has been shown that the actual mass is always less than the sum of the masses of the constituent nucleus. The difference is known as *mass defect*, which is related to the energy binding of particles in the nucleus. If m_p, m_n and m_e represent the masses of the proton, neutron and electron respectively, the sum of the masses of the constituents of an atom is $Zm_p + Zm_e + (A - Z)m_n$. Suppose the observed mass of the atom is M, then

$$\text{mass defect} = \left[Z\left(m_p + m_e\right) + (A - Z)m_n\right] - M = Zm_H + (A - Z)m_n - M \quad (1.1)$$

where $m_p + m_e = m_H$, the mass of the hydrogen atom.

Since $m_H = 1.008145$ amu* and $m_n = 1.008986$ amu

(*amu** → *atomic mass unit* is defined as exactly one-sixteenth of the mass of oxygen (O^{16}) atom or one-twelfth of the mass of carbon (C^{12}) atom.)

Therefore,

$$\text{mass defect} = [1.008145\,Z + (A - Z)\,1.008986 - M \quad (1.2)$$

The mass defect is a measure of the energy which would be released, if the individual Z protons and (A − Z) neutrons combined to form a nucleus. Conversely, it is numerically equal to the energy which would have to be supplied to break apart the nucleus into its constituents. Thus, the energy equivalent of the mass defect is called the *binding energy* of the nucleus; i.e. the energy, which holds together the nucleus of an atomic nucleus, is named the *binding energy*.

If m is the decrease in mass accompanying any particular process, then the equivalent amount of energy E released is given by the Einstein equation:

$$E = mc^2 \quad (1.3)$$

where m is mass in grams, c is velocity of light = 2.998×10^{10} cm/sec

Therefore,

$$E = m \times 8.99 \times 10^{20} \text{ ergs} \quad (1.4)$$

In the nuclear energy, energies are usually stated in terms of the *electron volt unit*, represented by ev. Since the electronic charge is 1.602×10^{-19} C.

$$\text{The electrical energy, } e = vQ\ J, \quad (1.5)$$

where v is in volts, and Q is the amount of electricity in coulomb.

If 1 ev = 1.602×10^{-19} J = 1.602×10^{-12} erg (since 1 J = 10^7 erg)

1 Mev (million electron volts) = 1.602×10^{-6} erg

Therefore,

$$\begin{aligned} E\,(\text{Mev}) &= m\,(\text{gm}) \times \left[\left(8.99 \times 10^{20}\right)/\left(1.602 \times 10^{-6}\right)\right] = 5.612 \times 10^{26}\ m \\ &= 5.612 \times 10^{26} \times 1.66 \times 10^{-24} m = 931.59\ m\,(\text{amu}) \end{aligned} \quad (1.6)$$

Since 1 amu = 1.66×10^{-24} gm

The numerical value of the nuclear binding energy in Mev can be obtained upon multiplying by 931.6, the mass defect in amu, as given by Eq. (1.2). A more useful quantity is the average binding energy per nucleon, which is equal to the total binding energy (BE) divided by the number of nucleons, i.e. by the mass number A; hence from Eqs. (1.2) and (1.6):

$$[\mathrm{BE/A}] = [(931.59/\mathrm{A}) \text{ x } \{1.008145 \text{ x } \mathrm{Z} + 1.008986 \text{ x } (\mathrm{A} - \mathrm{Z}) - \mathrm{M}\}] \tag{1.7}$$

Example 1.1 Determine the binding energy per nucleon in (a) Sn-120 for which M is 119.9401 amu and (b) U-235 for which M is 235.1175 amu.

Solution: (a) The atomic number of Sn is 50, i.e. Z = 50 and A = 120
Since [BE/A] = [(931.59/A) x {1.008145 x Z + 1.008986 x (A − Z) − M}]
Therefore,

$$\begin{aligned}[\mathrm{BE/A}] &= [(931.59/120) \text{ x } \{(1.008145 \text{ x } 50) + (1.008986 \text{ x } 70) - 119.9401\}] \\ &= \mathbf{8.51\ Mev\ per\ nucleon}\end{aligned}$$

(b) The atomic number of U is 92, i.e. Z = 92 and A = 235

$$\begin{aligned}[\mathrm{BE/A}] &= [(931.59/120) \text{ x } \{(1.008145 \text{ x } 92) + (1.008986 \text{ x } 143) - 235.1175\}] \\ &= \mathbf{7.59\ Mev\ per\ nucleon}\end{aligned}$$

1.4 Radioactivity

There are conventional electrostatic repulsive forces between the positively charged protons. When the atomic number is low, the repulsive force among the protons is small. Hence, the proton–proton, neutron–neutron, and proton–neutron forces are roughly equal; a neutron/proton ratio is close to unity, to be expected for stability. But with increasing atomic number, electrostatic repulsion between the protons, which varies as Z^2, becomes more and more important. Since, the total electrostatic repulsion force between all the protons in the nucleus is proportional to the square of their number, i.e. to Z^2.

In order to maintain stability, the nuclei must now contain an increased proportion of neutrons, so that the attractive neutron–neutron and neutron–proton forces can compensate for the rapidly increasing repulsive forces between the protons. But there is a limit. Consequently, the elements of atomic number 84 or larger have no stable isotopes; although elements 84 (polonium, Po) through 92 (uranium, U) exist in nature, they are unstable and exhibit the phenomenon of *radioactivity*.

Since properties of atomic nuclei depend on the total number of protons and neutrons (nucleons) in the nucleus, isotopes of one and the same element possess different nuclear properties. That is, some isotopes are stable, whereas others are unstable or radioactive.

Radioactive isotopes undergo spontaneous nuclear change (i.e. transformation) at a definite rate in the direction of increasing stability of the element. During this transformation the unstable nucleus emits characteristic particles (i.e. high velocity particles or radiation) and is thereby transformed into a different nucleus which may or may not also be a radioactive element. Isotopes which owe their instability to their high mass numbers emit either positively charged *alpha (α) particles* or negatively charged *beta (β) particles.*

(a) Alpha (α) particles are identical with helium nuclei ($_2He^4$) and consist of two protons and two neutrons. The product (or daughter) nucleus of alpha decay has two protons and two neutrons less than the parent nucleus, so that its mass number is four units less:

$$_ZM^A \rightarrow {}_{Z-2}M^{A-4} + {}_2He^4(\alpha) \tag{1.8}$$

In other words, it produces an element in the Periodic Table (Fig. 1.1) two places to the left of the parent isotope. For example, α disintegration of $_{92}U^{238}$ results in its transformation into $_{90}Th^{234}$:

$$_{92}U^{238} \rightarrow {}_{90}Th^{234} + {}_2He^4(\alpha) \tag{1.9}$$

(b) Beta (β) particles are identical to ordinary electrons. The nucleus itself does not contain electrons, and in radioactive beta decay the electron arises from the spontaneous conversion of a neutron into a proton and an electron:

$$\text{neutron} \rightarrow \text{proton} + \text{electron}\left(\beta\,\text{particle},\ {}_{-1}\beta^0\right) + \text{neutrino} \tag{1.10}$$

The additional neutral particle, with essentially zero mass, called a *neutrino*, carries out some of the energy liberated in the radioactive transformation. In beta decay the daughter nucleus has one neutron less and one proton more than its parent, but the mass number is unchanged; i.e. this produces an element in the Periodic Table (Fig. 1.1) one place to the right of the parent element:

$$_ZM^A \rightarrow {}_{Z+1}M^A + {}_{-1}\beta^0 \tag{1.11}$$

$$\text{e.g. } {}_{92}U^{239} \rightarrow {}_{93}Np^{239} + {}_{-1}\beta^0 \rightarrow {}_{94}Pu^{239} + {}_{-1}\beta^0 \tag{1.12}$$

A neutron is replaced in the nucleus by a proton, so that the neutron/proton ratio decreases, the daughter nucleus will be more stable than its parent, not necessarily completely stable.

(c) In many cases, although not always, radioactive decay is associated with the emission of gamma (γ) rays, in addition to an alpha (α) or beta (β) particles. Gamma (γ) rays are penetrating electromagnetic radiations of high energy, essentially identical with X rays. In fact, the only difference between γ rays and X rays is that the former originate from an atomic nucleus; whereas the later are produced by processes outside the nucleus.

γ rays occur in a radioactive change when the daughter nucleus is formed in what is called an excited state, i.e. a state in which it has a higher internal energy than the normal (or ground) state of that nucleus. The excess energy is then released almost instantaneously as γ radiation. Such an excited nucleus is known as an *isomer of the unexcited nucleus*.

1.5 Rate of Radioactive Decay

The rate of radioactive decay is a value characteristic of a given isotope and independent of the physical and chemical state of the element, at least under the conditions of temperature and pressure. In a given specimen, the rate of radioactive decay at any instant is always directly proportional to the number of radioactive atoms of the isotope under consideration present at that time. Thus, if N is the number of the particular radioactive atoms (or nuclei) present at any time t, the decay rate is given by:

$$(dN/dt) = -\lambda N \tag{1.13}$$

where λ is the radioactive decay (or disintegration) constant of the radioactive species, a measure of its decay probability. Minus sign indicates a decrease in the number of the particular radioactive atoms (or nuclei) i.e. N. This Eq. (1.13) permits a determination of N at a given time t, assuming that there were N_0 parent nuclei at the initial time t_0. By integrate Eq. (1.13):

$$N = N_0 e^{-\lambda t} \tag{1.14}$$

The rate of radioactive disintegration is conveniently expressed by means of a so-called *half-life period* ($t_{1/2}$). It is defined as the time required for the number of radioactive nuclei of a given kind to decay to half its initial value. Assuming $N = 0.5N_0$ and applying to Eq. (1.14):

$$t_{1/2} = (0.6931/\lambda) \tag{1.15}$$

The reciprocal of the decay constant, represented by t_m, is known as *mean life* (or *average life*) of the radioactive species; thus,

$$t_m = 1/\lambda \tag{1.16}$$

The half-life is thus inversely proportional to the decay constant or by Eq. (1.15), directly proportional to the mean life, i.e.

$$t_{1/2} = 0.6931 t_m \tag{1.17}$$

Some of the data for nuclides that are of direct or indirect interest as nuclear fuels are given in Table 1.1. The long half-life is important since it means that there are

Table 1.1 Radioactive characteristics of nuclear fuel isotopes

Nuclide	Radiation	Half-life, years
Th^{232}	α	1.39×10^{10}
U^{233}	α	1.62×10^{5}
U^{235}	α	7.13×10^{8}
U^{238}	α	4.51×10^{9}
Pu^{239}	α	2.44×10^{4}

no appreciable losses in storage over many hundreds of years. It is of interest that, when the fuel species with the shortest half-life, namely Pu^{239}, decays its daughter is U^{235} having a much longer half-life.

$$_{94}Pu^{239} \rightarrow {}_{2}He^{4} + {}_{92}U^{235} \tag{1.18}$$

Since the uranium (U^{235}) is also a nuclear fuel, there is no appreciable loss in usefulness of the material as a source of energy.

Example 1.2 If $(dN/dt) = -\lambda N$, then prove that: $N = N_0 e^{-\lambda t}$

Solution:

$$(dN/dt) = -\lambda N$$

$$\text{By integrating} \int_{N_0}^{N} (dN/dt) = -\int_{0}^{t} \lambda N$$

$$\text{Or} \quad \int_{N_0}^{N} (dN/N) = -\lambda \int_{0}^{t} dt \quad (\text{since } \lambda \text{ is constant})$$

$$\text{Or} \quad (\ln N - \ln N_0) = -\lambda t \quad \text{or} \quad \ln(N/N_0) = -\lambda t$$

Hence, $\mathbf{N = N_0 e^{-\lambda t}}$ **Prove**

1.6 Neutron Reaction

In addition to the spontaneous nuclear reactions due to radioactivity, in which one nucleus is changed into another as the result of a particle, there are many nuclear reactions which can be brought about artificially, resulting from the absorption of one particle or another. Among such reactions, those in which a neutron is absorbed are of special interest. Neutrons are normally bound in atomic nuclei, but it is possible to obtain these in the free state. Such free neutrons can interact in various ways with nuclei.

There are three types of neutron-nuclear interactions of importance in the operation of nuclear reactors, namely: (1) scattering; (2) capture; and (3) fission.

(1) *Scattering*: The nucleus absorbs the neutron to form a compound nucleus in an excited state of higher internal energy. This compound nucleus rapidly expels a neutron with a lower kinetic energy than the absorbed neutron, the excess energy remaining on the residual nucleus.

   (a) A compound nucleus may emit a particle identical with the one absorbed by the nucleus, the total kinetic energy of the struck nucleus and the bombarding particle remaining unchanged. Such a process is referred to as *elastic scattering*. This kind of reaction neither releases nor absorbs energy, and its only result is a redistribution of the kinetic energy of the collision particles.

$$\text{Reaction}: \quad {}_6\mathrm{C}^{12} + {}_0\mathrm{n}^1 \rightarrow {}_6\mathrm{C}^{12} + {}_0\mathrm{n}^1 \text{ or } \mathrm{C}^{12}(\mathrm{n},\mathrm{n})\mathrm{C}^{12} \tag{1.19}$$

   (b) It may happen that the incident particle and the ejected particle are identical, but the total kinetic energy of the colliding particles decreases after the collision. The energy difference is emitted by the compound nucleus as a γ radiation. Such a process is called *inelastic scattering*.

$$\text{Reaction}: \quad {}_{92}\mathrm{U}^{238} + {}_0\mathrm{n}^1 = {}_{92}\mathrm{U}^{238} + {}_0\mathrm{n}^1(\text{slow neutron}) + \gamma \tag{1.20}$$

   Let E_1 is the total kinetic energy of the neutron and target nucleus before collision, and E_2 is the kinetic energy after collision; if E_γ is the energy emitted as γ radiation, then $E_1 = E_2 + E_\gamma$ (as shown in Fig. 1.2).

(2) *Capture*: The bombarding particle may be trapped in the nucleus. In this case the excitation energy of the compound nucleus is carried away by the γ radiation emitted after absorption of the particle. Such a process is called *capture*. The capture of a neutron by a nucleus followed by the emission of γ radiation.

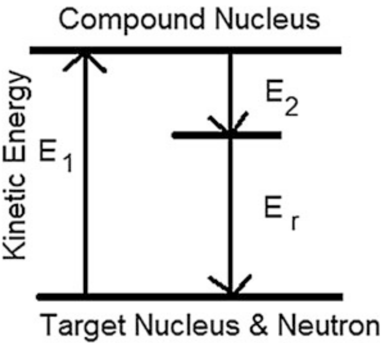

Fig. 1.2 Kinetic energy of the neutron and target nucleus before and after collision

In this reaction the neutron is taken up by the reacting (or target) nucleus in a high energy state. Within a small fraction of a second, the compound nucleus emits the excess energy as radiation, leaving a nucleus differing by one neutron from the target nucleus:

$$_{92}U^{238} + {_0n^1} = {_{92}U^{239}} + \gamma \tag{1.21}$$

The resulting nucleus, U-239, is radioactive and decays with the emission of a negative beta particle ($_{-1}\beta^0$). Thus:

$$_{92}U^{239} \rightarrow {_{-1}\beta^0} + {_{93}Np^{239}} \tag{1.22}$$

The product, Np-239 being an isotope of an element of atomic number 93, called neptunium (Np), which does not normally exist on earth. Np-239 is also beta active and decays fairly rapidly.

$$_{93}Np^{239} \rightarrow {_{-1}\beta^0} + {_{94}Pu^{239}} \tag{1.23}$$

(3) *Fission:* When a compound nucleus attains such a state of excitation that it splits into two parts of lighter masses. This process is called *fission*. When certain nuclei of higher atomic number and mass number capture neutrons, the resulting exited compound nucleus, instead of emitting its excess energy as γ radiation, splits into two lighter nuclei having masses very different from that of the original heavy nucleus.

$$_{92}U^{235} + {_0n^1} = {_{38}Sr^{89}} + {_{54}Xe^{145}} + \left({_0n^1} + {_0n^1}\right)\{\text{Fast neutrons}\} + \text{Heat energy} \tag{1.24}$$

When first liberated, free neutrons usually possess high kinetic energies, in the million electron volt range, and so they are called *fast neutrons*. The fission reaction is the basic to the operation of nuclear reactors. Fission occurs only with nuclei of high atomic (and mass) number, and the large value of Z^2, and hence the repulsive force within the nucleus, is an important contributory factor. When fission occurs, the excited compound nucleus formed after absorption of a neutron breaks up into two lighter nuclei, called *fission fragments*.

The important of fission, from the standpoint of the utilization of nuclear energy, lies in two facts: (a) the process is associated with the release of a large amount of energy per unit mass of nuclear fuel; and (b) the fission reaction, which is initiated by neutron, is accompanied by the liberation of neutrons.